UNDERSTANDING HUMAN IMPACTS ON THE PHOSPHORUS CYCLE: IMPLICATIONS FOR AGRONOMIC AND ENVIRONMENTAL MANAGEMENT AT MULTIPLE SCALES

Graham Kyle MacDonald

Department of Natural Resource Sciences
McGill University, Montreal

July 2012

A thesis submitted to McGill University in partial fulfilment
of the requirements of the degree of Doctor of Philosophy

ABSTRACT

Modern agriculture has led to fundamental changes in the phosphorus (P) cycle that pose agronomic and environmental challenges at all scales. Phosphorus is a non-renewable resource that is critical to food production because of its role as an essential plant nutrient. At the same time, P runoff from agricultural systems contributes to water quality degradation worldwide. Recent research has begun to illuminate the dimensions of P use in agriculture and its broader sustainability implications, yet there is limited understanding of how these disparate issues are connected across scales. In this thesis, I explore important knowledge gaps related to spatial and temporal changes in P flows due to human activity, their drivers, and some of the implications for large-scale management of soil P and water quality.

In the first study, I conducted a global analysis to better understand the contemporary distribution of agronomic P use for croplands. The spatially-detailed results revealed that disparities in the magnitude of P applied to cropland soils as fertilizer and manure relative to crop P use occur across most regions, but with considerable spatial variation. Although inputs of P fertilizer (14.2 Tg of P y^{-1}) and manure (9.6 Tg of P y^{-1}) collectively exceeded P removal by harvested crops (12.3 Tg of P y^{-1}) at the global scale, P deficits covered almost 30% of the global cropland area. High P fertilizer application relative to crop P use was more typically associated with the most intense P surpluses (>13 kg P ha y^{-1}) globally, whereas manure was often a more localized driver of P surpluses in areas with high livestock densities. A 21% reduction of P fertilizer use across areas with intense surpluses associated with fertilizer use would provide enough fertilizer savings to meet the crop P demands of all P-deficit croplands globally if this P were redistributed. Changes in nutrient management across many regions will be essential to increasing global agricultural P-use efficiency.

I then conducted a comprehensive analysis of the United States agricultural system to explore how globalization is exacerbating changes to regional P cycling. This study considered how limited national P fertilizer supplies are allocated at the interface between trade, biofuel production, and diets. Total mineral P used in the US (~1.9 Tg of P y^{-1}) can be traced predominantly to accumulation in domestic agricultural soils (28%), post-harvest losses (40%), or biofuel refining (10%). Only 8% of mineral P use was ultimately consumed in domestic diets. More than half of the mineral P used in the US was devoted to feed and livestock production, largely to produce meat for domestic consumption and corn or soybean exports. One quarter of

domestic P fertilizer use was allocated to producing exports, which subsequently resulted in a sizeable P flux (0.3 Tg of P y^{-1}) from the US to other countries. However, 16% (≈0.14 Tg P) of the P fertilizer used to produce food consumed in the US was applied in other countries for imports. Trade is an overlooked but highly influential component of the modern P cycle, with important potential environmental externalities and geo-political dimensions.

Agriculturally-driven changes in soil P pools could also have long-term ecological implications given the slow cycling of P in some soils. I conducted a comprehensive meta-analysis of previous studies to understand legacies of either enriched or depleted soil labile and total P pools following agricultural abandonment throughout the world. This global meta-analysis revealed potentially large and enduring legacies of past agriculture on soil P pools across different regions and soil types, but with some reduction in the magnitude of these changes over time since abandonment. Although soil P content was typically elevated after abandonment compared to levels in nearby reference ecosystems, it was reduced compared to soils that remained under agriculture. There were more pronounced differences in the legacies of past agriculture on soil P among regions than the types of land use practiced prior to abandonment (cropland, pasture, or forage grassland). This first quantitative synthesis of soil P legacies indicated that understanding how agricultural land-use history impacts soil P pools should incorporate the roles of temporal dynamics, soil texture, alteration of soil pH, and the type of successional vegetation.

Finally, I considered the roles of watershed anthropogenic and biophysical characteristics on P loading to lakes to aid in the development of large scale lake eutrophication risk models. I used a multi-faceted statistical approach with recent global land use and hydrological data to predict lakewater total phosphorus (TP) concentrations across a representative sample of >1000 lakes worldwide. Global lake TP predictions from three unique statistical methods explained from 50% to as much as 79% of the variation in observed TP, with relatively low error rates. These models indicated the need to account for complex interdependencies between watershed biophysical context (e.g., climate and soil type) and anthropogenic drivers of P loading (e.g., the proportion of the watershed in agriculture and population density) in the determination of TP across heterogeneous lakes. These global models of TP will enable spatially detailed prediction of eutrophication risk across diverse lakes and regions.

Collectively, this work sheds new insight on agricultural modifications to the global P cycle arising from past and present nutrient management as well as how these might influence both soil P pools and water quality over time. These multi-faceted and novel studies also revealed opportunities to mitigate critical P imbalances and reduce system-wide P losses. Tackling these disparate issues will require thinking holistically about P management across scales and system components (e.g., agricultural management and water quality management). There is growing need to balance local context (e.g., soil types, management histories, and livestock densities) with the cumulative implications these have on limited P reserves at the national or global level (e.g., closing regional P imbalances and accounting for the effects of trade). Greater attention to the spatial variation in both problems and solutions related to P, as well as their complex temporal dimensions, will be essential for advancing the science and policies needed to achieve greater P sustainability in agriculture while simultaneously ensuring healthy aquatic ecosystems.

RÉSUMÉ

L'agriculture moderne a fondamentalement changée le cycle du phosphore (P) d'une façon qui dorénavant pose des défis agronomiques et environnementaux à toutes les échelles. Le P est une ressource non renouvelable qui est d'une importance cruciale à la production alimentaire car c'est un nutriment essentiel pour les plantes. En même temps, les pertes de P à partir des terres agricoles dans l'eau de ruissellement contribuent à la dégradation de la qualité de l'eau dans le monde entier. Des recherches récentes ont commencées à éclairer ces dimensions de l'utilisation P dans l'agriculture et de ses implications plus larges dans le cadre du développement durable. Nous avons tout de même une compréhension limitée sur les façons dont ces dimensions du cycle du P, qui peuvent sembler disparates, sont connectées à travers les échelles temporales et spatiales. Dans cette thèse, j'explore des lacunes importantes au niveau de nos connaissances liées aux changements spatiaux et temporels dans le mouvement du P due à l'activité humaine, leurs causes, et quelques-unes des implications pour la gestion du P dans le sol et de la qualité de l'eau à grande échelle.

Dans la première étude, j'ai effectué une analyse à l'échelle globale sur les implications de l'utilisation du P agronomique sur les sols agricoles et sur la distribution actuelle du P dans les sols agricoles. Les résultats, qui sont exprimé spatialement et d'une façon détaillé, ont démontré qu'il y a une disparité entre la quantité de P appliqué aux terres agricoles comme engrais chimiques et comme fumier et le P incorporé dans les récoltes agricoles dans la plus part des régions du monde, mais que l'ampleur de cette disparité varie considérablement entre ces régions. Bien qu'à l'échelle global l'application de P comme engrais (14,2 Tg de P an^{-1}) et comme fumier (9,6 Tg de P an^{-1}) collectivement dépasse le P incorporé dans les récoltes agricoles (12,3 Tg de P an^{-1}), un déficit de P est présent sur près de 30% de la superficie mondiale des terres cultivées. Un haut tôt d'application de P sous formes d'engrais par rapport à utilisation du P par les plantes a put généralement être associée avec les plus intenses excédents de P (> 13 kg P ha an^{-1}) à l'échelle mondiale, tandis que l'application de P sous forme de fumier est souvent une cause plus localisée d'excédents de P dans des zones à forte densité de bétail. Une réduction de 21% de l'utilisation d'engrais de P dans des zones présentant des excédents intenses associés à l'utilisation d'engrais, si redistribué sur des terres en déficits de P, répondrait aux demandes en P des cultures de toutes les terres cultivées sous déficit de P. Des changements dans la gestion du P seront

essentielles a travers plusieurs régions du monde afin d’accroître l'efficacité de l'utilisation du P dans l'agriculture.

J'ai ensuite effectué une étude approfondie du système agricole des États-Unis et ses partenaires commerciaux afin de comprendre comment la mondialisation exacerbe les changements dans le cycle du P régionale. Cette étude a examiné l’allocation des réserves nationales limitées d'engrais de P à l'interface entre le commerce, la production de biocarburants, et la diète humaine. Le P minéral total utilisé aux États-Unis (~ 2,0 Tg de P an^{-1}) peut être tracée principalement à une accumulation dans les sols agricoles domestiques (28%), les pertes après la récolte (40%), ainsi que la production de bio carburant (10%). Seulement 8% de ce P minéral a été consommée comme nourritures aux États-Unis. Plus de la moitié du P minéral utilisé aux États-Unis a été consacré à aux fourrages pour l'élevage de bétail, en grande partie pour produire de la viande pour la consommation national et pour les exportations de maïs et de soja comme fourrage. Un quart de la demande national d'engrais de P a été alloué à la production d’exportations, ce qui représente un mouvement de P non négligeable (0,3 Tg de P an^{-1}) des États-Unis vers d'autres pays. Toutefois, 16% (≈ 0,14 Tg de P) de l'engrais de P utilisée pour produire la nourriture consommée aux Etats-Unis a été appliqué dans d'autres pays et puis importer. Le commerce est une composante souvent négligée, mais très importante dans le cycle du P moderne, et donc avec une influences importante sur les externalités environnementales et sur dimensions géopolitiques de la gestion du P.

Les changements causés par agriculture sur les réservoirs de P dans sols pourraient également avoir des implications écologiques à long terme contenu de la lente vitesse à laquelle le cycle du P a lieu dans certains sols. J'ai mené une méta-analyse exhaustive des études existantes pour comprendre le rôle de l’héritage de présence de culture agricoles sur les réservoirs de P total et de P mobiles dans des sols après l'abandon agricole à travers le monde. Cette méta-analyse à l’échelle globale révèle que l’héritage d’une production et d’une gestion agricole a des effets potentiellement importants et de longue durée sur la présence de P mobile et des réserves de P total dans le sol dans les différentes régions du monde et les différents types de sols, mais cette effet est réduit d'ampleur avec le nombre d’année depuis l'abandon de la culture agricole. Bien que le P contenu dans le sol était généralement élevé après l'abandon agricole par rapport aux niveaux de P dans le sol de référence dans les écosystèmes à proximité des sites étudiés, le niveau de P était réduit par rapport au sol qui était resté en production agricole. Il y avait des différences plus marquées dans l'effet de l'héritage agricole sur le P dans le sol entre les régions du monde qu'entre les types d'agriculture avant l'abandon (terres cultivées, pâturages, ou de fourrage des prairies). Cette première synthèse quantitative de l'impact de l'héritage agricole sur le P dans le sol a indiqué que pour comprendre comment utilisation des terres agricoles affects les réservoirs de P dans le sol devrait incorporer les rôles de la dynamique temporelle, de la texture du sol, de l'altération du pH du sol, et le type de végétation de succession.

Finalement, j'ai considéré les facteurs anthropologiques déterminants l'accumulation de P dans les lacs en comparaison avec le rôle des caractéristiques biophysiques des bassins versants afin d'aider à l'élaboration de modèles sur le risque d'eutrophisation des lacs qui seront largement applicables pour plusieurs régions du monde. J'ai utilisé une approche à multiples facettes statistique pour prédire les concentrations de phosphore total (PT) dans l'eau de lac pour d'un échantillon représentatif (> 1000) de lacs dans le monde entier, à partir de cartes d'utilisation des terres mondiale et de données hydrologiques courante. Mondialement les prédictions de PT dans les lacs, à partir de trois approches statistiques uniques, expliquent entre 50% et 79% de la variation observée dans le PT, avec des taux d'erreur relativement faible.
Ces modèles démontrent la nécessité de tenir compte des interdépendances complexes entre le contexte biophysique des bassins versants (par exemple le climat et le type de sol) et les causes anthropologiques du mouvement du P vers les lacs (par exemple la proportion du bassin versant en production agricole et de la densité de population) dans la détermination de PT dans des lacs hétérogènes. Ces modèles globaux de PT permettront des prédictions spatialement détaillées du risque d'eutrophisation des lacs à travers de diverses régions du monde.

Collectivement, ce travail illustre comment les modifications agricoles du cycle du P mondiale peuvent être comprit en examinant la gestion du P dans le passé et le présent ainsi que la façon dont cette gestion peut influencer les réserves du P dans les sols et la qualité de l'eau à travers le temps. Ces études originales et à multiple facettes mettent en évidence les possibilités d'atténuer les déséquilibres critiques de P et les pertes de P dans l'ensemble du système agricoles mondiale. En tenant compte de ces résultats quantitatifs, je discute comment il faudra reconsidérer la gestion du P comme un tout, à travers les échelles temporales et spatiales ainsi qu'a travers le system d'alimentation (du la gestion agricole, jusqu'à la qualité de l'eau), pour faire face à ces défis. Il y a un besoin d'équilibrer le contexte local (par exemple les types de sol, les historiques de gestion, et des densités d'élevage) avec les effets cumulatifs que la gestion locale a sur les réserves limité de P au niveau national et mondial (par exemple diminuer les déséquilibres de P régionaux en tenant compte des effets du commerce). Une plus grande attention à la variation spatiale dans les deux dimensions de cette problématique et les solutions liés au P, ainsi que leurs dimensions temporelles complexes, sera essentielle pour faire progresser à la fois la science et les politiques nécessaires pour parvenir à une plus grande durabilité dans la gestion du P dans l'agriculture tout en veillant à la santé des écosystèmes aquatiques.

Thesis Contents

ACKNOWLEDGMENTS

Sometimes we are fortunate to encounter great teachers in life. Elena Bennett has been an inspirational mentor, an understanding supervisor, a patient writing coach, and an ever insightful educator. Thank you, Elena, for everything that you have done to help me develop as a scientist and for the invaluable guidance you have imparted along the way.

I appreciate helpful feedback and encouragement from Navin Ramankutty at committee meetings, as well as his expert guidance on the analysis of global agricultural data. Rene Gregory-Eaves provided numerous opportunities for me to collaborate with her lab, which was always an enriching experience, and her support with my professional development was a big help throughout this process. I am grateful to have had the opportunity to work with Stephen Carpenter and appreciate the insight he offered on my early research ideas. Advice from Joann Whalen and Bernhard Lehner on specific aspects of my research was very helpful.

It has been a pleasure to have had the opportunity to collaborate with several great colleagues during the past few years. In particular, I thank Günther Grill and Zofia Taranu for their unflinching support, as well as their time and dedication to our collaborative research (despite the big turns it sometimes took). I also thank Phil Potter for sharing his template computer code, used in Chapter 2, and for helping me figure out MATLAB.

Everyone in the Bennett Lab has been so great to work with. In particular, I thank Geneviève Metson, Meagan Schipanksi, Matthew Mitchell, and Kate Liss, who not only discussed aspects of my research on numerous occasions, but also commented on drafts and provided moral support. G. Metson also translated the abstract of this thesis. And to all of my friends who have listened to me talk about phosphorus throughout my graduate studies, I thank you for your unwavering encouragement and support—especially Pete.

Assistance from Jan Kowalewski with data compilation and georeferencing in Chapter 5 was extremely helpful to this research. I also thank Shauna Mahajan for some help with data compilation in Chapter 4.

I gratefully acknowledge doctoral research scholarships from the Natural Resource Sciences and Engineering Research Council of Canada (NSERC) and Le fonds de recherche du Québec - Nature et technologies (FQRNT), as well as funding through Elena Bennett's research programme.

A major thank you is owed to my parents, Lewis & Cynthia MacDonald, who helped me get to this stage and provided so much moral support and reassurance over the years. And to my brother, Alex MacDonald, who encouraged me to pursue science, I am very grateful for everything you have done for me—especially the many patient discussions we have had about my research along the way.

THESIS STRUCTURE AND AUTHOR CONTRIBUTIONS

This manuscript-based thesis is prepared in accordance with McGill University guidelines. Formatting follows the *Global Change Biology* style. The thesis contains four research articles, as well as brief literature review and synthesis chapters. I am first author on each of the chapters; in all cases, I lead the research design and topic development, lead or conducted the data compilation, conducted all statistical analyses, and lead the preparation of the manuscript text. However, several co-authors made important contributions to each of the chapters, which are detailed below.

Chapter 2 has been published:
MacDonald, G. K., Bennett, E. M., Potter, P. A., Ramankutty, N. (2011) Agronomic phosphorus imbalances across the world's croplands. *Proceedings of the National Academy of Sciences,* **108,** 3086-3091.

- Elena Bennett contributed ideas for the data analysis and rationale, and guided the manuscript preparation and revision. Phil Potter provided a template for the MATLAB computer code and the basic data structure used for the analysis. Navin Ramankutty provided feedback on the analysis (including presentation of results and discussion of uncertainty), and helped to prepare the manuscript.

Chapter 3 is under consideration for publication:
MacDonald, G. K., Bennett, E. M., Carpenter, S. R. Embodied phosphorus and the global connections of United States agriculture. Revised version of manuscript submitted to *Environmental Research Letters* (September 2012).

- Elena Bennett helped focus the topic, offered advice on various aspects of the analysis, and provided guidance on the manuscript preparation. Stephen Carpenter helped foster the idea for the research topic and provided feedback during the manuscript preparation.

Chapter 4 has been published:
MacDonald, G. K., Bennett, E. M. & Taranu, Z. E. (2012) The influence of time, soil characteristics, and land-use history on soil phosphorus legacies: a global meta-analysis. *Global Change Biology,* **18,** 1904-1917.

- Elena Bennett helped foster the idea for studying land-use legacies on soil P and provided guidance on the manuscript preparation. Zofia Taranu provided advice on the statistical analysis and feedback throughout the manuscript preparation.

Chapter 5 is in preparation for journal consideration:
MacDonald, G. K., Grill, G., Bennett, E. M., Lehner, B, Kowalewski, J. The role of watershed drivers in global predictive models of lake total phosphorus. *In preparation.*

- Günther Grill collaborated in the conceptualization of the study, contributed to the research design, conducted the watershed analysis using his hydrological models, shared data preparation responsibilities with G. MacDonald, and assisted with the chapter preparation. Elena Bennett provided guidance on the research design and chapter preparation. Bernhard Lehner provided feedback on the hydrological analysis and contributed data. Jan Kowalewski compiled data from several government lake nutrient databases and conducted the core literature search under supervision of G. MacDonald, G. Grill, and B. Lehner.

CHAPTER 1: INTRODUCTION

1.0. THE PHOSPHORUS PREDICAMENT

Phosphorus (P) plays an indispensable role in modern society, yet our management of the P cycle is riddled with simultaneous problems of over- and under-abundance (Smil 2000; Cordell *et al.* 2009; Elser & Bennett 2011). All plants and animals need P to grow. P is a critical macro-nutrient that, at the micro-scale, is a building block of cells (forming a component of DNA & RNA) and, at the macro-scale, limits primary production of some whole ecosystems (Elser 2012; Elser et al. 2007). While P is not typically thought of as a direct concern for human nutrition, we are dependent on a reliable source of P to sustain modern agriculture (Smil 2000). Inadequacies of P in certain regions, as well as general uncertainties about future access to mineral P reserves, could constrain food production (Vitousek *et al.* 2009; Townsend & Porder 2011). At the same time, excess P use that characterizes agriculture in some areas is an inefficiency that is also linked to pollution of aquatic ecosystems (Sharpley & Withers 1994; Heathwaite 2010; Kleinman *et al.* 2011). These issues require a multi-faceted understanding of how human activity is affecting the P cycle and what opportunities exist to mitigate this (Childers *et al.* 2011).

P stands alongside nitrogen and potassium as an essential plant nutrient for agricultural production and an integral component of soil fertility (i.e., the chemical characteristics of soils that support crop growth) (Brady & Weil 2004; Kirkby & Johnston 2008). For centuries, P and other nutrients have been applied to agricultural lands in soil amendments such as manure, as well as less conventional sources including fish bone and guano (Newman 1997; Verheyen *et al.* 1999; Smil 2000; Liu *et al.* 2008). However, in the absence of P inputs external to the farming system, and given very slow chemical release of P from soil parent material, historic agriculture in some regions was essentially P-limited and unsustainable in the longer-term (Newman 1997).

The natural P cycle is characterized mainly by processes occurring on a geologic time scale (Compton *et al.* 2000; Falkowski *et al.* 2000). The global P cycle has a relatively small gaseous component, relying instead on processes such as chemical weathering of soil parent material to release bioavailable P that slowly moves from land into the oceans (Compton *et al.* 2000). Human activity has fundamentally altered this slow process by introducing large quantities of bioavailable P to the biosphere (Bennett *et al.* 2001).

During the past 50 to 100 years, there has been a rapid increase in the use of mineral P fertilizes globally, coinciding with intensification of agriculture in general (Smil 2000; Tilman *et al.* 2002; Bouwman *et al.* 2011). These P fertilizers are derived from ancient sedimentary 'phosphate rocks', largely found in the calcium-phosphate form of apatite [$Ca_5(PO_4)_3OH$], that offer a readily bioavailable P source for agriculture (Liu *et al.* 2008; Villalba *et al.* 2008). Mining of phosphate rock has in turn tripled or even quadrupled P mobilization relative to background levels (Smil 2000; Bennett *et al.* 2001; Villalba *et al.* 2008). Because phosphate rocks are formed over millions of years as ocean sediment is uplifted, this critical source of mineral P is also a non-renewable resource on a human time scale—and one for which there is no substitute (Smil 2000).

In many soils, plant available P concentrations increase in step with any fertilizer P applied in excess of crop demands (Barber 1979; Sharpley 2003; Ekholm *et al.* 2005). The ability to build soil P stocks means that increases in P fertilizer use helped to alleviate soil P deficiencies for agriculture in places such as the Midwestern US and parts of Europe, where historical reliance on stocks of P in soils was not viable in the long-term (Newman 1997). More recently, there have been large increases in P fertilizer use in some rapidly developing regions, such as parts of East and South Asia, which are important for improving crop productivity but also raise environmental concerns (Shigaki *et al.* 2006; Chen *et al.* 2008; Heffer 2009; Pathak *et al.* 2010; Wang *et al.* 2011). At the same time, farmers and regions lacking access to these costly P fertilizers continue to be affected by agricultural P deficiencies (Cordell *et al.* 2009), which is a major concern for food security and malnutrition in regions such as sub-Saharan Africa (Sanchez 2002; Vitousek *et al.* 2009).

Agricultural P use is also intricately related to water quality management (Carpenter *et al.* 1998; Kleinman *et al.* 2011). Excess P can be lost from agricultural fields via runoff, sorped to eroded soil particles, or even through leaching in soils that are very P-rich (Heathwaite & Dils 2000; McDowell 2012). Primary production in many lakes is ultimately regulated by external delivery of P, wherein excessive P inputs essentially act as a fertilizer that can increase algal growth (Schindler *et al.* 2008). Elevated anthropogenic P flux in rivers has also increasingly been viewed as a major concern for coastal water quality problems, such as hypoxia (Conley 2009; Rabalais 2010; Jacobson *et al.* 2011).

Unlike carbon and nitrogen, which may cycle through soils over relatively short timescales, agriculturally-induced changes in soil P are often more prolonged (McLauchlan

2006). This is because surplus P can move to more slowly-cycling inorganic or organic forms that are less available to plants in the short term, whereas depleted P may require years of weathering to be replenished depending on soil age and inherent P supplies (e.g., Syers *et al.* 2008; Negassa & Leinweber 2009). There is typically a threshold in most soils where continued P fertilization fails to improve crop yields, but excess P accumulates and becomes more prone to moving into surface waters over time (Kleinman *et al.* 2000; Kirkby & Johnston 2008; Syers *et al.* 2008). Yet, because some aquatic systems are particularly susceptible to P inputs, recovery from past nutrient enrichment may be more difficult than simply adjusting nutrient applications on farms (Carpenter 2008; Vitousek *et al.* 2009). Mitigating eutrophication risk necessitates thinking about where and what forms of P are being applied relative to crop P uptake, potential historical legacies of past P fertilization on current soil P, and the biophysical or edaphic characteristics that might cause this P to be lost from soils via runoff (Heathwaite 2010; Kleinman *et al.* 2011).

There has been growing concern among the scientific community over potential future shortages of phosphate rock that could undermine agricultural productivity (Elser 2012). While projections of phosphate rock depletion vary from decades to hundreds of years (Cordell *et al.* 2009; Van Vuuren *et al.* 2010), and are subject to change with new mineral discoveries, this issue is nonetheless alarming given lack of substitutes and rapidly growing demand for mineral fertilizers (Elser & Bennett 2011). The distribution of phosphate rock is heavily concentrated in a handful of regions, with the Western Sahara territory in northwestern Africa possessing by far the greatest reserves (Childers *et al.* 2011). Agricultural P use therefore has an important geo-political component.

An important opportunity to mitigate the pace of either domestic or global phosphate rock depletion involves increasing the efficiency with which new mineral P is used in agriculture and ability to recycle existing P that has already entered the biosphere or food systems (Cordell *et al.* 2012). However, assessing agricultural P-use efficiency requires a detailed understanding of how P fertilizer is used in relation to agricultural productivity among different locations (Bouwman *et al.* 2009), the influence of past P use on current soil P levels (Townsend *et al.* 2002), as well as the demand for specific commodities and the magnitude of P flows associated with producing them (Schröder *et al.* 2011).

1.1. Rationale and thesis framework

The interrelated issues of 'too much' and 'too little' facing our management of the modern, human-dominated P cycle are increasingly being viewed as a critical sustainability challenge for 21st century agriculture (Childers *et al.* 2011; Elser & Bennett 2011). This challenge requires systematic consideration of the efficiency with which P is used in agriculture, its implications at different spatial and temporal scales for distinct components of the P cycle, as well as opportunities to recover P losses for reuse.

In this thesis, I explore the theme of *human alteration of P flows globally* (Fig. 1.1) with two synergistic sub-themes that are related to environmental management (*agriculture as a driver of soil P* and *lake eutrophication risk related to drivers of P loading*). I identified three important knowledge gaps in our understanding of how human activity is influencing these aspects of the modern P cycle at large spatial and temporal scales (*global agricultural P management*; *demand for P fertilizers and its relationship to agricultural trade*; as well as *land-use legacies on soil P*). I discuss how we might achieve more efficient agricultural P use globally, as well as some of the broader implications of human activity for management of soil P and freshwater quality at different scales.

1.1.1. Human alteration of P flows

A fundamental aspect of the human dominated P cycle relates to how we have accelerated or entirely changed the locations where P is introduced on the landscape and in what quantities. The natural P cycle relies on processes such as wind or water transportation to move P over the landscape, or slow, geologic processes to release bioavailable P to ecosystems (Compton *et al.* 2000). Use of mineral P fertilizers allows for this slow cycling to be bypassed (Villalba *et al.* 2008), with the 'translocation' of P from concentrated phosphate rock deposits in one region to potentially disparate locations globally. In other words, changes in soil P related to farming in one region, such as northern Europe (where there are no substantial phosphate rock reserves) may involve P imported from another region (such as northern Africa, where reserves are concentrated) (Beaton *et al.* 1995; Cordell *et al.* 2009).

Understanding the geographic dimension of P necessitates pinpointing how P is being used in different places and why (Potter *et al.* 2010). This is increasingly possible with more detailed global land use data (e.g., Monfreda *et al.* 2008; Foley *et al.* 2011). The issue of 'why' P fertilizer is used further relates to changes in agricultural demand (e.g., growing meat consumption from industrial production and increased use of biofuels such as corn ethanol), as well as globalization of agricultural systems via trade.

However, our ability to translate this understanding of altered P cycling into useable models for ecosystem management is inhibited by two important factors: (1) a relatively limited understanding of the temporal dimensions of soil P relating to past agriculture (i.e., land-use legacies); and (2) limited large-scale models of P loading to lakes necessary to assess spatial patterns of eutrophication risk globally. My research offers insight to help bridge these divides.

Global agricultural P management

While our aggregate understanding of how human activity is impacting the global P cycle has been informed by several studies (e.g., Smil 2000; Bennett *et al.* 2001; Liu *et al.* 2008; Cordell *et al.* 2009), these studies have not offered detailed insight on the geographic patterns of agricultural P use. Because agricultural production and management is often spatially heterogeneous, it is likely to have a large impact on the spatial patterns of P use and their implications (Bouwman *et al.* 2009; Foley *et al.* 2011). A simple indicator of this is the soil-surface nutrient balance, which examines the directly managed inputs of P to agricultural soils via fertilizer and manure relative to how much P is removed via crop harvest (Oenema *et al.* 1999; Ekholm *et al.* 2005), providing insight on the potential for soil P accumulation or whether soil P stocks may be subject to gradual declines. In Chapter 2, I explore this using a spatially detailed assessment of soil-surface P balances globally. Specifically, I ask: ***What are the geographic patterns and implications of P management for potentially elevating or depleting regional soil P?***

Demand for P fertilizer use and agricultural trade

Changing consumption patterns for agricultural commodities, and particularly growth in agricultural trade, requires a shift in our thinking about the global P cycle. For example, non-renewable P fertilizers are increasingly being used for biofuel production (Hein & Leemans 2012), while considerably more fertilizer use may be required to produce meat than simply producing crops directly for human consumption (Foley *et al.* 2011). Intensive livestock production exacerbates regional P cycling primarily because the mineral P from livestock feeds is poorly recycled, with manure use being concentrated on farms nearby to high-density livestock holdings (Naylor *et al.* 2005; Menzi *et al.* 2010). Increasing demand for meat is also related to growing international trade in feed crops and meat (Galloway *et al.* 2007). Trade has the potential to greatly alter regional P fluxes simply by moving P contained in traded goods (Grote *et al.* 2005), but also results in linkages in P fertilizer and manure use among regions (Matsubae *et al.* 2011; Schipanski & Bennett 2012). While previous studies have touched on these issues, there is a need to elucidate how trade relates to various aspects of agricultural production within and among regions from a full-system perspective (Wang *et al.* 2011; Senthilkumar *et al.* 2012). In Chapter 3, I build on work from Chapter 2 to examine this topic for a key agricultural producing and consuming nation (the United States). I ask the following questions: ***What implications does the allocation of mineral P use to the production of certain commodities have on overall P flows and losses in the US agricultural system?;*** and ***How much of these P flows are linked to international agricultural trade?***

1.1.2. Implications for environmental management: soil P and water quality

Agricultural land-use legacies on soil P

If agricultural amendments or crop harvest drive changes in soil P, the altered supply of P in soils could have important implications for future land use. For example, it could alter the amount of future P fertilization required to meet crop requirements (Sattari *et al.* 2012), the quantity of P accumulated in the soil matrix that could move into adjacent surface waters (Kleinman *et al.* 2011), or the composition of the ecological communities occupying a post-agricultural ecosystem if land-use change occurs (Baeten *et al.* 2010). These long-term dynamics of soil P are considerably less well understood compared to other elements, such as carbon, with studies from individual locations often finding conflicting trends for soil P legacies (McLauchlan 2006). Confounding this further is the fact that the exchange of soil P between different forms (inorganic/organic, readily available/fixed) occurs on shorter timescales than previously thought and is potentially influenced by land use (Richter *et al.* 2006; Syers *et al.* 2008; Negassa & Leinweber 2009). In Chapter 4, I tackle this knowledge gap by considering the response of soil P (both the relatively small portion that is plant available and the total stock of phosphorus) to past agriculture across a large number of studies from around the world, asking: ***Are there generalizable legacies of past agriculture on current soil P across regions?;*** and ***What factors contribute most to these land-use legacies on soil P?***

P as a driver of lake eutrophication risk

A major emphasis in limnology has been to understand P cycling within lakes of different regions (Kalff 2002). Numerous empirical models have been developed to predict lake total phosphorus (TP) concentrations based on factors such as external P inputs, P uptake or release from lake sediments, and the P flux from lakes in discharge (e.g., Ahlgren *et al.* 1988; Håkanson 1995). A key gap in these models relates to quantifying the *external* P inputs, which is determined by the confluence of complex human activities and biophysical processes within a lake's watershed (Heathwaite 2010). While many studies have addressed watershed sources of P to understand factors contributing to lake eutrophication risk, most studies assessing anthropogenic influence on P loading have considered lakes within relatively small geographic regions (see review by Taranu & Gregory-Eaves 2008) or have used aggregate models that are not specific to individual lakes (e.g., global work by Carpenter & Bennett (2011)). In Chapter 5, I develop broadly applicable predictive models of lake TP using a large dataset of lakes located around the world, in conjunction with various global land use, hydrological, and biophysical data. In particular, I emphasize the usefulness of key indicators of anthropogenic P loading (agriculture and population density), integrating across concepts from Chapters 2, 3, and 4. This research explores the following questions: ***How do watershed biophysical characteristics and human drivers interact to determine lake TP across a diverse set of lakes?;*** and ***Is it possible to obtain accurate predictions of TP for individual lakes by using relatively coarse estimates of watershed characteristics derived from various global datasets?***

A sample of some recent large-scale studies on P shows that these often correspond to specific subsystems of the modern P cycle. For example, several studies have focussed on P use relating primarily to agricultural soils (e.g., Liu *et al.* 2008; Bouwman *et al.* 2009; Bateman *et al.* 2011; Bouwman *et al.* 2011; Sattari *et al.* 2012; Schipanski & Bennett 2012), sustainability issues surrounding more distributional aspects of P use (e.g., Villalba *et al.* 2008; Cordell *et al.* 2009; Suh & Yee 2011; Wang *et al.* 2011; Hein & Leemans 2012), or the ecological effects of P use (e.g., Harrison *et al.* 2010; Carpenter & Bennett 2011). Each of the novel studies in my thesis lays groundwork for collectively addressing these components of the P cycle, which is intended to help bridge gaps in current P research for a more cross-disciplinary understanding of the human-dominated P cycle.

THESIS CONCEPTUAL MODEL

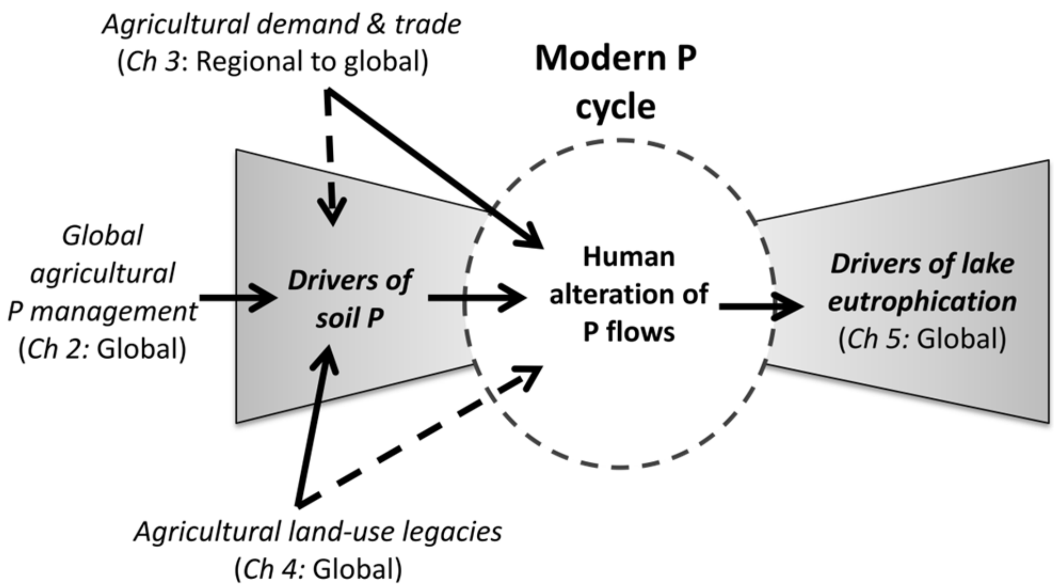


Fig. 1.1. Conceptual model identifying interactions among the thesis research topics (Ch. 2, 3, 4, and 5), with emphasis on how these relate to understanding human alteration of the modern P cycle in general. The sides of the diagram shaded in grey indicate issues related to ecosystem management (drivers of soil P and drivers of lake water quality). Solid arrows indicate direct linkages among themes while dotted arrows show indirect linkages.

1.2. References

Ahlgren, I., Frisk, T. & Kamp-Nielsen, L. (1988) Empirical and theoretical models of phosphorus loading, retention and concentration vs. Lake trophic state. *Hydrobiologia,* **170,** 285-303.

Baeten, L., Vanhellemont, M., De Frenne, P., De Schrijver, A., Hermy, M. & Verheyen, K. (2010) Plasticity in response to phosphorus and light availability in four forest herbs. *Oecologia,* **163,** 1021-1032.

Barber, S. A. (1979) Soil phosphorus after 25 years of cropping with five rates of phosphorus application. *Communications in Soil Science and Plant Analysis,* **10,** 1459-1468.

Bateman, A., van der Horst, D., Boardman, D., Kansal, A. & Carliell-Marquet, C. (2011) Closing the phosphorus loop in england: The spatio-temporal balance of phosphorus capture from manure versus crop demand for fertiliser. *Resources, Conservation and Recycling,* **55,** 1146-1153.

Beaton, J., Roberts, T., Halstead, E. & Cowell, L. (1995) Global transfers of p in fertilizer materials and agricultural commodities. *Phosphorus in the global environment: Transfers, cycles and management scope 54* (eds. H. Tiessen). Wiley, Chichester.

Bennett, E. M., Carpenter, S. R. & Caraco, N. F. (2001) Human impact on erodable phosphorus and eutrophication: A global perspective. *Bioscience,* **51,** 227-234.

Bouwman, A. F., Beusen, A. H. W. & Billen, G. (2009) Human alteration of the global nitrogen and phosphorus soil balances for the period 1970-2050. *Global Biogeochemical Cycles,* **23**.

Bouwman, L., Goldewijk, K. K., Van Der Hoek, K. W., Beusen, A. H. W., Van Vuuren, D. P., Willems, J., Rufino, M. C. & Stehfest, E. (2011) Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period. *Proceedings of the National Academy of Sciences,* **Early Edition,** 1-6.

Brady, N. C. & Weil, R. R. (2004) *Elements of the nature and properties of soils*. Prentice Hall, Upper Saddle River, NJ.

Carpenter, S. R. (2008) Phosphorus control is critical to mitigating eutrophication. *Proceedings of the National Academy of Sciences,* **105,** 11039-11040.

Carpenter, S. R. & Bennett, E. M. (2011) Reconsideration of the planetary boundary for phosphorus. *Environmental Research Letters,* **6,** 014009.

Carpenter, S. R., Caraco, N. F., Correll, D. L., Howarth, R. W., Sharpley, A. N. & Smith, V. H. (1998) Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecological Applications,* **8,** 559-568.

Chen, M., Chen, J. & Sun, F. (2008) Agricultural phosphorus flow and its environmental impacts in china. *Science of The Total Environment,* **405,** 140-152.

Childers, D. L., Corman, J., Edwards, M. & Elser, J. J. (2011) Sustainability challenges of phosphorus and food: Solutions from closing the human phosphorus cycle. *Bioscience,* **61,** 117-124.

Compton, J., Mallinson, D., Glenn, C. R., Filippelli, G., Follmi, K., Shields, G. & Zanin, Y. (2000) Variations in the global phosphorus cycle. *Marine Authigenesis: From Global to Microbial. Special publication of the Society for Sedimentary Geoglogy (SEPM),* **66,** 21-34.

Conley, D. J. (2009) Controlling eutrophication: Nitrogen and phosphorus. *Science,* **323,** 1014-1015.

Cordell, D., Drangert, J. O. & White, S. (2009) The story of phosphorus: Global food security and food for thought. *Global Environmental Change,* **19,** 292-305.
Cordell, D., Neset, T. S. S. & Prior, T. (2012) The phosphorus mass balance: Identifying 'hotspots' in the food system as a roadmap to phosphorus security. *Current Opinion in Biotechnology,* ***In Press***.
Ekholm, P., Turtola, E., Grönroos, J., Seuri, P. & Ylivainio, K. (2005) Phosphorus loss from different farming systems estimated from soil surface phosphorus balance. *Agriculture, Ecosystems & Environment,* **110,** 266-278.
Elser, J. & Bennett, E. (2011) Phosphorus cycle: A broken biogeochemical cycle. *Nature,* **478,** 29-31.
Elser, J. J. (2012) Phosphorus: A limiting nutrient for humanity? *Current Opinion in Biotechnology,* ***In press***.
Falkowski, P., Scholes, R. J., Boyle, E., Canadell, J., Canfield, D., Elser, J., Gruber, N., Hibbard, K., Högberg, P., Linder, S., Mackenzie, F. T., Moore III, B., Pedersen, T., Rosenthal, Y., Seitzinger, S., Smetacek, V. & Steffen, W. (2000) The global carbon cycle: A test of our knowledge of earth as a system. *Science,* **290,** 291-296.
Foley, J. A., Ramankutty, N., Brauman, K. A., Cassidy, E. S., Gerber, J. S., Johnston, M., Mueller, N. D., O'Connell, C., Ray, D. K., West, P. C., Balzer, C., Bennett, E. M., Carpenter, S. R., Hill, J., Monfreda, C., Polasky, S., Rockstrom, J., Sheehan, J., Siebert, S., Tilman, D. & Zaks, D. P. M. (2011) Solutions for a cultivated planet. *Nature,* **478,** 337-342.
Galloway, J. N., Burke, M., Bradford, G. E., Naylor, R., Falcon, W., Chapagain, A. K., Gaskell, J. C., McCullough, E., Mooney, H. A., Oleson, K. L. L., Steinfeld, H., Wassenaar, T. & Smil, V. (2007) International trade in meat: The tip of the pork chop. *AMBIO: A Journal of the Human Environment,* **36,** 622-629.
Grote, U., Craswell, E. & Vlek, P. (2005) Nutrient flows in international trade: Ecology and policy issues. *Environmental Science & Policy,* **8,** 439-451.
Håkanson, L. (1995) Models to predict lake annual mean total phosphorus. *Journal of Aquatic Ecosystem Stress and Recovery (Formerly Journal of Aquatic Ecosystem Health),* **4,** 25-58.
Harrison, J. A., Bouwman, A. F., Mayorga, E. & Seitzinger, S. (2010) Magnitudes and sources of dissolved inorganic phosphorus inputs to surface fresh waters and the coastal zone: A new global model. *Global Biogeochem. Cycles,* **24,** GB1003.
Heathwaite, A. L. (2010) Multiple stressors on water availability at global to catchment scales: Understanding human impact on nutrient cycles to protect water quality and water availability in the long term. *Freshwater Biology,* **55,** 241-257.
Heathwaite, A. L. & Dils, R. M. (2000) Characterising phosphorus loss in surface and subsurface hydrological pathways. *Science of The Total Environment,* **251–252,** 523-538.
Heffer, P. (2009) Assessment of fertilizer use by crop at the global level 2006/07 – 2007/08. pp. 1-12. International Fertilizers Industry Association, Paris, France.
Hein, L. & Leemans, R. (2012) The impact of first-generation biofuels on the depletion of the global phosphorus reserve. *AMBIO: A Journal of the Human Environment***,** 1-9.
Jacobson, L. M., David, M. B. & Drinkwater, L. E. (2011) A spatial analysis of phosphorus in the mississippi river basin. *J. Environ. Qual.,* **40,** 931-941.
Kalff, J. (2002) *Limnology: Inland water ecosystems*. Prentice-Hall., Upper Saddle River, NJ.

Kirkby, E. A. & Johnston, A. E. (2008) Soil and fertilizer phosphorus in relation to crop nutrition the ecophysiology of plant-phosphorus interactions. (eds. P. J. White & J. P. Hammond), pp. 177-223. Springer Netherlands.

Kleinman, P. J. A., Bryant, R. B., Reid, W. S., Sharpley, A. N. & Pimentel, D. (2000) Using soil phosphorus behavior to identify environmental thresholds. *Soil Science,* **165,** 943-950.

Kleinman, P. J. A., Sharpley, A. N., McDowell, R. W., Flaten, D. N., Buda, A. R., Tao, L., Bergstrom, L. & Zhu, Q. (2011) Managing agricultural phosphorus for water quality protection: Principles for progress. *Plant and Soil,* **349,** 169-182.

Liu, Y., Villalba, G., Ayres, R. U. & Schroder, H. (2008) Global phosphorus flows and environmental impacts from a consumption perspective. *Journal of Industrial Ecology,* **12,** 229-247.

Matsubae, K., Kajiyama, J., Hiraki, T. & Nagasaka, T. (2011) Virtual phosphorus ore requirement of japanese economy. *Chemosphere,* **84,** 767-772.

McDowell, R. W. (2012) Minimising phosphorus losses from the soil matrix. *Current Opinion in Biotechnology,* ***In press***.

McLauchlan, K. (2006) The nature and longevity of agricultural impacts on soil carbon and nutrients: A review. *Ecosystems,* **9,** 1364-1382.

Menzi, H., Oenema, O., Burton, C., Shipin, O., Gerber, P., Robinson, T. & Franceschini, G. (2010) Impacts of intensive livestock production and manure management on the environment. (eds. H. Steinfeld, H. Mooney, F. Schneider & L. Neville), pp. 416. Island Press, Washington, DC.

Monfreda, C., Ramankutty, N. & Foley, J. A. (2008) Farming the planet: 2. Geographic distribution of crop areas, yields, physiological types, and net primary production in the year 2000. *Global Biogeochemical Cycles,* **22**.

Naylor, R., Steinfeld, H., Falcon, W., Galloway, J., Smil, V., Bradford, E., Alder, J. & Mooney, H. (2005) Losing the links between livestock and land. *Science,* **310,** 1621-1622.

Negassa, W. & Leinweber, P. (2009) How does the hedley sequential phosphorus fractionation reflect impacts of land use and management on soil phosphorus: A review. *Journal of Plant Nutrition and Soil Science,* **172,** 305-325.

Newman, E. I. (1997) Phosphorus balance of contrasting farming systems, past and present: Can food production be sustainable? *Journal of Applied Ecology,* **34,** 1334-1347.

Oenema, O., Heinen, M., Smaling, E. M. & Fresco, L. O. (1999) Nutrient disequilibria in agroecosystems. *Uncertainties in Nutrient Budgets Due to Biases and Errors: Concepts and Case Studies***,** 75-97.

Pathak, H., Mohanty, S., Jain, N. & Bhatia, A. (2010) Nitrogen, phosphorus, and potassium budgets in indian agriculture. *Nutrient Cycling in Agroecosystems,* **86,** 287-299.

Potter, P., Ramankutty, N., Bennett, E. M. & Donner, S. D. (2010) Characterizing the spatial patterns of global fertilizer application and manure production. *Earth Interactions,* **14,** 1-22.

Rabalais, N. N., Díaz, R. J., Levin, L. A., Turner, R. E., Gilbert, D., and Zhang, J. (2010) Dynamics and distribution of natural and human-caused hypoxia. *Biogeosciences,* **7,** 585-619.

Richter, D., Allen, H., Li, J., Markewitz, D. & Raikes, J. (2006) Bioavailability of slowly cycling soil phosphorus: Major restructuring of soil p fractions over four decades in an aggrading forest. *Oecologia,* **150,** 259-271.

Sanchez, P. A. (2002) Soil fertility and hunger in africa. *Science,* **295,** 2019-2020.

Sattari, S. Z., Bouwman, A. F., Giller, K. E. & van Ittersum, M. K. (2012) Residual soil phosphorus as the missing piece in the global phosphorus crisis puzzle. *Proceedings of the National Academy of Sciences,* **109,** 6348-6353.

Schindler, D. W., Hecky, R. E., Findlay, D. L., Stainton, M. P., Parker, B. R., Paterson, M. J., Beaty, K. G., Lyng, M. & Kasian, S. E. M. (2008) Eutrophication of lakes cannot be controlled by reducing nitrogen input: Results of a 37-year whole-ecosystem experiment. *Proceedings of the National Academy of Sciences,* **105,** 11254-11258.

Schipanski, M. & Bennett, E. (2012) The influence of agricultural trade and livestock production on the global phosphorus cycle. *Ecosystems,* **15,** 256-268.

Schröder, J. J., Smit, A. L., Cordell, D. & Rosemarin, A. (2011) Improved phosphorus use efficiency in agriculture: A key requirement for its sustainable use. *Chemosphere,* **84,** 822-831.

Senthilkumar, K., Nesme, T., Mollier, A. & Pellerin, S. (2012) Conceptual design and quantification of phosphorus flows and balances at the country scale: The case of france. *Global Biogeochem. Cycles,* **26,** GB2008.

Sharpley, A. N., T. Daniel, T. Sims, J. Lemunyon, R. Stevens, and R. Parry. (2003) Agricultural phosphorus and eutrophication, 2nd ed., pp. 44. U.S. Department of Agriculture, Agricultural Research Service.

Sharpley, A. N. & Withers, P. J. A. (1994) The environmentally-sound management of agricultural phosphorus. *Fertilizer Research,* **39,** 133-146.

Shigaki, F., Sharpley, A. & Prochnow, L. I. (2006) Animal-based agriculture, phosphorus management and water quality in brazil: Options for the future. *Scientia Agricola,* **63,** 194-209.

Smil, V. (2000) Phosphorus in the environment: Natural flows and human interferences. *Annual Review of Energy and the Environment,* **25,** 53-88.

Suh, S. & Yee, S. (2011) Phosphorus use-efficiency of agriculture and food system in the us. *Chemosphere,* **84,** 806-813.

Syers, J. K., Johnson, A. H. & Curtin, D. (2008) Efficiency of soil and fertilizer phosphorus use: Reconciling changing concepts of soil phosphorus behaviour with agronomic information. FAO Fertilizer and Plant Nutrient Bulletin: 18. Food and Agriculture Organization of the United Nations, Rome, Italy.

Taranu, Z. & Gregory-Eaves, I. (2008) Quantifying relationships among phosphorus, agriculture, and lake depth at an inter-regional scale. *Ecosystems,* **11,** 715-725.

Tilman, D., Cassman, K. G., Matson, P. A., Naylor, R. & Polasky, S. (2002) Agricultural sustainability and intensive production practices. *Nature,* **418,** 671-677.

Townsend, A. R., Asner, G. P., Cleveland, C. C., Lefer, M. E. & Bustamante, M. M. C. (2002) Unexpected changes in soil phosphorus dynamics along pasture chronosequences in the humid tropics. *Journal of Geophysical Research,* **107,** 8067.

Townsend, A. R. & Porder, S. (2011) Boundary issues. *Environmental Research Letters,* **6,** 011001.

Van Vuuren, D. P., Bouwman, A. F. & Beusen, A. H. W. (2010) Phosphorus demand for the 1970-2100 period: A scenario analysis of resource depletion. *Global Environmental Change,* **20,** 428-439.

Verheyen, K., Bossuyt, B., Hermy, M. & Tack, G. (1999) The land use history (1278-1990) of a mixed hardwood forest in western belgium and its relationship with chemical soil characteristics. *Journal of Biogeography,* **26,** 1115-1128.

Villalba, G., Liu, Y., Schroder, H. & Ayres, R. U. (2008) Global phosphorus flows in the industrial economy from a production perspective. *Journal of Industrial Ecology,* **12,** 557-569.

Vitousek, P. M., Naylor, R., Crews, T., David, M. B., Drinkwater, L. E., Holland, E., Johnes, P. J., Katzenberger, J., Martinelli, L. A., Matson, P. A., Nziguheba, G., Ojima, D., Palm, C. A., Robertson, G. P., Sanchez, P. A., Townsend, A. R. & Zhang, F. S. (2009) Nutrient imbalances in agricultural development. *Science,* **324,** 1519-1520.

Wang, F., Sims, J. T., Ma, L., Ma, W., Dou, Z. & Zhang, F. (2011) The phosphorus footprint of china's food chain: Implications for food security, natural resource management, and environmental quality. *J. Environ. Qual.,* **40,** 1081-1089.

CHAPTER 2: AGRONOMIC PHOSPHORUS IMBALANCES ACROSS THE WORLD'S CROPLANDS

Graham K. MacDonald, Elena M. Bennett, Philip P. Potter, Navin Ramanutty

Published in *Proceedings of the National Academy of Sciences (PNAS)*

2.0. ABSTRACT

Increased phosphorus (P) fertilizer use and livestock production has fundamentally altered the global P cycle. We calculated spatially-explicit P balances for cropland soils at 0.5 degree resolution based on the principal agronomic P inputs and outputs associated with production of 123 crops globally for the year 2000. Although agronomic inputs of P fertilizer (14.2 Tg P yr^{-1}) and manure (9.6 Tg P yr^{-1}) collectively exceeded P removal by harvested crops (12.3 Tg P yr^{-1}) at the global scale, P deficits covered almost 30% of the global cropland area. There was massive variation in the magnitudes of these P imbalances across most regions, particularly Europe and South America. High P fertilizer application relative to crop P use resulted in a greater proportion of the intense P surpluses (>13 kg P ha^{-1} yr^{-1}) globally than manure P application. High P fertilizer application was also typically associated with areas of relatively low P-use efficiency. Although manure was an important driver of P surpluses in some locations with high livestock densities, P deficits were common in areas producing forage crops used as livestock feed. Resolving agronomic P imbalances may be possible with more efficient use of P fertilizers and more effective recycling of manure P. Such reforms are needed to increase global agricultural productivity while maintaining or improving freshwater quality.

2.1. INTRODUCTION

Disparities between the nutrients applied to agricultural soils via fertilizer or manure and the nutrients removed by harvested crops result in nutrient imbalances that can influence environmental quality and productivity of agricultural systems (Vitousek *et al.* 2009). Growing consumption of inorganic phosphorus (P) fertilizers derived from mining of non-renewable phosphate rock (Cordell *et al.* 2009) has contributed to major increases in crop yields since the 1950s (Tilman *et al.* 2002). Concurrent growth in fertilizer use and livestock production has more than tripled global P flows to the biosphere over preindustrial levels (Smil 2000), resulting in P accumulation in some agricultural soils that acts as a driver of eutrophication in freshwater and coastal systems (Sharpley & Withers 1994; Bennett *et al.* 2001; Rabalais 2010). At the same time, limited availability of P fertilizers in other regions has contributed to prolonged P deficits that can deplete soil P and limit crop yields (Vlek *et al.* 1997; Roy *et al.* 2003; Sheldrick & Lingard 2004). Although agricultural P surpluses and deficits have been documented for several regions (e.g., Gerber *et al.* 2005; Cobo *et al.* 2010), there is still limited understanding of the spatial patterns of P imbalances at the global scale.

Patterns of nutrient imbalances across agricultural systems may reflect contrasting agricultural practices, economic development, and broader agricultural policies (Oenema *et al.* 1999; Vitousek *et al.* 2009). Understanding agricultural P use is key to managing global phosphate rock reserves (Van Vuuren *et al.* 2010) and mitigating the risk for potentially irreversible eutrophication of lakes (Carpenter 2008). Despite considerable advances in the development of spatially-explicit global nitrogen balances (e.g., Liu *et al.* 2010), most previous global P balance studies have relied on globally or regionally aggregated data (Smil 2000; Bennett *et al.* 2001; Sheldrick *et al.* 2002; Liu *et al.* 2008), limiting our ability to infer spatial patterns of surpluses and deficits. The only spatially-explicit global P balance study that we are aware of used estimates of inputs and outputs based primarily on regional or national agricultural statistics distributed over four aggregated cropping systems using the IMAGE model (Bouwman *et al.* 2009). Here, we use empirical data to calculate P balances for croplands circa the year 2000 at 0.5 degree resolution in latitude and longitude (~50 x 50 km^2) to examine patterns of agronomic P imbalances globally. These P balances were calculated using spatial estimates of the principal agronomic P inputs (P fertilizer and manure applications) and outputs (P in harvested crops) for cropland soils based on spatially-explicit global maps of more than 100 crops.

2.2. RESULTS

2.2.1. Spatial patterns of agronomic P imbalances

We classified P surpluses and deficits by quartiles to compare P imbalances across all regions globally (Fig. 1). In total, 29% of the global cropland area had overall P deficits and 71% of the cropland area had overall P surpluses. A sizeable fraction of the global cropland area (~31%) had only small negative or positive imbalances (within ±2 kg P ha^{-1} yr^{-1} from zero), corresponding to the lowest two quartiles for deficits and the lowest quartile for surpluses (Fig. 2). These minor imbalances occurred in every region, but were most prevalent in Africa.

Moderate P imbalances (lower-middle and upper-middle quartiles of surpluses (3 to 13 kg P ha^{-1} yr^{-1}) and upper-middle quartile for deficits (-2 to -3 kg P ha^{-1} yr^{-1})) were characteristic of croplands in every region except Africa, occurring in 47% of croplands globally. The largest share of moderate surpluses occurred in South Asia (India, Pakistan and Thailand) and North and Central America (United States, Canada and Mexico). Moderate deficits occurred in only 8% of the global cropland area, largely in Eastern Europe (Russia and Ukraine) and West Africa, as well as smaller tracts of other regions, such as south-eastern Australia.

The largest imbalances of agronomic P, corresponding to the top quartiles of both deficits and surpluses (-3 to -39 kg P ha^{-1} yr^{-1} and 13 to 840 kg P ha^{-1} yr^{-1}, respectively), were spatially concentrated in certain areas. Just 10% of the global cropland area with the largest P deficits contributed 65% of the cumulative global P deficit (Fig. 3). The most widespread large deficits were in South America (particularly Argentina and Paraguay), the northern United States, and Eastern Europe. Similarly, 10% of the cropland area with the largest surpluses contributed 45% of the cumulative global P surplus. These large surpluses (which had a median value of 26 kg P ha^{-1} yr^{-1}) covered most of East Asia, as well as sizeable tracts of Western and Southern Europe, the coastal United States, and southern Brazil, but less than 2% of the cropland in Africa (Fig. 2). A more detailed breakdown of the P balance quartile ranges and variations by continent highlights the particularly large intra-regional variation in agronomic P imbalances in Europe and South America (Fig. S1).

2.2.2. Global agronomic P flows

Fertilizer application to croplands in the year 2000 totalled 14.2 Tg P yr^{-1}, of which more than half was applied to cereal crops. The largest P fertilizer application rates occurred predominantly in East Asia, Western Europe, the Midwestern United States, and southern Brazil (Fig S2 and see Potter et al. (2010)). Approximately 9.6 Tg P yr^{-1}, or 40% of total manure P excreted by livestock in 2000 (Potter *et al.* 2010), was used for cropland application based on estimates of recoverable manure for 12 regions (Sheldrick *et al.* 2003) and for U.S. states (Kellogg *et al.* 2000). Recoverable manure P shows much greater spatial variation than P fertilizer applications (Fig. S2), with clusters of more intense manure P applications occurring in many countries (such as the United States and Brazil) and more widespread high manure P applications in East Asia and Western Europe.

The production of 123 crops in the year 2000 (Monfreda *et al.* 2008) removed 12.3 Tg P yr^{-1} from cropland soils. The greatest crop P removal occurred in the northern United States, Western Europe, East Asia, South America (particularly southern Brazil and Argentina), and Australia, largely reflecting crop yields. Cereal crops accounted for about half and by far the largest share of P removal, most of which was attributable to harvest of wheat, maize, and rice.

Our global estimate of total P inputs to cropland soils exceeds P removed by harvested crops, resulting in a global agronomic surplus of 11.5 Tg P yr^{-1}. We also calculated P balances based on contrasting crop residue management scenarios using plausible high and low residue recycling and removal estimates from Smil (Smil 1999) that reflect broad differences in residue management between developed and developing countries (detailed in the SI text). The high residue removal scenario resulted in a slight decrease in our global P balance estimate (11.1 Tg P yr^{-1}), whereas the low residue removal scenario resulted in a considerable increase (to 12.5 Tg P yr^{-1}) due to the influence of residue P recycling inputs. These crop residue scenarios had minimal influence on the spatial patterns of P surpluses and deficits (Fig. S3).

2.2.3. Agronomic drivers of cropland P imbalances

We found considerable spatial variation in the main drivers of P surpluses based on the magnitudes of fertilizer and manure inputs relative to crop P use (using crop P removal as a proxy for crop use) (Fig. 4A). Fertilizer alone exclusive of manure inputs exceeded crop P use in the largest fraction of P-surplus cropland in all continents except Africa (Fig. 4B), and particularly in intensive agricultural regions of Asia and North America (40% of the cropland area in each continent). The combination of fertilizer and manure was the primary driver of P surpluses in about 30% of the global cropland area with P surpluses; manure and fertilizer each individually exceeded crop use in half of this area, particularly in southern China and eastern Brazil, whereas the sum of fertilizer plus manure exceeded crop use in the remaining half. Manure P alone exclusive of fertilizer P exceeded crop use in only 11% of croplands globally, particularly in areas with high livestock densities but relatively limited cropland areas (e.g., parts of the United States) or in regions with relatively low P fertilizer application and low P surpluses (e.g., across central Africa).

Half of the cropland area with the largest P surpluses (>13 kg P ha^{-1} yr^{-1}) globally corresponded to locations where P fertilizer and manure applications each individually exceeded crop P use, but P fertilizer application alone was particularly influential in some regions. When summed across both categories (Fig. 4B), P fertilizer applications exceeding crop P use coincided with a greater proportion (87%) of the global cropland area that had large P surpluses compared to manure P applications in excess of crop P use (62%). In particular, a much greater proportion of the large P surpluses in Asia and South America corresponded to locations where P fertilizer application alone exceeded crop P use compared to areas where manure P alone exceeded crop P use. Roughly the same proportion of cropland areas with large P surpluses in Europe and North America corresponded to locations where either fertilizer or manure P, or both, exceeded crop P use.

The types of crops grown contributed substantially to the locations of deficits. Forage crops, and particularly grasses, were associated with large P deficits in several regions. These crops received approximately 5% of the total global P fertilizer application in countries with crop-specific fertilizer data in 2000 (which collectively represent 95% of global P fertilizer inputs), yet they accounted for more than 20% of global crop P removal. Approximately 13% of crop P removal globally was attributable to mixed leguminous grasses and alfalfa, which may receive manure applications but are only fertilized in a few countries (IFA/IFDC/IPI/PPI/FAO 2002). Non-forage croplands in several areas had small or moderate P surpluses (e.g., throughout the United States and Australia) (Fig. S4), confirming that some P deficits (Fig. 1) were linked to harvest of forage crops. Forage crops were less influential for P deficits in other locations (e.g., Argentina and Nigeria) (Fig. S4; see SI text for further explanation). For example, the concentration of top quartile deficits in South America was primarily related to soybean harvest in Argentina, and to a lesser extent, harvest of grasses and wheat. Soybean received on average 2.5 kg P ha^{-1} yr^{-1} of P fertilizer in Argentina circa 2000 (10% of the reported P fertilizer rate for soybean in neighbouring Brazil (IFA/IFDC/IPI/PPI/FAO 2002)), which was a small fraction of the P removal rate for soybean (15 kg P ha^{-1} yr^{-1}).

2.2.4. Phosphorus-use efficiency

To assess the relationship between P imbalances and overall crop productivity, we calculated a map of agronomic P-use efficiency (PUE), defined here as total crop dry-matter production per unit of P applied (kg crop · kg P input^{-1}; Fig. 5) (Dobermann & Cassman 2005). This method incorporates the contribution of both agronomic P inputs and existing soil nutrients to crop production (Dobermann & Cassman 2002). High PUE values can therefore indicate large crop production returns per unit of P applied (e.g., where high PUE coincides with P surpluses) or reliance on soil P depletion for crop production (e.g., where high PUE coincides with P deficits). Variation in PUE also reflects the types of crops grown and differences in productivity across regions (Fig. S5A).

Approximately 45% of the global cropland area in 2000 showed medium-high or high PUE, but there was considerable variation in the drivers of these higher PUE values. Locations with P deficits were overwhelmingly associated with higher PUE (61% of areas with medium-high or high PUE had P deficits), indicating that overall crop production in these areas was more reliant on soil P drawdown than on agronomic P inputs to meet crop requirements. This situation was particularly extensive in Eastern Europe, southern South America, as well as West and Central Africa. The remaining fraction (39%) of the global cropland area with medium-high or high PUE was associated with P surpluses, indicating high crop production returns per unit of P input without dependence on soil P drawdown. Large tracts of South and Southeast Asia as well as parts of the central United States and Western Europe showed these higher PUE values in areas with P surpluses and relatively high overall crop production (Fig. S5A). Widespread parts of Africa and isolated areas in other regions with low overall crop production also had relatively high PUE, typically with small surpluses related to low P inputs. Approximately 13% of cropland with higher PUE also had high (top quartile) manure applications (e.g., in parts of the United States), whereas less than 6% had high P fertilizer applications (e.g., parts of Germany) (Fig. S2).

Medium-low and low PUE (55% of the global cropland area) were associated most with locations of high fertilizer use and larger P surpluses, where excess P fertilizer use likely provided little additional benefits for improving crop productivity. Approximately 46% of the cropland area with lower PUE worldwide corresponded to areas with high fertilizer P applications, which is almost double the proportion of cropland area with lower PUE that had high manure P applications (25%) (Fig. S2). This is particularly evident in China, the Midwestern United States, and Southern Europe. Other areas with lower PUE often had relatively low P fertilizer applications and relatively low overall crop production (Fig. 5 and Fig. S5A), such as East Africa and northern Brazil, indicating potential constraints on crop productivity other than agronomic P inputs, including fixation of applied fertilizer P to less plant-available forms in soils, deficiencies of other soil properties (Sanchez *et al.* 2003), or lack of adequate irrigation. Relatively low agronomic P inputs may have contributed minimally to augmenting crop production in these areas compared to other areas with low crop production.

2.3. Discussion

2.3.1. Implications for regional phosphorus management

Our study provides a consistent global account of the spatial pattern of agronomic P surpluses and deficits, highlighting areas of potential soil P accumulation that can increase P loading to aquatic ecosystems (Carpenter *et al.* 1998; Kleinman *et al.* 2000), as well as areas of P deficits that could impose constraints on crop productivity and food security (Vlek *et al.* 1997; Garcia 2001; Csathó & Radimszky 2009). Large disparities between agronomic P inputs and outputs pose major challenges for long-term management of water quality and agricultural productivity at all scales. However, regional agronomic P imbalances are difficult to address because they represent the aggregate effects of many complex factors, including nutrient management decisions by individual farmers, socioeconomic conditions, government policies, and environmental setting (Smil 2000; Lesschen *et al.* 2007; Cobo *et al.* 2010).

Improving global P-fertilizer use efficiency will be pivotal to making agricultural P use more sustainable. Global consumption of P fertilizers increased in the past decade by >10% over 2000 levels (based on an average of 2007-2008 estimates) (FAO 2010). Farmers may apply P fertilizer in excess of crop requirements to build soil P concentrations, which may be important in P-deficient soils, yet there are typically diminishing returns of additional P fertilizer application for crop yields above a critical level at which plant-available P is maximized (Sharpley & Withers 1994; Kleinman *et al.* 2000). On average, developing countries had P deficits during the mid-20th century (Bennett *et al.* 2001), but our results suggest that current P fertilizer use may be contributing to soil P accumulation and relatively low PUE in some rapidly developing areas. Although there is considerable uncertainty in the rate of depletion for current economically-extractable phosphate rock reserves, increases in the cost of P fertilizer in the coming decades is likely (Cordell *et al.* 2009; Van Vuuren *et al.* 2010). Potential constraints on future access to P fertilizers could be better reflected in current P fertilizer use, particularly in regions with low PUE.

Redistribution of P fertilizers from certain areas with more intense P surpluses to P-deficit croplands could be particularly effective at resolving global P imbalances. We estimate that a 21% reduction in P fertilizer use in all locations with top quartile surpluses (>13 kg P ha^{-1} yr^{-1}) associated with P fertilizer inputs (Fig. 4B) in 2000 could have been achieved without causing any of these areas to transition to P deficits. This hypothetical targeted reduction in P fertilizer use would provide a net fertilizer savings of 1.2 Tg P yr^{-1} globally (reducing the global P surplus by 10%), while also increasing average PUE across these locations by 14%. If this fertilizer P were instead redistributed across all P-deficit cropland, it would effectively meet the total crop P requirements in these locations, eliminating all P deficits globally. These findings highlight the inherent interconnectedness of solutions to both P surpluses and deficits at the global scale.

Opportunities to more effectively capture and recycle manure in mixed livestock-cropping systems could also help move some sub-regional P deficits closer to net zero P balances, particularly in forage croplands with extensive P deficits (Menzi *et al.* 2010). Our results show that manure P was typically associated with P surpluses in areas with high livestock densities but insufficient cropland to effectively assimilate the manure P produced by these animals (Kellogg *et al.* 2000; Naylor *et al.* 2005). In some regions with low manure recoverability (e.g., South Asia and Africa) (Sheldrick *et al.* 2003), there may be a large magnitude of potentially underutilized manure P currently lost from agricultural systems that could serve as a useful organic fertilizer source to reduce reliance on inorganic P fertilizers. Achieving more effective manure P recycling at the global scale may require broader management or structural changes in livestock farming, such as improved access to manure collection and treatment technologies, changes in livestock diets, or even reductions in livestock densities (Menzi *et al.* 2010).

Areas with relatively balanced P situations (first quartile surplus or deficit), and coincident medium-high or high PUE and crop production represent a model of optimal P management that could provide insight on how to resolve P imbalances in other areas. Locations meeting these criteria were scattered throughout every region, across a wide range of development statuses and types of agricultural systems (Fig. S5B). Examples of larger contiguous areas with optimal P management were in Southeast Asia, Central and Eastern Europe, the central United States, and the Caribbean (Fig. 5). One pattern common to most of these locations (85%) is that neither fertilizer nor manure P applications alone exceeded crop P use (Fig. 4A); also, less than 10% of these areas had top quartile P fertilizer applications, and less than 20% had top quartile manure P applications (Fig. S2). This indicates that practices such as integrated nutrient management, which more effectively recycle manure nutrients from livestock and reduce fertilizer use accordingly (Shigaki *et al.* 2006; Ju *et al.* 2009), are important for mitigating P surpluses in areas with high livestock densities and high fertilizer application rates.

2.3.2. Evaluation of results and limitations

Our global agronomic P balance estimates for 2000 are broadly comparable to those calculated by other studies using a similar agronomic balance approach (Table S1). Our estimate of global P fertilizer application compares particularly well to reported values from other studies for this time period. Smil's (2000) estimate of manure P applied to croplands for the mid-1990s (6-8 Tg P yr^{-1}) is lower than our recoverable manure P estimate of 9.6 Tg P yr^{-1} due to the higher total manure P production used as a baseline in our study (Potter *et al.* 2010). We also estimated greater crop P removal (12.3 Tg P yr^{-1}) than Smil (2000) (8-9 Tg P yr^{-1}) given our inclusion of several additional crops (Monfreda *et al.* 2008). Bouwman et al. (2009) estimated P balances separately for grasslands and all other croplands in 2000; however, they grouped some cultivated grasses, such as hay crops, with their estimate for non-cultivated pasture lands, making comparison somewhat challenging because our study addresses only cultivated lands. They estimated a total agronomic P surplus of 15 Tg P yr^{-1} for agricultural soils globally (11 and 4 Tg P yr^{-1} in croplands and grassland soils, respectively) when omitting losses to hydrological systems of 2 Tg P yr^{-1} (Bouwman *et al.* 2009). This is within the range of our estimate of 11.5 Tg P yr^{-1} given that we included inputs and outputs associated with cultivated grasses and several additional crops, but not with pastures, therefore our crop P removal estimates are higher and our manure P estimates are lower. Our overall agronomic P balance results are close to Smil's (2000) lower estimate of 12 Tg P yr^{-1}, whereas we estimate a higher agronomic P balance than Sheldrick et al. (2002) due to their lower manure and fertilizer estimates (Table S1).

The spatial patterns in our P balance results are also generally consistent with spatially-explicit P balances for Asia and Europe by Gerber et al. (Gerber *et al.* 2002; Gerber *et al.* 2005), for China by Shen et al. (Shen *et al.* 2005), and for India by Pathak et al. (Pathak *et al.* 2010). Gerber et al. (2005) found similar large P surpluses driven by fertilizer use in eastern China and northern India, as well as P deficits or small P surpluses in Burma, parts of Malaysia, Indonesia, and northern China. However, they found P deficits throughout southern India and in pockets of southern China and Vietnam where we found mostly larger P surpluses, which is likely attributable to our use of a more simplified method to calculate manure P applications to ensure global consistency. Our P balance results also generally agree with Gerber et al. (2002) for sub-national jurisdictions in Western Europe, showing larger P surpluses driven primarily by manure P inputs in the Netherlands, Belgium, northern Italy, western Germany, Ireland, and Finland, although we found slightly higher P surpluses for parts of France and Italy. Pathak et al. (2010) found P surpluses ranging from small to large across different states in southern and northern India that are more consistent with our results for that region, except that they found P deficits in the north-eastern state of Assam and the central state of Madhya Pradesh. Shen et al. (2005) calculated P surpluses >10 kg P ha^{-1} yr^{-1} throughout most provinces in China that correspond to our top quartile P surpluses calculated for that region (>13 kg P ha^{-1} yr^{-1}), particularly in eastern China; however, they generally found lower P surpluses in southern China. We also compared our results to those of Bouwman et al. (2009) based on slight modifications of their P balance calculations to provide greater comparability with our study (see SI text for a detailed description; Fig. S6). Our P balance results show greater spatial variation than Bouwman et al. (2009) due to our use of data derived from additional sub-national sources for inputs and outputs (Wint 2007; Monfreda *et al.* 2008). Other differences are most likely related to our more detailed calculation of P removal for 123 individual crops and their inclusion of manure P produced by pasture-grazed animals not included in our calculations for croplands.

Our study is based on a soil-surface balance approach (Oenema *et al.* 1999; Sheldrick *et al.* 2002) that considers direct agronomic P inputs and outputs but not potential losses from cropland soils. Estimates of total P losses due to agricultural soil erosion range from 12.5 to 22.5 Tg P yr^{-1}, with particularly large P erosion losses in Africa and Southeast Asia (Smil 2000;

Quinton *et al.* 2010). Bouwman et al. (2009) used a more conservative estimate of agricultural P loss via runoff and leaching based on 10% of total P inputs (roughly 2.4 Tg P yr^{-1} using our estimates). Accounting for P losses from soils to water in areas with small P surpluses, such as sub-Saharan Africa, would possibly lead to results of small deficits in many locations, reflecting the small P deficits typically found in studies that considered P losses in that region (Sheldrick & Lingard 2004; Lesschen *et al.* 2007; Cobo *et al.* 2010). Occlusion of P in soils to less plant-available forms may also limit the effectiveness of surplus agronomic P to supplement crop growth, especially in highly weathered and P-limited tropical soils, such as those in parts of Brazil and East Africa (Sanchez *et al.* 2003), which may partially explain lower PUE in these areas. Additional limitations related to data constraints are described in the SI text.

2.3.3. Moving forward

Although P deficits and P surpluses may appear to be geographically separate and essentially opposite problems, our analysis indicates that global solutions to both types of P imbalances should be approached in tandem. Closing the gaps between areas with the most intense P surpluses and deficits may be achievable with more efficient P fertilizer use, which would help redistribute P fertilizers to P-deficit cropland, and more effective recycling of manure P that would promote tighter P cycling in agricultural landscapes and ease reliance on inorganic P sources.

Global food production may need to increase by up to 70% over year 2000 levels to meet demands from population growth by 2050 (Bruinsma 2009). Much of this increase will likely need to come from increasing crop yields in developing countries (Bruinsma 2009). Agricultural P management is central to maximizing agricultural productivity while simultaneously reducing threats to water quality due to P loading from agricultural lands, as well as accounting for uncertainties in future access to inorganic P fertilizers. Although improvements in P-use efficiency with more integrated management of fertilizer and manure will be helpful, long-term solutions to regional P surpluses and deficits may require transformations in underlying agricultural policies and management practices (Vitousek *et al.* 2009; Menzi *et al.* 2010), as well as better recycling of P from human and agricultural waste streams (Cordell *et al.* 2009; Van Vuuren *et al.* 2010). The challenge of supplying sufficient P to meet agricultural demands worldwide without degrading freshwater resources will be a key issue for agriculture in the 21st century.

2.4. METHODS

2.4.1. Agronomic P inputs and outputs

We estimated P fertilizer applications to croplands in 2000 based on a slight modification of methods from Potter et al. (2010). For 88 countries with crop-specific average national P fertilizer rates from the International Fertilizer Industry Association (IFA) (IFA/IFDC/IPI/PPI/FAO 2002), the spatial patterns of 104 individual crop maps and 30 grouped crop maps (Monfreda *et al.* 2008) were used to estimate the distribution of P fertilizer applications (Potter *et al.* 2010). For 73 countries without crop-specific data, we distributed total national P fertilizer consumption estimates for the year 2000 (or 2002 in some cases) from the Food and Agriculture Organization of the United Nations (FAO) based on the spatial patterns of a generic cropland map (Ramankutty *et al.* 2008). We also added sub-national P fertilizer estimates for four major P-fertilizer consuming countries each representing >5% of the total global P fertilizer consumption circa 2002 (China, United States, India, and Brazil; (FAO 2010)), as well as adjustments for P fertilizer applications to cultivated grasslands, and national or crop-specific P fertilizer application updates for certain countries (described in the SI text).

Total manure P excreted by animals in each grid cell was estimated using the method of Potter et al. (2010) based on global livestock distribution maps (Wint 2007) for major livestock species (cows, pigs, poultry, sheep, goats, and buffalo). The number of animals in each grid cell was multiplied by species-specific manure P production coefficients, which were scaled based on regional livestock weights to account for inter-regional differences (Potter *et al.* 2010). The fraction of total manure P produced by animals that is available for cropland application may vary based on factors such as the degree of livestock confinement or pasture grazing, transportation costs, and agricultural technology (Sheldrick *et al.* 2003). We estimated manure P used for cropland application in each grid cell based on regional estimates of manure recoverability from Sheldrick et al. (2003) for 12 regions. For the United States, we used recoverable manure P fractions for cattle, pigs, and poultry from Kellogg et al. (2000) for each state based on comprehensive surveys of livestock confinement and manure collection. The approximate fraction of manure used as fuel was also excluded from the total recoverable manure P based on regional estimates for cattle, buffaloes, and chickens for West, South, and Southeast Asia (assuming that buffaloes were representative of dairy cattle) (Mosier *et al.* 1998).

Total P removed from cropland soils by harvested crops circa 2000 was estimated using spatially-explicit crop production maps for 123 individual crops (Monfreda *et al.* 2008) and P harvest removal fractions for each of these crops. The Monfreda et al. (2008) crop maps are based largely on subnational crop production statistics (or national in some countries without sub-national data), which is the most comprehensive spatial source showing inventories for individual crops globally that we are aware of. We calculated P removal based on this spatial crop production data for each cropland grid cell by multiplying the total production (in kg) for each individual crop by the dry matter (DM) content and P content of that crop (for all crops and all grid cells, *Total P removal (kg) = Crop yield (kg) x (%DM/100) x (%P/100)*). We used crop-specific P content and dry matter content of grains or harvested portions for 80 crops from USDA-NRCS (2009), and averages based on crop groups for 14 crops when crop-specific P contents were unavailable. For remaining crops where P data was not available from this source, we used P contents from IFA (1992) and Lechessen et al. (2007) and dry matter contents from Monfreda et al. (2008). Our method for estimating crop residue production, as well as the contrasting high and low residue P recycling and removal scenarios based on Smil (1999) are detailed in the SI text.

2.4.2. P balance and P-use efficiency calculations

The P balance for each cropland grid cell was calculated as the difference between total inputs and outputs, divided by the total cropland area in that cell. Total cropland areas from Ramankutty et al. (2008) were used as the basis for cropland areas; however, in addition to cultivated areas, this map includes substantial amounts of temporary pasture and fallow in some regions. To test the sensitivity of our results to this, we also calculated P balances based on total harvested areas summed for all 123 individual crops from Monfreda et al. (2008) (Fig. S4). This and other alternative P balance calculations related to known uncertainties in our analysis are described in the SI text; differences in the spatial patterns of P surpluses and deficits based on these alternative calculations were typically minimal, but we found larger surpluses and deficits in most regions when using total harvested area for all crops as the denominator. PUE was calculated as total crop dry-matter crop production divided by total P fertilizer and manure application in each grid cell (Bouwman *et al.* 2009). The calculations of quartile ranges for the P input, output, P balance, and PUE maps excluded grid cells with less than 5% cropland area in order to avoid marginal agricultural areas that typically showed extreme values for each term.

2.5. ACKNOWLEDGEMENTS

We thank Meagan Schipanski for many helpful discussions and Simon Donner for his contribution to the development of the P fertilizer dataset by Potter et al. (Potter *et al.* 2010) and suggestions for this work. Funding was provided by the Natural Sciences and Engineering Research Council of Canada (NSERC).

2.6. References

Bennett, E. M., Carpenter, S. R. & Caraco, N. F. (2001) Human impact on erodable phosphorus and eutrophication: A global perspective. *Bioscience,* **51,** 227-234.

Bouwman, A. F., Beusen, A. H. W. & Billen, G. (2009) Human alteration of the global nitrogen and phosphorus soil balances for the period 1970-2050. *Global Biogeochemical Cycles,* **23**.

Bruinsma, J. (2009) The resource outlook to 2050: By how much do land, water and crop yields need to increase by 2050? Food and Agriculture Organization of the United Nations, Rome.

Carpenter, S. R. (2008) Phosphorus control is critical to mitigating eutrophication. *Proceedings of the National Academy of Sciences of the United States of America,* **105,** 11039-11040.

Carpenter, S. R., Caraco, N. F., Correll, D. L., Howarth, R. W., Sharpley, A. N. & Smith, V. H. (1998) Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecological Applications,* **8,** 559-568.

Cobo, J. G., Dercon, G. & Cadisch, G. (2010) Nutrient balances in african land use systems across different spatial scales: A review of approaches, challenges and progress. *Agriculture, Ecosystems and Environment,* **136,** 1-15.

Cordell, D., Drangert, J. O. & White, S. (2009) The story of phosphorus: Global food security and food for thought. *Global Environmental Change,* **19,** 292-305.

Csathó, P. & Radimszky, L. (2009) Two worlds within eu27: Sharp contrasts in organic and mineral nitrogen-phosphorus use, nitrogen-phosphorus balances, and soil phosphorus status: Widening and deepening gap between western and central europe. *Communications in Soil Science and Plant Analysis,* **40,** 999-1019.

Dobermann, A. & Cassman, K. G. (2002) Plant nutrient management for enhanced productivity in intensive grain production systems of the united states and asia. *Plant and Soil,* **247,** 153-175.

Dobermann, A. & Cassman, K. G. (2005) Cereal area and nitrogen use efficiency are drivers of future nitrogen fertilizer consumption. *Science in China. Series C, Life sciences / Chinese Academy of Sciences.,* **48 Spec No,** 745-758.

FAO (2010) Faostat: Fao statistical databases. Food and Agriculture Organization of the United Nations, Rome.

Garcia, F. (2001) Phosphorus balance in the argentinean pampas. *Better Crops International,* **15,** 22-24.

Gerber, P., Chilonda, P., Franceschini, G. & Menzi, H. (2005) Geographical determinants and environmental implications of livestock production intensification in asia. *Bioresource Technology,* **96,** 263-276.

Gerber, P., Franceschini, G. & Menzi, H. (2002) Livestock density and nutrient balances across europe. FAO Livestock, Environment and Development Inititative (LEAD), Rome.

IFA (1992) *World fertilizer use manual*. International Fertilizer Industry Association, Paris.

IFA/IFDC/IPI/PPI/FAO (2002) Fertilizer use by crops. *(International Fertilizer Industry Association, Rome),* **5th Ed.**

Ju, X. T., Xing, G. X., Chen, X. P., Zhang, S. L., Zhang, L. J., Liu, X. J., Cui, Z. L., Yin, B., Christie, P., Zhu, Z. L. & Zhang, F. S. (2009) Reducing environmental risk by improving n management in intensive chinese agricultural systems. *Proceedings of the National Academy of Sciences of the United States of America,* **106,** 3041-3046.

Kellogg, R. L., Lander, C. H., Moffitt, D. C. & Gollehon, N. (2000) Manure nutrients relative to the capacity of cropland and pastureland to assimilate nutrients: Spatial and temporal trends for the united states. (Natural Resources Conservation Service/Economic Research Service/US Department of Agriculture, Washington, DC).

Kleinman, P. J. A., Bryant, R. B., Reid, W. S., Sharpley, A. N. & Pimentel, D. (2000) Using soil phosphorus behavior to identify environmental thresholds. *Soil Science,* **165,** 943-950.

Lesschen, J. P., Stoorvogel, J. J., Smaling, E. M. A., Heuvelink, G. B. M. & Veldkamp, A. (2007) A spatially explicit methodology to quantify soil nutrient balances and their uncertainties at the national level. *Nutrient Cycling in Agroecosystems,* **78,** 111-131.

Liu, J., You, L., Amini, M., Obersteiner, M., Herrero, M., Zehnder, A. J. B. & Yang, H. (2010) A high-resolution assessment on global nitrogen flows in cropland. *Proceedings of the National Academy of Sciences of the United States of America,* **107,** 8035-8040.

Liu, Y., Villalba, G., Ayres, R. U. & Schroder, H. (2008) Global phosphorus flows and environmental impacts from a consumption perspective. *Journal of Industrial Ecology,* **12,** 229-247.

Menzi, H., Oenema, O., Burton, C., Shipin, O., Gerber, P., Robinson, T. & Franceschini, G. (2010) Impacts of intensive livestock production and manure management on the environment. (eds. H. Steinfeld, H. Mooney, F. Schneider & L. Neville), pp. 416. Island Press, Washington, DC.

Monfreda, C., Ramankutty, N. & Foley, J. A. (2008) Farming the planet: 2. Geographic distribution of crop areas, yields, physiological types, and net primary production in the year 2000. *Global Biogeochemical Cycles,* **22**.

Mosier, A., Kroeze, C., Nevison, C., Oenema, O., Seitzinger, S. & Van Cleemput, O. (1998) Closing the global N_20 budget: Nitrous oxide emissions through the agricultural nitrogen cycle: OECD/IPCC/IEA Phase II development of IPCC guidelines for national greenhouse gas inventory methodology. *Nutrient Cycling in Agroecosystems,* **52,** 225-248.

Naylor, R., Steinfeld, H., Falcon, W., Galloway, J., Smil, V., Bradford, E., Alder, J. & Mooney, H. (2005) Losing the links between livestock and land. *Science,* **310,** 1621-1622.

Oenema, O., Heinen, M., Smaling, E. M. & Fresco, L. O. (1999) Nutrient disequilibria in agroecosystems. *Uncertainties in Nutrient Budgets Due to Biases and Errors: Concepts and Case Studies***,** 75-97.

Pathak, H., Mohanty, S., Jain, N. & Bhatia, A. (2010) Nitrogen, phosphorus, and potassium budgets in indian agriculture. *Nutrient Cycling in Agroecosystems,* **86,** 287-299.

Potter, P., Ramankutty, N., Bennett, E. M. & Donner, S. D. (2010) Characterizing the spatial patterns of global fertilizer application and manure production. *Earth Interactions,* **14,** 1-22.

Quinton, J. N., Govers, G., Van Oost, K. & Bardgett, R. D. (2010) The impact of agricultural soil erosion on biogeochemical cycling. *Nature Geosci,* **3,** 311-314.

Rabalais, N. N., Díaz, R. J., Levin, L. A., Turner, R. E., Gilbert, D., and Zhang, J. (2010) Dynamics and distribution of natural and human-caused hypoxia. *Biogeosciences,* **7,** 585-619.

Ramankutty, N., Evan, A. T., Monfreda, C. & Foley, J. A. (2008) Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000. *Global Biogeochemical Cycles,* **22**.

Roy, R. N., Mirsa, R. V., Lesschen, J. P. & Smaling, E. M. (2003) *Assessment of soil nutrient balance: Approaches and methodologies*. FAO, Rome.

Sanchez, P. A., Palm, C. A. & Buol, S. W. (2003) Fertility capability soil classification: A tool to help assess soil quality in the tropics. *Geoderma,* **114,** 157-185.
Sharpley, A. N. & Withers, P. J. A. (1994) The environmentally-sound management of agricultural phosphorus. *Fertilizer Research,* **39,** 133-146.
Sheldrick, W., Keith Syers, J. & Lingard, J. (2003) Contribution of livestock excreta to nutrient balances. *Nutrient Cycling in Agroecosystems,* **66,** 119-131.
Sheldrick, W. F. & Lingard, J. (2004) The use of nutrient audits to determine nutrient balances in africa. *Food Policy,* **29,** 61-98.
Sheldrick, W. F., Syers, J. K. & Lingard, J. (2002) A conceptual model for conducting nutrient audits at national, regional, and global scales. *Nutrient Cycling in Agroecosystems,* **62,** 61-72.
Shen, R. P., Sun, B. & Zhao, Q. G. (2005) Spatial and temporal variability of n, p and k balances for agroecosystems in china. *Pedosphere,* **15,** 347-355.
Shigaki, F., Sharpley, A. & Prochnow, L. I. (2006) Animal-based agriculture, phosphorus management and water quality in brazil: Options for the future. *Scientia Agricola,* **63,** 194-209.
Smil, V. (1999) Nitrogen in crop production: An account of global flows. *Global Biogeochemical Cycles,* **13,** 647-662.
Smil, V. (2000) Phosphorus in the environment: Natural flows and human interferences. *Annual Review of Energy and the Environment,* **25,** 53-88.
Tilman, D., Cassman, K. G., Matson, P. A., Naylor, R. & Polasky, S. (2002) Agricultural sustainability and intensive production practices. *Nature,* **418,** 671-677.
USDA-NRCS (2009) Crop nutrient tool: Nutrient content of crops. United States Department of Agriculture, Natural Resource Conservation Service, Washington.
Van Vuuren, D. P., Bouwman, A. F. & Beusen, A. H. W. (2010) Phosphorus demand for the 1970-2100 period: A scenario analysis of resource depletion. *Global Environmental Change,* **20,** 428-439.
Vitousek, P. M., Naylor, R., Crews, T., David, M. B., Drinkwater, L. E., Holland, E., Johnes, P. J., Katzenberger, J., Martinelli, L. A., Matson, P. A., Nziguheba, G., Ojima, D., Palm, C. A., Robertson, G. P., Sanchez, P. A., Townsend, A. R. & Zhang, F. S. (2009) Nutrient imbalances in agricultural development. *Science,* **324,** 1519-1520.
Vlek, P. L. G., KuÌˆhne, R. F. & Denich, M. (1997) Nutrient resources for crop production in the tropics. *Philosophical Transactions of the Royal Society B: Biological Sciences,* **352,** 975-985.
Wint, G. R., and T. P. Robinson (2007) Gridded livestock of the world - 2007. *(Food and Agriculture Organization of the United Nations/Rome, Italy).*

2.7. FIGURES

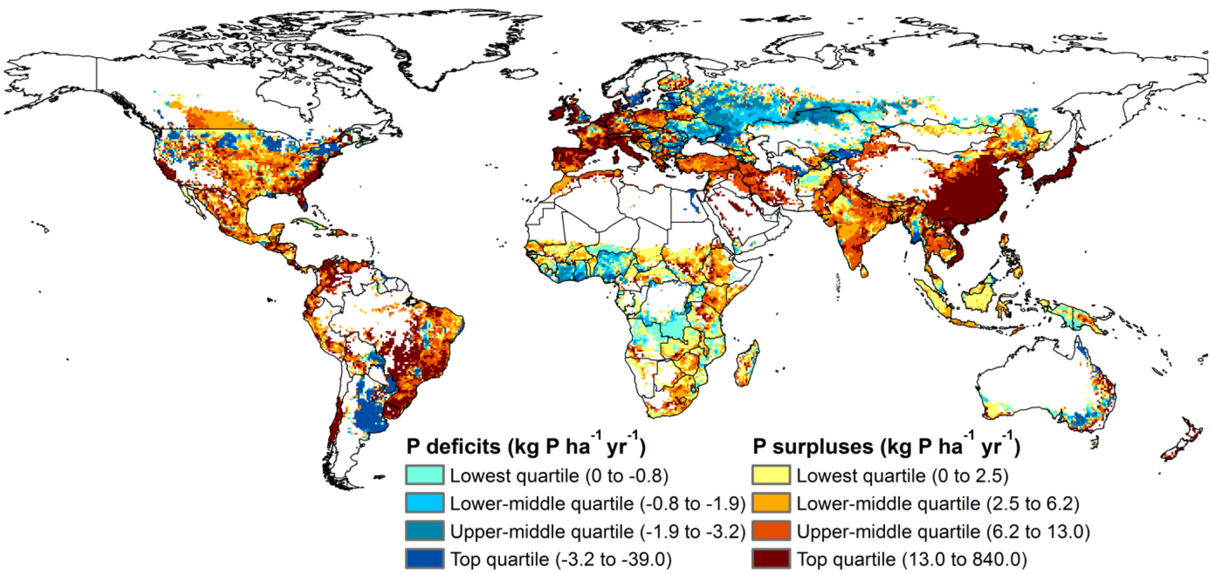


Fig. 1. Global map of agronomic P imbalances for the year 2000 expressed per unit of cropland area in each 0.5 degree grid cell. The P surpluses and deficits are each classified according to quartiles globally (0-25th, 25-50th, 50-75th, and 75-100th percentiles)

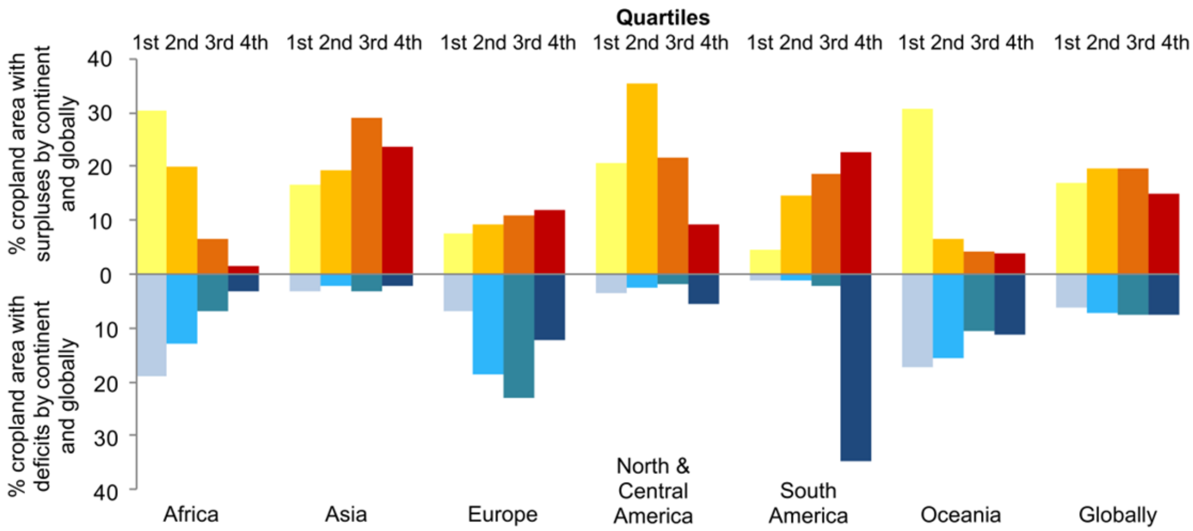


Fig. 2. Distributions of P surpluses and deficits by quartiles shown as percent of total cropland area in each continent and as percent of global cropland area.

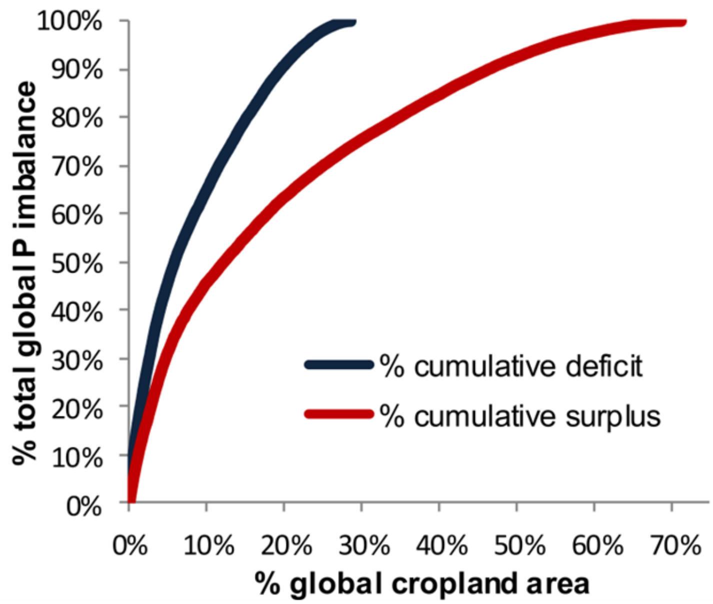


Fig. 3. Cumulative distributions of global cropland P imbalances (surpluses or deficits, sorted from largest to smallest) in relation to cumulative global cropland area.

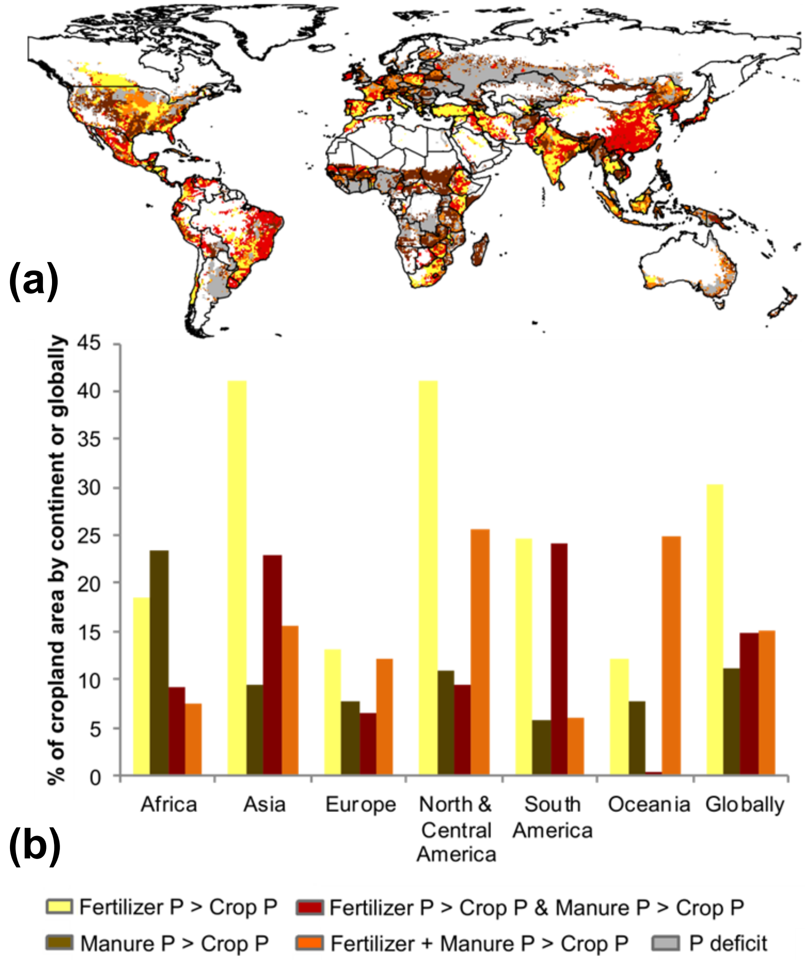


Fig. 4. Agronomic drivers of P surpluses based on the magnitude of fertilizer or manure P applied relative to crop P use in different locations (*a*) and summarized according to percent of cropland area by continent and globally (*b*). Each category is mutually exclusive based on locations where either fertilizer alone or manure alone exceeded crop P use, where fertilizer and manure each individually exceeded crop P use, or where only the sum of fertilizer and manure exceeded crop P use.

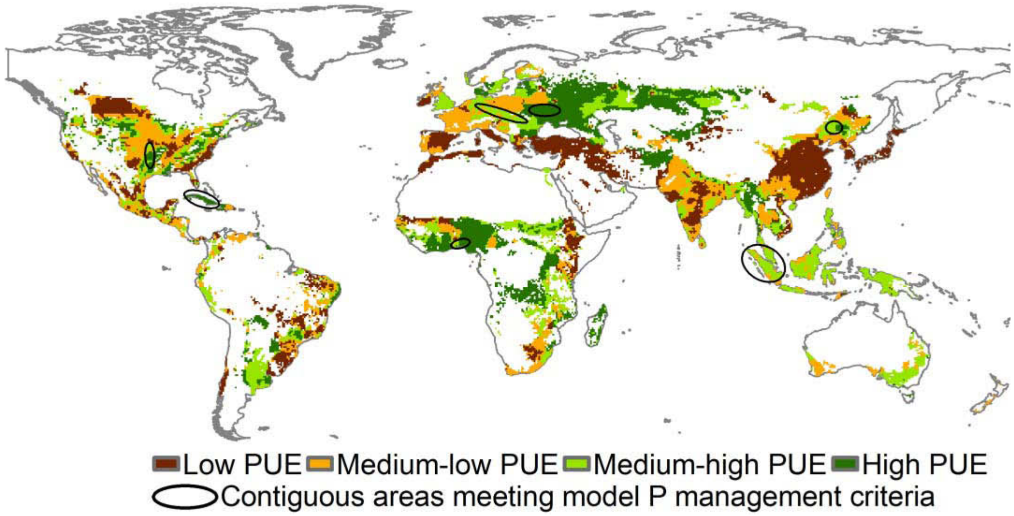


Fig. 5. Map of total agronomic P-use efficiency (kg crop · kg P input^{-1}; PUE) for 123 crops in 2000 classified from low to high based on quartiles globally. Ovals indicate examples of contiguous areas with model P management (relatively balanced P situations, with PUE and crop production each above the global median; based on Fig. S5B).

2.8. SUPPORTING INFORMATION TEXT

Calculation of P fertilizer applications

We followed the basic method of Potter et al. (2010) with some minor adjustments to estimate P fertilizer application to croplands. Potter et al. (2010) used the spatial patterns of 104 individual crop maps and 30 grouped crops from Monfreda et al. (2008) to spatially disaggregate national crop-specific P fertilizer application rates for 88 countries based on harvested or fertilized areas from the International Fertilizer Industry Association (IFA) (IFA/IFDC/IPI/PPI/FAO 2002). Several countries reported fertilizer applications to pastures, cultivated grasslands (e.g., hay crops), and other forage crops, but sometimes grouped all three uses into one category (IFA/IFDC/IPI/PPI/FAO 2002). Our study is representative of only croplands, including cultivated grasslands and other forage crops, but excluding uncultivated pastures. Accordingly, we omitted P fertilizer inputs to pastures in countries that listed P fertilizer rates separately for this use. In twelve countries that grouped P fertilizer applications for pasture and other forage crops together into a single category, we estimated the fraction that was applied to just forage crops based on the ratio of total national pasture area to total forage crop area in each country (Monfreda *et al.* 2008; Ramankutty *et al.* 2008). We assumed that the P fertilizer category 'other' crops (IFA/IFDC/IPI/PPI/FAO 2002) in New Zealand was for pastures and thus excluded all P inputs for this category. We also used the cropland map from Ramankutty et al. (Ramankutty *et al.* 2008) to distribute P fertilizer application for the 'other' crops category for India (IFA/IFDC/IPI/PPI/FAO 2002). Corrections for the crop-specific P fertilizer application rates in Argentina and Turkey were made to account for inconsistencies in the percentage of harvested areas that were fertilized, as compared to other countries reported by IFA (2002). Finally, we added an update for the P fertilizer rate for wheat in Bulgaria based on data from IFA (2002).

For 70 countries without crop-specific data, we used total national P fertilizer consumption estimates for the year 2000 from the Food and Agriculture Organization of the United Nations (FAO) and the spatial pattern of the cropland map by Ramankutty et al. (2008) to estimate the sub-national distribution of P fertilizer applications (Potter *et al.* 2010). We also added 2002 FAO national P fertilizer consumption estimates for Angola, Afghanistan, and Suriname that were not reported by the FAO for the year 2000 (FAO 2010); no fertilizer data was available for Yugoslavia in either year, so P fertilizer application was set to zero for that country.

We used 2000 FAO national P fertilizer consumption estimates for Moldova instead of the IFA (2002) crop-specific values because of large differences in the total P fertilizer consumption estimates between the two sources (43 million tons of P fertilizer reported by FAO compared to 0.1 million tons P from IFA).

For all countries that individually contributed 5% or more to the total global P fertilizer consumption in 2002 (China, India, Brazil, and the United States) (FAO 2010), we used additional sub-national P fertilizer data to estimate fertilizer applications. Crop-specific, sub-national P fertilizer application data was only directly available for the United States (USDA-ERS 2010), but this data was limited to four crops and only certain states (compared to the national P fertilizer rates listed for approximately 50 crops in the IFA fertilizer database for that country (IFA/IFDC/IPI/PPI/FAO 2002)). Given these data limitations, and to ensure consistency across countries, we used sub-national total P fertilizer rates aggregated for all crops to scale the IFA crop-specific national rates in these countries. Sub-national total P fertilizer application rates (kg ha^{-1} yr^{-1}) aggregated for all crops were reported directly for India (DAC 2003) and Brazil (FAO 2004). Only the total P fertilizer use (tonnes yr^{-1}) for all crops was consistently reported sub-nationally for China and the United states, so we had to calculate P fertilizer rates using the most appropriate data available for the cultivated or fertilized land areas. For the United States, we divided total P fertilizer use for each state (AAPFCO 2003) by the total fertilized area reported by the U.S. Census of Agriculture (USDA-NASS 2008), thus accounting for substantial unfertilized cropland areas in some states. For China, we used 1997 total provincial P fertilizer use in conjunction with final revised 1996 total cultivated area estimates (NBS 2006) in order to estimate sub-national P fertilizer rates aggregated for all crops matching the time-frame of the IFA (2002) data. Next, we calculated aggregate national P fertilizer application rates in each country based on the crop-specific IFA (2002) data by taking weighted-averages of P fertilizer application rates for all crops, with total harvested area of each crop as the weighting factor. We then scaled the IFA (2002) crop-specific fertilizer rates sub-nationally in each country using the quotients resulting from dividing each sub-national aggregate P fertilizer rate by the IFA national weighted-average rate. This scaling method was chosen iteratively to ensure that total national P fertilizer applications were as close as possible to the values reported by IFA (2002).

Calculation of manure P applications

Total manure P production for all major livestock types was calculated *sensu* Potter et al. (2010) with spatially-explicit global livestock data from the Gridded Livestock of the World (Wint 2007) datasets, and then adjusted for the fraction of manure available for cropland application using a method similar to Liu et al. (2010). The total amount of manure P produced by livestock that is available for cropland application may vary based on factors such as the degree of livestock confinement or pasture grazing, transportation costs, and agricultural technology (Sheldrick *et al.* 2003). Accordingly, we multiplied the total manure P production estimates by recoverable manure P coefficients for 12 world regions from Sheldrick et al. (2003); for cattle, pigs, and poultry in the U.S., we instead used more detailed recoverable manure P estimates for individual states from Kellogg et al. (2000). We also used U.S. averages for cattle, pigs, and poultry (Kellogg *et al.* 2000) to estimate recoverable manure in Canada. Average recoverability factors for cattle, pigs, and poultry from Sheldrick et al. (2003) range from a low of 50% in Africa, Central and South America to a high of 82% in Western Europe. Average recoverability for cattle, pigs, and poultry in the U.S. from Kellogg et al. (2000) was 73%. However, recoverability for animals that are more commonly grazed in some regions (e.g. cattle and ruminants) is typically much lower than these averages for both sources, reflecting the higher recoverability of confined animals (e.g. chickens and pigs).

Alternative P balance map calculations

We calculated a series of alternative P balance maps in order to evaluate the potential effects of known uncertainties on our overall results (Fig. S3 and Fig. S4). In particular, we looked at the potential effects of: crop residue management on the spatial patterns of P surpluses and deficits; possible under-reporting of P fertilizer rates for crops in some countries with limited data; forage crop harvest on the location of P deficits; and incorporation of fallow lands in the cropland area denominator used to calculate the P balances.

Crop residue management scenarios

Crop residues, including straw or stover material not incorporated in the economic harvest of grains and other crops, may contribute substantially to cropland nutrient flows in some regions (Smil 1999; Liu *et al.* 2010); however, the amount of crop reside that remains in fields or is removed for other purposes varies widely by crop type within and between regions and is largely untracked by national or international statistics. We estimated residue production within each grid cell based on residue multipliers from Smil (1983; 1999) that adjust total wet-matter crop production to residue dry-matter production, and then multiplied this by the P content of residues for 9 major crops (USDA-NRCS 2009) and crop groups for all remaining crops (Smil 2000). To account for the potential effects of crop residue management on our results, we recalculated P balances based on two contrasting global scenarios of crop residue management that each account for broad differences in residue management between high-income (developed) and low-income (developing) countries (FAO 2003). In the high residue removal scenario, we assumed that 40% of residues were removed from croplands in developed countries and 60% were removed in developing countries (20), therefore resulting in additional P outputs from cropland soils. The remainder of the residues were considered to be recycled as additional P inputs to cropland soils. In the low residue removal scenario, we assumed that 30% and 40% of residues were removed in developed and developing countries, respectively, and the remainder of residues were recycled to soils (20). Based on these parameters for residue removal and recycling, the additional P input and output terms associated with residues were calculated using the equations described by Liu et al. (2010).

At the global scale, we estimate that approximately 3.9 Tg P yr^{-1} was contained in crop residues associated with crops harvested in 2000. In the high global residue removal scenario, 1.9 Tg P yr^{-1} was recycled as additional P inputs to cropland soils and 2.0 Tg P yr^{-1} was removed from croplands as additional P outputs, reducing the global agronomic P surplus slightly to 11.1 Tg P yr^{-1}. In the low residue removal scenario, 2.5 Tg P yr^{-1} was recycled as inputs and 1.4 Tg P yr^{-1} was removed, increasing the P surplus to 12.5 Tg P yr^{-1} (~9% greater).The inclusion of these additional P inputs and outputs for crop residues had very minimal influence on our spatial P balance results (Fig. S3A and S3B), with very slight additions of P deficits in small areas of some countries in the high residue removal scenario (e.g., India) and typically larger P surpluses in both scenarios for some developed countries due to the additional P inputs associated with residue recycling (e.g., the Midwestern United States). Actual residue removal may be substantially higher in some developing countries than our global estimates, for example residue removal averages as much as 80% for major crops in India (Ravindranath *et al.* 2005), but much of this removal is associated with residue burning in fields, up to 75% of which can be returned to soils during burning (23).

Fertilizer reporting and forage crop assessments

Although the IFA fertilizer database (IFA/IFDC/IPI/PPI/FAO 2002) provides detailed breakdowns for P fertilizer applications to crops in many countries, the detailed sub-national crop production data from Monfreda et al. (2008) contains several additional crops not reported as fertilized in some countries by IFA (2002). In these cases, we assumed that there were zero P fertilizer applications to these crops; however, it is possible that these crops were fertilized but that data was simply not listed, in which case artificial P deficits could result. To better reflect the P deficits associated with low P fertilizer application rather than potentially missing fertilizer data, we recalculated global P balance maps including only the P removal from crops that received P fertilizer inputs, as well as adjustments for the percentage of national harvested areas of these crops that were fertilized. The resulting P balance map shows broadly consistent spatial patterns of P surpluses and deficits compared to our overall P balance map for all crops in countries with crops-specific P fertilizer data (compare Fig S4A to Fig. 1). This suggests that P fertilizer application data potentially missing from the IFA statistics for certain crops is unlikely to have any substantial influence on our results. One exception is that when limiting the P balance calculations to only crops that are listed in the IFA fertilizer database, some areas where overall P surpluses were more prevalent in our main results (Fig. 1) instead show P deficits (e.g., northern France and eastern Germany; Fig. S4A). The reason for these inconsistent results between the two maps is that we scaled the Monfreda et al. (2008) harvested areas to match the IFA (2002) reported areas for this alternative analysis to provide consistency with the harvested areas used for our P fertilizer estimates (Potter *et al.* 2010). This resulted in greater P removal due to the higher national harvested areas reported by IFA (2002) than Monfreda et al. (2008) for some crops in these countries. These disparities are therefore more likely to reflect uncertainties in the IFA harvested areas for France and Germany rather than uncertainties in our overall P balances for those countries.

Some areas with P deficits in our original results considering all crops show P surpluses in our results using just the crops with P fertilizer rates reported by IFA (2002), which largely correspond to the location of forage crops. We therefore calculated a P balance map excluding P fertilizer applications and crop P removal for all forage crops (in countries where crop-specific fertilizer data was available) in order to confirm that forage crops were acting as important drivers of P deficits in our overall P balance map (compare Fig. S4B and Fig. 1). The resulting map excluding forage crop P inputs and outputs shows many P surpluses that are consistent with those in Fig. S4A (i.e., the northern United States and southern Australia, where forage crops are prevalent) and several additional P surpluses in areas where forage crops are fertilized (e.g., parts of Germany and the Czech Republic), confirming that P removal by forage crops is an important driver of P deficits in our overall P balance results.

Influence of fallow areas on cropland P balances

Our choice of denominator for estimating P surpluses or deficits per unit of cropland area is subject to the uncertainties of the underlying cropland data used in this study (Monfreda *et al.* 2008; Ramankutty *et al.* 2008). In particular, we had a choice between using the total harvested areas for all crops from Monfreda et al. (2008) or the generic cropland area map by Ramankutty et al. (2008). We chose to use the latter map for our overall P balance results in order to more accurately reflect the physical cropland area in different regions and thus the potential rate of P accumulation or depletion in soils. This is particularly important because the Monfreda et al. (2008) harvested areas include multiple cropping of the same land within a single year. However, the Ramankutty et al. (2008) map includes temporary pasture, market and kitchen gardens, and temporarily fallow lands that may over-estimate the actual annual cultivated area in some regions, particularly in tropical South America and Africa where fallowing of croplands is most common. Use of total harvested area (Monfreda *et al.* 2008) as the denominator instead of total cropland area (Ramankutty *et al.* 2008) results in larger P imbalances (either surpluses or deficits) in almost all regions, but particularly in South America and Africa (Fig. S4C), whereas the broad groupings of our P balances by quartile ranges stayed fairly consistent in areas with multiple cropping, such as eastern China. As a result, it is possible that our P surpluses and deficits underestimate the potential P accumulation or depletion in some tropical regions where there are substantial areas of fallow lands.

Comparison with IMAGE global P balance estimates

Bouwman et al. (2009) used the IMAGE 2.4 model to estimate P balances for grasslands (including pastures and some cultivated grasses) and all other croplands in 2000. The model estimates P inputs for fertilizer and manure and P outputs for crops and grasses using spatially-disaggregated source data from national or regional scales for most countries, but sub-national data was used for the U.S. and China (Bouwman *et al.* 2006; Bouwman *et al.* 2009). The inputs are aggregated over three broad cropping systems (wetland rice, legumes, and upland crops), a mixed grassland-cropping system, and three pastoral systems (one for crops and two for grasslands), whereas crop P export is aggregated over all systems for crops and grasses (Bouwman *et al.* 2006; Bouwman *et al.* 2009). Bouwman et al. (2009) presented P balances for 1970, 2000, and model projections for 2030 in kg km^{-2} yr^{-1} of total 0.5 degree grid-cell area, rather than by agricultural area within a grid cell. To allow for better comparability between our study and their results, we obtained the P input and output data generated by the IMAGE model and recalculated P balances using the same quartile ranges used for our P balance map, but excluding the areas of pastoral grassland systems because pastures were not included in our study. Briefly, we multiplied their total P inputs and outputs by the total areas of the three cropping systems and one mixed system described above in order to calculate the grid-cell P balances, which we then divided by the total combined area for these systems to estimate a total P balance per unit of agricultural area. In general, the resulting P balance map (Fig. S6) covers less of the global land area due to the fact that much of the land mass in the IMAGE model is associated primarily with the pastoral grazing system that we excluded for this comparison.

Many of the locations of surpluses and deficits in our study are similar to the model results from Bouwman et al. (2009), but there are some important exceptions that relate to differences in methodology between the studies. We found similar large or moderate surpluses compared to Bouwman et al. (2009) for China, as well as much of Brazil, India and the United States, but our results show more spatial variation in each of these countries, except China, and typically lower P surpluses for India. Although Bouwman et al. (2009) found P surpluses throughout Western Europe, they typically estimated smaller P surpluses in most countries, but our results of extensive P deficits in parts of Eastern Europe (including Russia) and Western Asia are consistent with their findings. Although our results support Bouwman et al.'s (2009) findings of widespread P deficits in grasslands, we calculated large P deficits in several areas with extensive grass and forage crop production (e.g., the northern United States, southern Australia, and southern Argentina) where they calculated P surpluses, which is most likely attributable to their inclusion of manure P deposited on uncultivated pastures by grazing animals that was omitted from our study due to our emphasis on arable croplands. Although our results are broadly or closely comparable to those of Bouwman et al. (2009) for many countries in Africa, they estimated more extensive P deficits throughout parts of Eastern Africa. However, they also found P surpluses throughout Kenya and isolated surpluses surrounded by deficits in some countries (e.g., Ethiopia and Sudan) that more are consistent with our results.

Data limitations and uncertainties

Although we attempted to ensure global-consistency as much as possible in this analysis, limited data availability for P fertilizer applications may affect our results. Crop-specific P fertilizer rates were unavailable for some large countries (e.g., Russia and Ukraine), so we instead distributed total national P fertilizer estimates across the cropland area within these countries following the method of Potter et al. (2010), which may not reflect actual distributions of specific crops and relative fertilizer applications sub-nationally. Additionally, our method for scaling P fertilizer estimates sub-nationally for China, India, Brazil, and the United States assumes that crop-specific P fertilizer applications within a country are directly proportional to sub-national P fertilizer rates aggregated for all crops, although P fertilizer application rates for specific crops may vary in different regions of a country (e.g., USDA-ERS 2010). The denominators used to calculate sub-national P fertilizer application rates also differ for each of the four countries for which we scaled fertilizer rates sub-nationally, i.e., "total cultivated area" was used in China; "total fertilized cropland area" was used in the United States; "gross cropped area" was used in India; and "total planted area" was used in Brazil. Potential inclusion or exclusion of fallow or other unfertilized land areas in these differing estimates of cropland area may result in inaccuracies in sub-national P fertilizer rates, particularly for China and India. Finally, there are occasional disparities between the total national P fertilizer application reported by IFA (2002) and the national P fertilizer applications estimated by taking the summation of each P fertilizer rate multiplied by fertilized area for all crops. Although our method for scaling P fertilizer in these countries estimates national totals closer to those reported by IFA (2002) than Potter et al. (2010), our totals are still lower for India (8%) and Brazil (14%) and higher for China (12%) and the United

States (11%) than IFA (2002). This disparity reflects uncertainty in average P fertilizer rates for individual crops in several countries based on available data. Overall, we feel that the improved spatial pattern of our P fertilizer results sub-nationally for these countries warrants the use of this method despite potential limitations.

There is even less global data available on national and sub-national cropland application of manure. Consistent sub-national manure recoverability data was only available for the United States, so we relied on relatively coarse regional manure recoverability estimates for other countries. Some recoverable manure in developed countries such as the United States may also be applied to permanent pastures (Kellogg *et al.* 2000; Liu *et al.* 2010), but we assumed that all recoverable manure was applied to cultivated lands, given uncertainty about actual pasture-application rates. This may lead to over-estimates of manure P applications in some areas with substantial amounts of permanent pasture relative to cropland area, but we chose to use this approach due to lack of reliable national or sub-national statistics for manure applications to pastures. Finally, we did not exclude potential post-collection losses of recoverable manure P for non-U.S. regions, following Sheldrick et al. (2003) and given data limitations; however, they assume that as much as 10% of recoverable manure could be lost after collection, making it unavailable for cropland application.

Our study draws on existing fertilizer, livestock, and crop harvest data from hundreds to thousands of political jurisdictions worldwide in order to present a spatially-detailed account of global and regional agricultural P imbalances, their drivers, and potential implications. Our findings provide new insight on how agricultural nutrient management is impacting global and regional P flows, but there is some uncertainty in our findings that is largely attributable to lack of detailed sub-national agricultural statistics. The location of crops and livestock are the least uncertain factors in our study given the greater use of sub-national data from the original sources of this data (Wint 2007; Monfreda *et al.* 2008), but there is greater uncertainty regarding the spatial variation in P fertilizer rates and recoverability of manure for cropland application, which may explain some of the differences in our results and those of other studies (e.g., (Gerber *et al.* 2005)). This stresses the importance of collecting more detailed and globally-consistent spatial statistics for fertilizer and manure, which will be essential for improving the accuracy of future global studies for P and other nutrients (Liu *et al.* 2010).

SI REFERENCES

AAPFCO (2003) Commercial fertilizers 2002. Association of American Plant Food Control Officials (AAPFCO), Lexington.

Bouwman, A. F., Beusen, A. H. W. & Billen, G. (2009) Human alteration of the global nitrogen and phosphorus soil balances for the period 1970-2050. *Global Biogeochemical Cycles,* **23**.

Bouwman, A. F., Kram, T. & Goldewijk, K. K. (2006) Integrated modelling of global environmental change. An overview of image 2.4. Netherlands Environmental Assessessment Agency, Bilthoven.

DAC (2003) National income and socio-economic indicators: Inputs. M. o. A. Department of Agriculture & Co-operation, Government of India. New Dehli.

FAO (2003) Compendium of agricultural – environmental indicators 1989-91 to 2000. Statistics Analysis Service, Statistics Division, Food and Agriculture Organization of the United Nations, Rome.

FAO (2004) Fertilizer use by crop in brazil Food and Agriculture Organization of the United Nations, Rome.

FAO (2010) Faostat: Fao statistical databases. Food and Agriculture Organization of the United Nations, Rome.

Gerber, P., Chilonda, P., Franceschini, G. & Menzi, H. (2005) Geographical determinants and environmental implications of livestock production intensification in asia. *Bioresource Technology,* **96,** 263-276.

IFA/IFDC/IPI/PPI/FAO (2002) Fertilizer use by crops. *(International Fertilizer Industry Association, Rome),* **5th Ed.**

Kellogg, R. L., Lander, C. H., Moffitt, D. C. & Gollehon, N. (2000) Manure nutrients relative to the capacity of cropland and pastureland to assimilate nutrients: Spatial and temporal trends for the united states. (Natural Resources Conservation Service/Economic Research Service/US Department of Agriculture, Washington, DC).

Liu, J., You, L., Amini, M., Obersteiner, M., Herrero, M., Zehnder, A. J. B. & Yang, H. (2010) A high-resolution assessment on global nitrogen flows in cropland. *Proceedings of the National Academy of Sciences of the United States of America,* **107,** 8035-8040.

Monfreda, C., Ramankutty, N. & Foley, J. A. (2008) Farming the planet: 2. Geographic distribution of crop areas, yields, physiological types, and net primary production in the year 2000. *Global Biogeochemical Cycles,* **22**.

NBS (2006) China statistical yearbook. National Bureau of Statistics of China, Beijing.

Potter, P., Ramankutty, N., Bennett, E. M. & Donner, S. D. (2010) Characterizing the spatial patterns of global fertilizer application and manure production. *Earth Interactions,* **14,** 1-22.

Ramankutty, N., Evan, A. T., Monfreda, C. & Foley, J. A. (2008) Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000. *Global Biogeochemical Cycles,* **22**.

Ravindranath, N. H., Somashekar, H. I., Nagaraja, M. S., Sudha, P., Sangeetha, G., Bhattacharya, S. C. & Abdul Salam, P. (2005) Assessment of sustainable non-plantation biomass resources potential for energy in india. *Biomass and Bioenergy,* **29,** 178-190.

Sheldrick, W., Keith Syers, J. & Lingard, J. (2003) Contribution of livestock excreta to nutrient balances. *Nutrient Cycling in Agroecosystems,* **66,** 119-131.

Smil, V. (1983) *Biomass energies*. Plenum Press, New York.

Smil, V. (1999) Crop residues: Agriculture's largest harvest. *Bioscience,* **49,** 299-308.
Smil, V. (2000) Phosphorus in the environment: Natural flows and human interferences. *Annual Review of Energy and the Environment,* **25,** 53-88.
USDA-ERS (2010) Fertilizer use and price. United States Department of Agriculture, Economic Research Service, Washington.
USDA-NASS (2008) United states census of agriculture - fertilizers and chemicals applied: 2002 and 1997. United States Department of Agriculture, National Agricultural Statistics Service, Washington.
USDA-NRCS (2009) Crop nutrient tool: Nutrient content of crops. United States Department of Agriculture, Natural Resource Conservation Service, Washington.
Wint, G. R., and T. P. Robinson (2007) Gridded livestock of the world - 2007. *(Food and Agriculture Organization of the United Nations/Rome, Italy)*.

2.9. SUPPORTING INFORMATION TABLES AND FIGURES

SI TABLES

Table S1. Comparison of total global agronomic P inputs, outputs and balances for this study and those calculated from other global studies. Studies are differentiated based on whether they were calculated for total croplands, croplands and grasslands (including pastures and cultivated grasslands), or total agricultural lands (the sum of all cultivated lands and pastures).

		Bouwman et al. (2009) ***Agricultural soils*** *2000 estimates*			***Cropland soils***		***Agricultural soils***
	This study	Croplands soils (excluding grasslands)	Grassland soils (including pastures)	Total	Sheldrick et al. (2002) *1996 estimates*	Smil (2000) *circa 1995 estimates*	Liu et al. (2008) *2004 estimates*
Fertilizer P applications	14.2	>13	<1	14	12.7	14 to 15	14.7
Manure P applications	9.6	7	10	17	7.7	6 to 8	2.5
Crop P removal	12.3	10	6	16	11.6	8 to 9	8.2
Agronomic balance	11.5	11	4	15	8.8	12 to 14	8.7

SI FIGURES

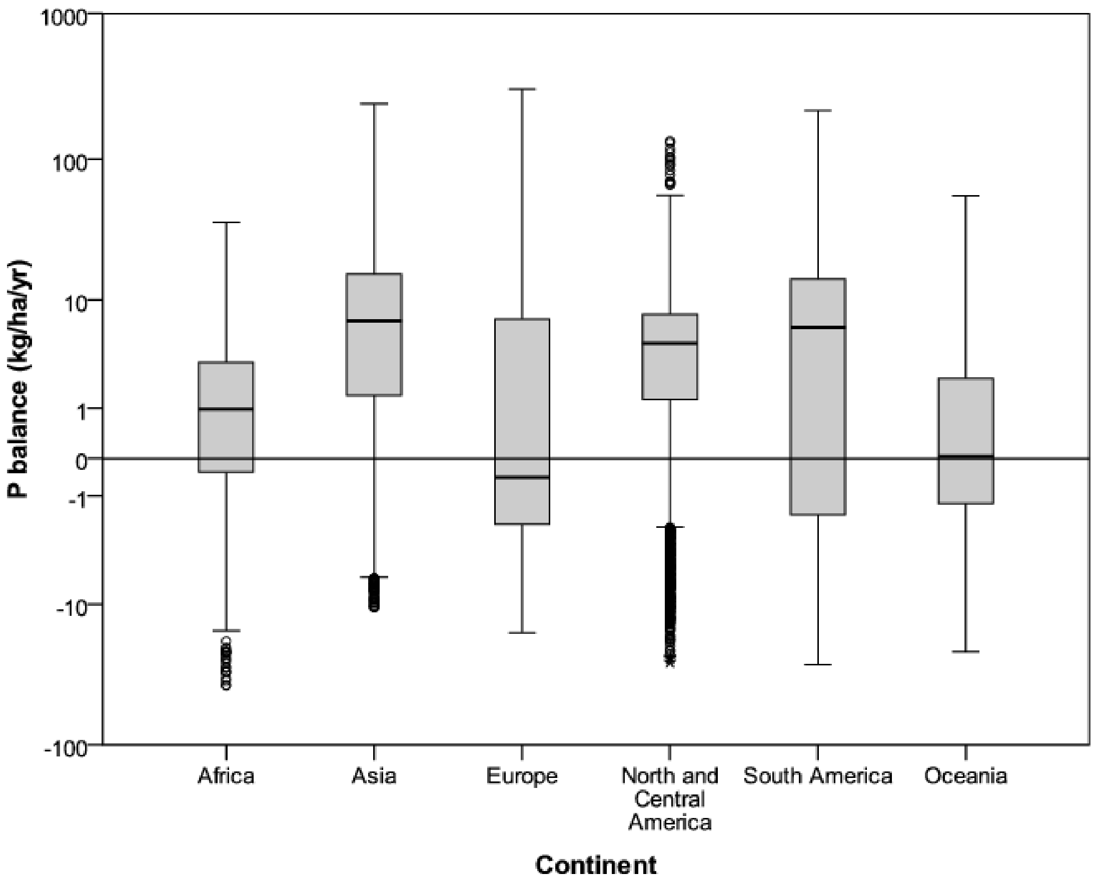


Fig. S1. Box-and-whiskers plot showing median grid-cell P balances, quartile ranges, and outliers for different continents (for all grid cells with >5% cropland area). Note the logarithmic scale on the y-axis. Russia is included in the Europe grouping.

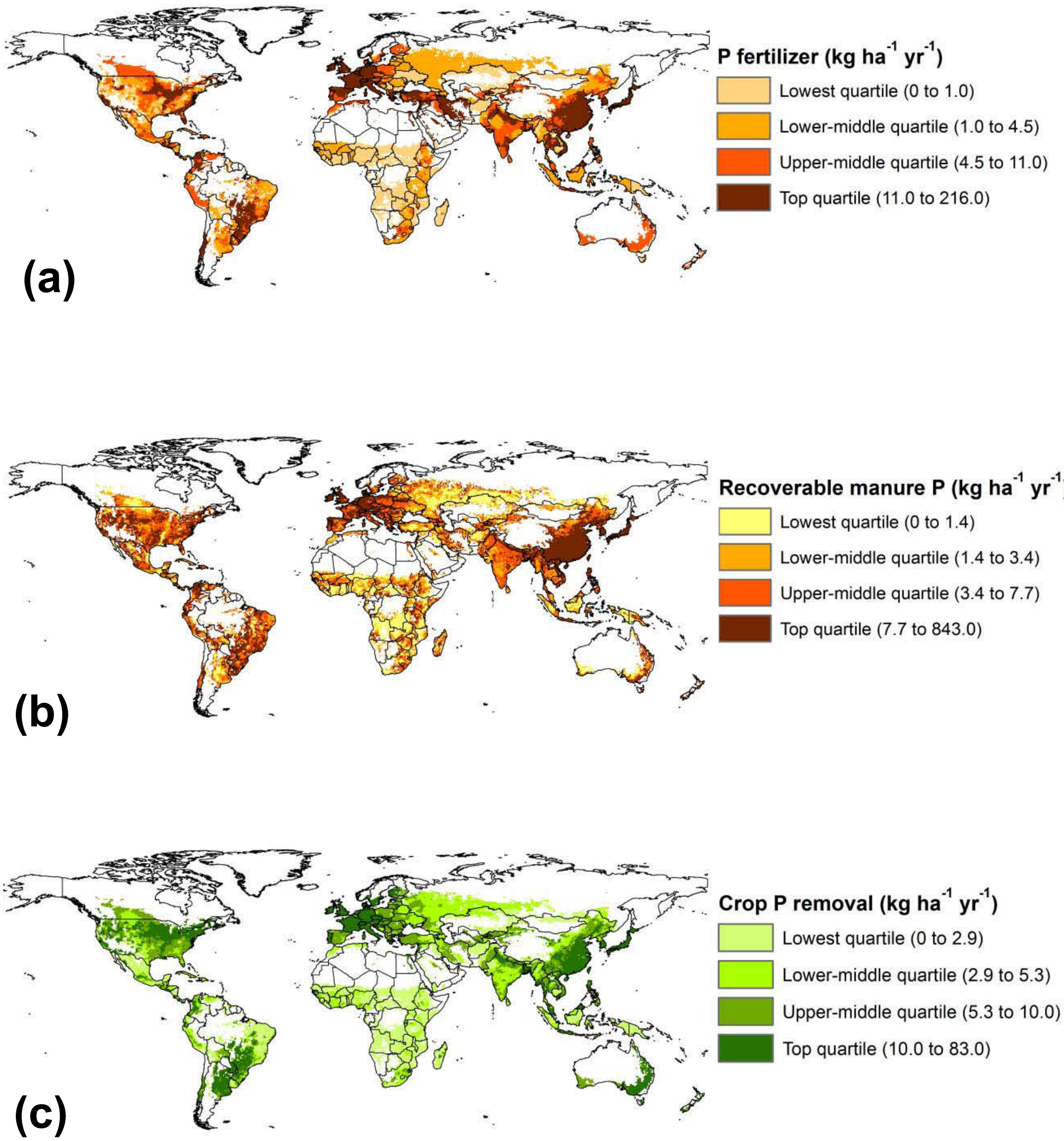


Fig. S2. The main agronomic P inputs (fertilizer (*a*) and recoverable manure applied to croplands (*b*)) and outputs (removal of P by harvested crops (*c*)) considered in this study, based on data for approximately the year 2000. All flows are expressed in kg P ha^{-1} yr^{-1} of total cropland area (Ramankutty *et al.* 2008a), which includes temporary fallow lands in some countries.

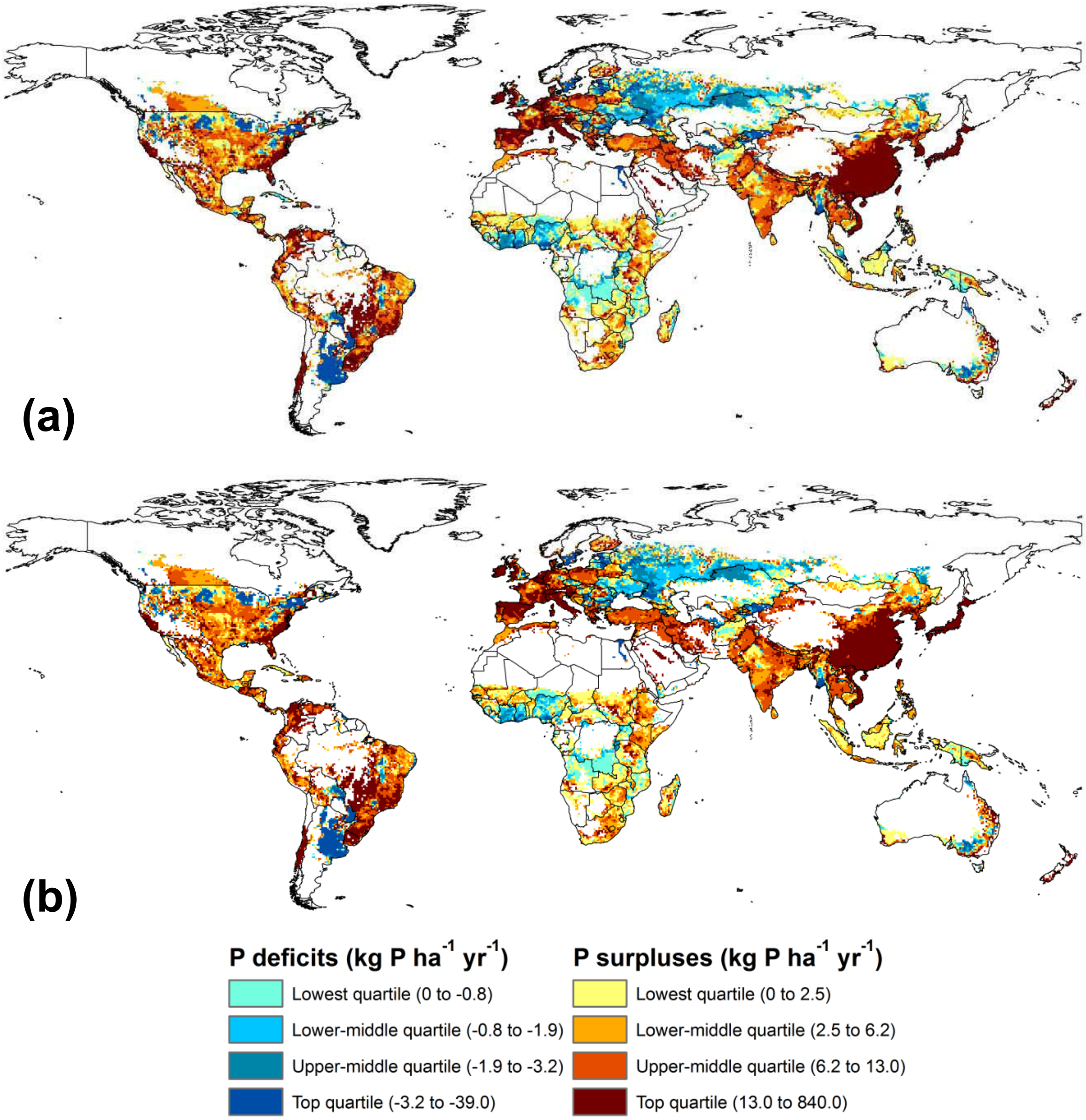


Fig. S3. Results of global agronomic P balances based on contrasting scenarios for crop residue recycling (as additional P input) and removal (as additional P output) for developed and developing countries globally. High crop residue removal scenario with low residue recycling (*A*) and low crop residue removal scenario with high residue recycling (*B*).

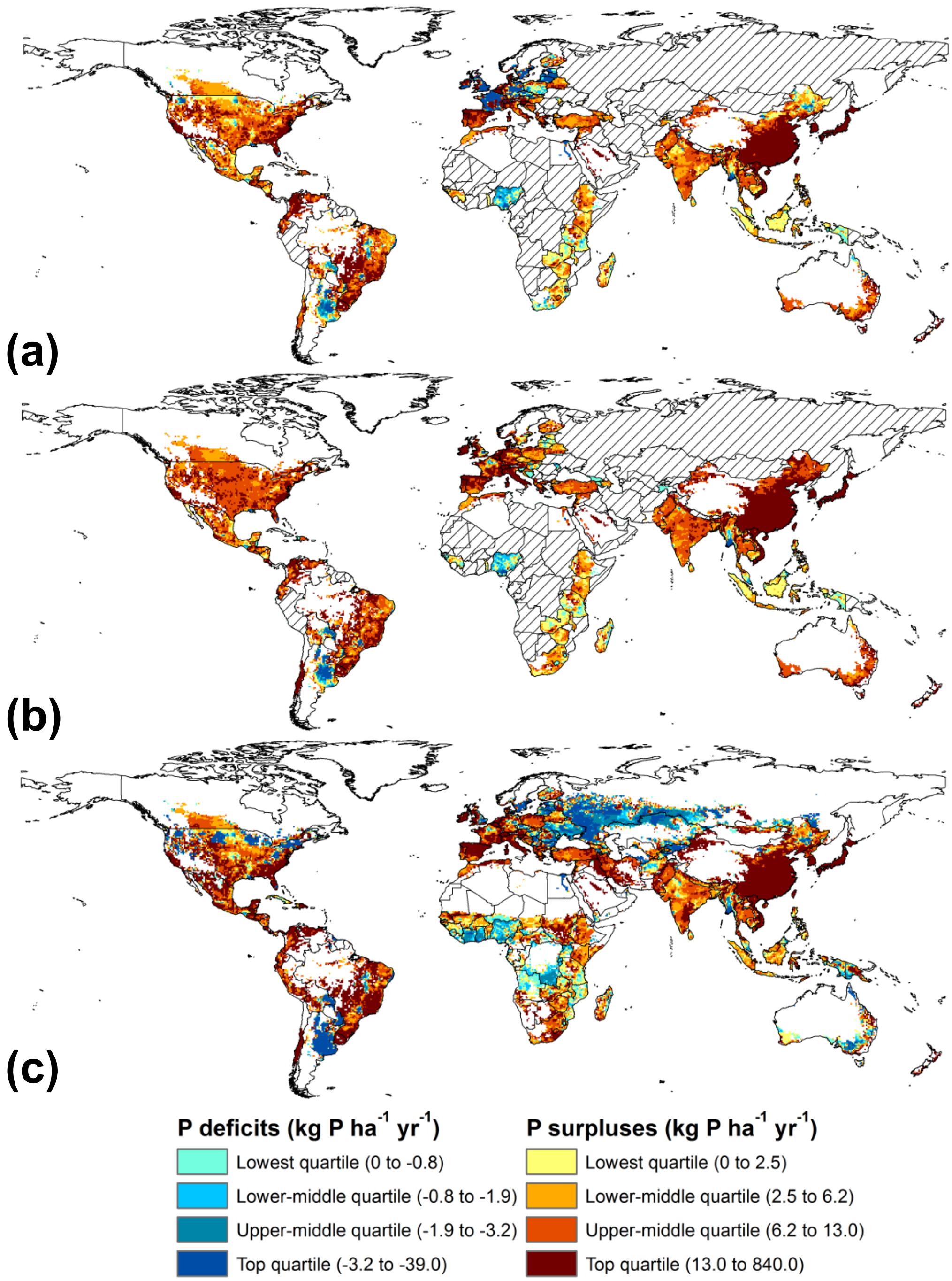
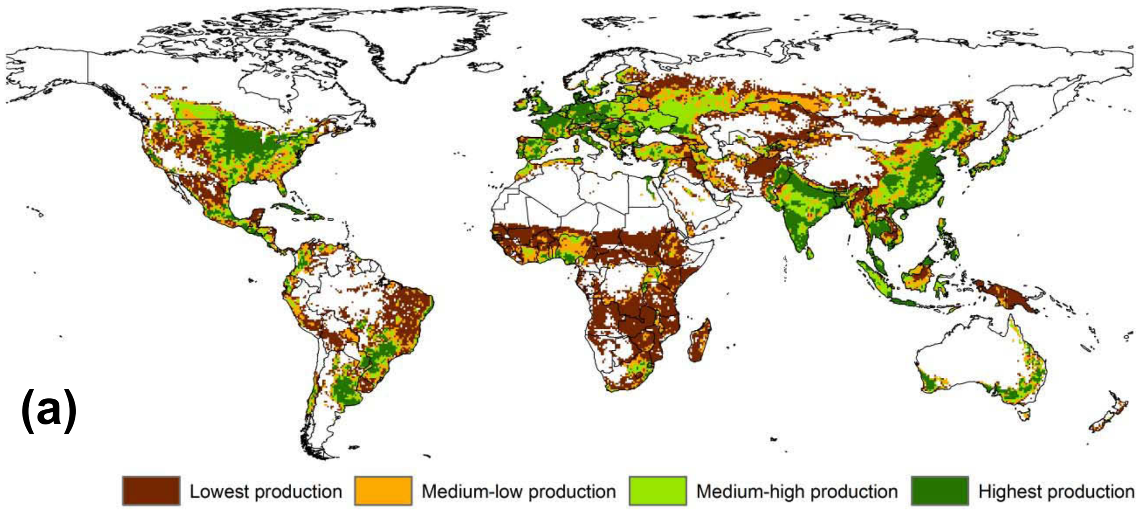


Fig. S4. Agronomic P balance map based on calculations of crop P removal limited to crops that received P fertilizer applications circa 2000 based on IFA (2002) for all countries with crop-specific P fertilizer data (*a*). Agronomic P balance map for all non-forage croplands in countries with crop-specific fertilizer data (*b*); these results are based on excluding all P fertilizer inputs and crop P removal associated with forage crops. P balance map using the total harvested areas from Monfreda et al. (2008) as the denominator to adjust the balances per unit of cropland area (*c*).



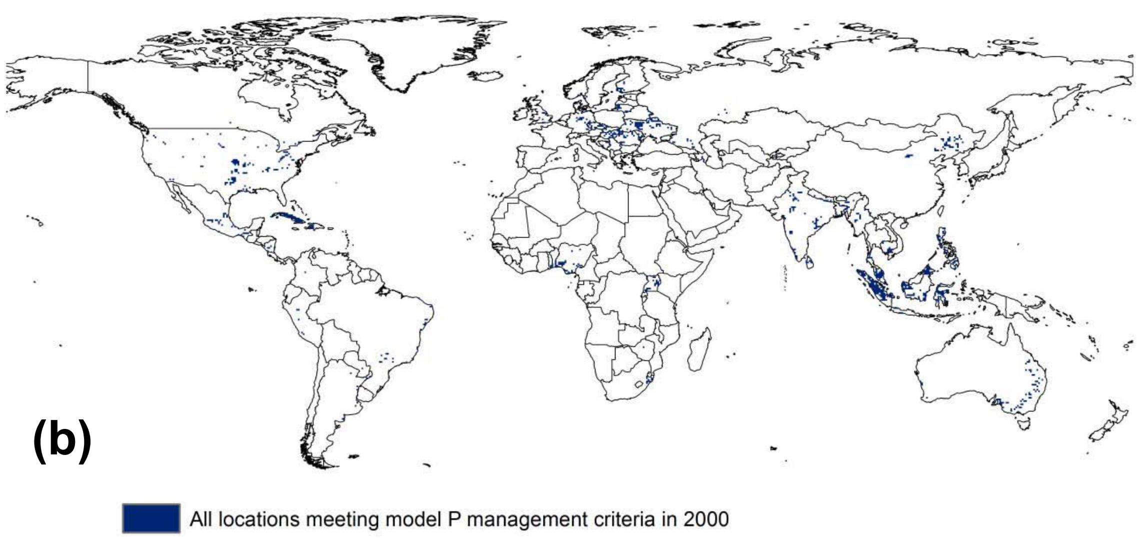


Fig. S5. Total crop production (dry-matter basis) for 123 crops from Monfreda et al. (2008) classified from low to high based on global quartile ranges (*a*). All locations globally meeting the model P management criteria described in the Discussion section of the main text (i.e., first quartile surplus or deficit, medium-high or high P-use efficiency, and medium-high or high crop production) (*b*).

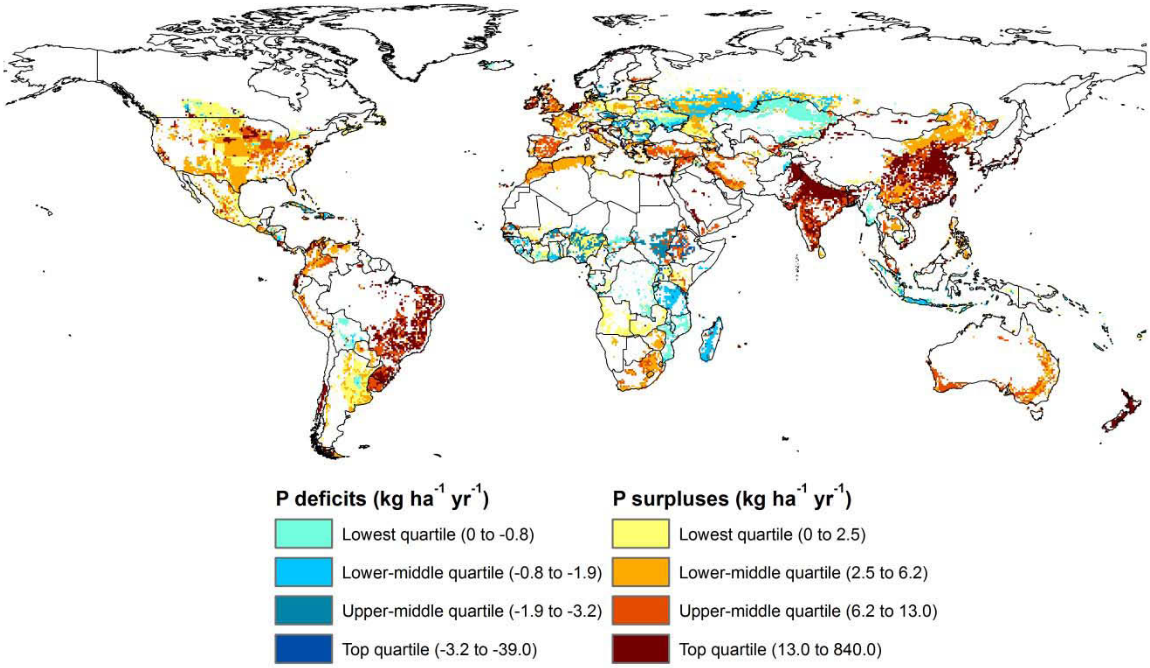


Fig. S6. Agricultural P balances calculated using data from the IMAGE (Bouwman *et al.* 2009) model for P inputs and outputs adjusted to provide comparison with our global agronomic P balance results. The P surpluses and deficits are grouped according to the same quartile ranges calculated in this study.

CONNECTING STATEMENT

In Chapter 2, I considered the geographic patterns of agricultural P management at the global scale and discussed the cumulative implications of P imbalances at smaller scales. Chapter 3 extends this by examining some of the underlying drivers of agricultural P use regionally, using a study of the United States agricultural system. In particular, I examine how the P imbalances identified in Chapter 2 might be linked across regions via the P use that is 'embodied' in agricultural trade.

This study provides a detailed assessment of the commodities that are driving demand for P fertilizer use between the US and its trading partners globally. Some of the main P losses along the agricultural supply chain are assessed, including P that is (temporarily) lost from the food system due to accumulation in agricultural soils. I then discuss opportunities to leverage P-use efficiency by considering P flows throughout the full US agricultural system, from the farm to consumer level, and how these are influenced by trade.

CHAPTER 3: EMBODIED PHOSPHORUS AND THE GLOBAL CONNECTIONS OF UNITED STATES AGRICULTURE

Graham K. MacDonald, Elena M. Bennett, Stephen R. Carpenter

Submitted to: *Environmental Research Letters*

3.0. ABSTRACT

Phosphorus (P) use in agriculture is intricately linked to food security and water quality. Globalization of agricultural systems and changing diets alter these relationships, yet the implications of these changes for managing non-renewable P reserves are unclear. We assessed P fertilizer used for production of food crops, livestock, and biofuels in the US agricultural system, emphasizing P flows embodied in the production of imports and exports. By far the largest demand for P fertilizer was for feed and livestock production (56% of all P use for domestic and imported products). Most of the mineral P inputs to US domestic agriculture in 2007 (1905 Gg P) were retained in agricultural soils (28%) or associated with post-harvest losses (40%) and biofuel refining (10%). One quarter of P fertilizer in the US was used for producing exports, particularly crops, driving a large net P flux out of the country (338 Gg P) that had varying influences on domestic cropland P balances. However, US meat consumption involved considerable reliance on P fertilizer use in other countries to produce imported red meat. Changes in domestic farm management and consumer waste could together reduce the P fertilizer needed to produce food consumed in the US by half, which is comparable to the P fertilizer reduction attributable to cutting meat consumption (44%).

3.1. INTRODUCTION

Agricultural phosphorus (P) use has multiple agronomic, environmental, and economic dimensions that are central to both agricultural productivity and sustainability (Childers *et al.* 2011). Growth in P fertilizer use has been an important component of improving agricultural production (Tilman *et al.* 2002), particularly in the US, where yields for many crops are among the highest in the world (Foley *et al.* 2011). However, changes in the agricultural sector, especially more intensive crop and livestock production, have resulted in sustained large P surpluses in many key producing countries (Bouwman *et al.* 2011; Schipanski & Bennett 2012). Long-term agricultural P surpluses pose difficulties for regulation of surface water quality (Kleinman *et al.* 2011) that has major economic costs in the US (Dodds *et al.* 2008). For example, P runoff from agricultural lands is an important driver of hypoxia in the Gulf of Mexico (Jacobson *et al.* 2011).

Modern agriculture is ultimately dependent on inputs of mineral P, a key plant nutrient derived from finite supplies of phosphate rock that have no substitute (Smil 2000; Cordell *et al.* 2009). Rising meat consumption, population growth, and continued diversion of crops to biofuel use pose additional pressures for managing these reserves (Simpson *et al.* 2008; Van Vuuren *et al.* 2010; Hein & Leemans 2012). At the same time, the food system is subject to large P losses between the farm and consumer (Matsubae *et al.* 2011; Suh & Yee 2011; Wang *et al.* 2011). These losses of mineral P inputs represent potential waste of an essential non-renewable resource, often with presently limited opportunity for recovery (Cordell *et al.* 2009). Together, these issues raise serious concerns about future costs or access to P fertilizers (Cordell *et al.* 2009), although global depletion rates are subject to considerable uncertainty (Van Vuuren *et al.* 2010).

Food production is becoming progressively globalized, increasing the distance between locations of food production and consumption (Naylor *et al.* 2005; Krausmann *et al.* 2008). This spatial disconnection can concentrate negative externalities on non-food ecosystem services in producing regions (Bennett & Balvanera 2007; Erb *et al.* 2009), while introducing new nutrient sources to importing regions that can be difficult to recycle (Schipanski & Bennett 2012; Senthilkumar *et al.* 2012). This situation is exacerbated by growing trade in food, feed, and animal products. However, previous national P budget studies have typically only given basic treatment to the role of trade on recovery of mineral P inputs from farm to food (Cordell *et al.* 2012), despite potentially large linkages across national boundaries (Schipanski & Bennett 2012).

Physical movement of P contained in traded agricultural commodities has the potential to vastly alter P fluxes between regions (Beaton *et al.* 1995; Grote *et al.* 2005) due to limited mobility of P in the biosphere (Smil 2000). Yet, the indirect, or 'embodied', component of P fertilizer use needed to produce traded goods may have even greater implications for both the agronomic and environmental dimensions of managing non-renewable P supplies (Matsubae *et al.* 2011; Schipanski & Bennett 2012). The importance of incorporating the resource flows embodied in the production of traded commodities has been established for the carbon and nitrogen cycles (Ciais *et al.* 2007; Galloway *et al.* 2007; Davis & Caldeira 2010), as well as for water footprints of agricultural commodities (Hoekstra & Mekonnen 2012).

The US is an exemplary case for full-system accounting of agricultural P use, including trade components, and its implications for managing non-renewable P supplies. The US produces and consumes a large share of global agricultural production, and is a leading exporter of many staple agricultural commodities (USDA-FAS 2012c). The US is also the largest manufacturer and exporter of inorganic P fertilizers (FAO 2012; USDA-ERS 2012c), and currently the third largest global consumer of P fertilizers (Heffer 2009). However, disparities in the locations of remaining phosphate rock reserves globally (Childers *et al.* 2011) mean that it is both producing and consuming P fertilizers at a high rate relative to its own reserves.

We investigated P fertilizer use in the US at the interface between trade, biofuel production, and diets given the context of limited national phosphate rock reserves and environmental concerns. We consider P fertilizer used for specific commodities throughout major components of the US agricultural system, including domestic or foreign production and consumption, thereby expanding considerably on previous modelling efforts (Suh & Yee 2011). Using detailed national data, we examined three questions: 1) how is P use allocated to the production of specific commodities within the US agricultural system?; 2) how does this distribution influence the fate of mineral P inputs from agricultural soils to the consumer level?; and 3) what magnitude of these P flows are linked to international agricultural trade? Finally, we discuss opportunities to mitigate excess P fertilizer use based on quantitative scenarios.

3.2. Methods

Our model tracks P flows through major components of the entire US agricultural system. We determined the quantities of 52 crops used for food, feed, or biofuels (corn ethanol and soybean biodiesel) domestically, as well as the quantity of these crops and major livestock products that were imported or exported. P flows associated with each use were calculated based on total supply (production adjusted for annual stocks, imports and exports) of each crop in 2007 using national statistics from the US Department of Agriculture (USDA) and the United Nations Food and Agriculture Organization (FAO) (see supporting information for a detailed overview of all sources and methods). We specifically compared the magnitude of P flows related to US production for domestic consumption, foreign production for US consumption, and US production for foreign consumption. We refer to the P associated with the production of, but not necessarily contained in, traded crop and livestock commodities as 'embodied' P use.

3.2.1. P inputs to domestic agricultural soils and livestock systems

Soil P balances were calculated for each crop as the difference between fertilizer and manure P inputs as well as P outputs in crop harvest. We also considered aggregate P balances for all agricultural soils, including manure P deposited on pastures, P uptake by pasture-grazed livestock, and P removed in crop residues used as livestock feed. Removal of P in harvested crops was estimated using the typical dry-matter P contents for the harvested portions of each crop (USDA-NRCS 2009) and crop-specific production data (USDA-NASS 2009).

The quantity of P fertilizer applied to major crops nationally in 2007 was obtained from the USDA (USDA-ERS 2010) (corn, soybean, cotton, and wheat) and from the International Fertilizer Industry Association (Heffer 2009) (rice and sugar crops). A source of recent national P fertilizer applications for other individual crops was unavailable. We therefore disaggregated total national P fertilizer applied to groups of crops in 2007 (i.e., fruits and vegetables, other oilseeds, and other cereals) (Heffer 2009) using crop-specific P fertilizer use data from the late 1990s (IFA/IFDC/IPI/PPI/FAO 2002). A detailed description of data and assumptions regarding P fertilizer application to silage corn, hay, and pasture is provided in the SI text.

Manure P use was estimated based on detailed livestock inventories (USDA-NASS 2008a; USDA-NASS 2009) and national management data. Total manure P production was calculated using inventories for 14 livestock classes multiplied by annual manure P excretion factors for US livestock (Kellogg *et al.* 2000; ASAE 2003; Maguire *et al.* 2007) (see SI text). Manure collected on farms for cropland application was estimated using livestock-specific fractions for manure recovery from housed animals, assuming subsequent 15% losses on farms (Kellogg *et al.* 2000). We assumed that remaining manure was deposited on pastures (for ruminants) or lost on farms (poultry and pigs). Manure application to specific crops was then estimated using national farm survey estimates (Macdonald *et al.* 2009) reported for 8 major field crops (72% of the total applications) and hay crops (26.3%). Application of remaining cropland-available manure was estimated according to the relative fraction of crop P demands from each remaining crop.

To relate P fertilizer use for feed crops to the production of each livestock class, we first calculated total P consumed by animals and the sources it was derived from. Total P intake was estimated using mass-balance, where P requirements are assumed to equal total manure P produced plus the amount of P stored in livestock bodies and yielded in products (Schussler *et al.* 2007). P contained in live animals and products was estimated based on average lifetime weights (Lander *et al.* 1998) and annual egg or milk production per animal (USDA-ERS 2012d). We assumed that all available feed crops, silage, and hay (adjusted for imports and exports) were fed to livestock in the US, accounting for ~4% crop spoilage (Gustavsson *et al.* 2009). Other P intake sources included industry data on US mineral P supplement consumption (PotashCorp 2006), reported use of animal by-product protein feeds (USDA-ERS 2011a), and approximate crop residues available for livestock consumption from 17 major feedstocks (Milbrandt 2005; Gallagher *et al.* 2006) (detailed in the SI text). All remaining P requirements were assumed to be met by pasture grazing.

To account for the relative fractions of total P intake from pasture-grazed versus feed or other forage for each livestock class, we used data on the fraction of grain or roughage in diets of seven livestock categories (USDA-ERS 2011a). We differentiated between P intake from pasture and other forms of roughage using the ratio of P available from hay and silage sources to P from pasture grazing.

3.2.2. P contained in traded products

Flows of P directly associated with imported and exported crop commodities were estimated using the fraction of production that was traded and product specific P contents. Imports and exports of live animal, meat, and dairy were obtained from a USDA trade database, with imports adjusted for re-export (i.e., subsequent export of imported commodities, possibly after processing) (USDA-FAS 2012b). We also calculated the P contained in traded phosphate rock and fertilizers using fertilizer trade statistics and typical P contents (USDA-ERS 2012c).

3.2.3. P flows embodied in US exports

P flows associated with the production of exported crops and livestock products were estimated based on the share of total domestic production destined for export. For livestock, we added the net domestic fertilizer, manure, and crop P uptake associated with feed or pasture intake by animals that were ultimately exported. Uses of crops produced in the US but consumed abroad were estimated based on national averages of food, feed and other uses from FAO (2012), weighted according to the fraction of exports for each major destination country (≥5% of exports for a given crop).

To assess the influence of these embodied P flows on soil balances, we calculated soil P use efficiencies for exported crops (PUE; crop P removed per unit of P input (Schipanski & Bennett 2012), expressed as a percentage) and cropland P balances for traded commodities (net P inputs minus net crop P outputs associated with imports or exports).

3.2.4. P flows embodied in US imports

We calculated the P fertilizer used abroad to produce the quantity of each crop product imported, as well as the P removal associated with that product. For each crop and each major source country, we used 2007 yields (FAO 2012), circa year 2000 P fertilizer application rates (IFA/IFDC/IPI/PPI/FAO 2002), and crop P contents to determine how much P fertilizer was applied. These calculations were weighted according to the share of imports from each country with ≥5% of imports for a given crop to reflect differences in production.

For P fertilizer applied to feed crops used to produce imported livestock, we first calculated the live animals associated with livestock product imports and the approximate sources of feed intake for these animals. Annual manure P production and P intake (by mass-balance) was estimated using average P excretion and live animal weights for OECD countries (OECD 2008; FAOSTAT 2011). We then used feed composition data from Statistics Canada (2003) for the relative fraction of intake from grains, forages, and non-grain feeds because Canada is the principal exporter of livestock to the US. P fertilizer applied to these feed sources was next estimated, as above, for each source country that accounted for ≥5% of livestock imports for each product. Application of manure P to either cropland or pasture by animals associated with imported livestock products was estimated following the basic method of Schipanski & Bennett (2012) (as described in the SI text).

3.2.5. Domestic food consumption and alternative scenarios for 2007

We estimated domestic consumption of P, kilo-calories, and protein in food for each crop and livestock product after adjusting for traded fractions, losses, and exclusion of crops used for seed or other industrial purposes (e.g., cotton fibre). Meat availability was estimated based on the number of livestock slaughtered on farms and commercial operations (USDA-NASS 2008b), excluding animals that were exported. Estimates of apparent processing losses were based on differences in the weights and P contents of the root commodity (harvested crop or live animal) and processed weights and product specific P contents, using conversion factors where necessary (FAOSTAT 2011; USDA-ERS 2011b). Total food loss estimates at the retail to consumer level from the USDA (USDA-ERS 2011b) for individual commodities (or product groups) were then used to determine food availability.

We examined three dimensions of P fertilizer used to produce food (kg of P fertilizer per unit of consumption) in terms of kg of dietary P, million kilo-calories, and kg of protein consumed after losses. These estimates exclude P fertilizer used to feed breeding animals. Because beef cows, sheep, and goats typically require multiple years of feed prior to being consumed as meat, we included a conservative adjustments for the fertilizer used during the life-cycle of these animals equivalent to ~2 years (detailed in the SI text).

Finally, we applied our model to compare the relative amount of P fertilizer used for domestic consumption (including that embodied in imports but excluding export production) under a series of hypothetical alternative scenarios for 2007. These scenarios illustrate opportunities to reduce P fertilizer use based on cumulative supply-side (farm level) or demand-side (consumer level) changes and the sensitivity of our results to changes in underlying factors.

3.3. RESULTS

Our results show that approximately 8% of the total mineral P fertilizer and livestock feed supplements used in the US was consumed in food domestically during 2007 (Fig. 1). The remaining P can be accounted for through accumulation in agricultural soils (28%), apparent post-farm losses from the food system (27%), P contained in US agricultural exports for consumption in other countries (21%), other on-farm losses (13%), and P embedded in biofuel refinement (10%).

3.3.1. Detailed account of P flows for domestic agriculture

Production of feed and livestock resulted in by far the greatest cumulative P flows in the US (Fig. 2). Fertilizer application to feed crops (859 gigagrams, Gg, P) was 42% greater than that applied to food crops (605 Gg P). Feed crops and pasture also received considerably more manure P inputs than other croplands (87% of the 1,466 Gg P total), and had greater overall P uptake via crop harvest and grazing than that from food or biofuels. Biofuels accounted for 11% of total domestic P fertilizer applications (196 Gg), almost entirely for corn ethanol. This is comparable to the magnitude of mineral P used as livestock feed supplements (193 Gg).

There was an overall surplus of P applied to US agricultural soils (534 Gg across croplands and pasture). The soil P balance of crops was on average 3.5 kg P ha^{-1} nationally when weighted according to the harvested area of each crop, but varied greatly among crops (simple average of 18.0 kg P ha^{-1} with a standard deviation of 25.0 kg P ha^{-1}: see Table S1 for a detailed breakdown). This can largely be explained by the disproportionate effects of 3 major crops: corn (surplus of 437 Gg P) and soybean (deficit of 219 Gg P) for feed and biofuel, and wheat (surplus of 84 Gg P) for food crops.

Post-harvest farm losses, apparent processing losses, and losses via distribution or consumer waste considerably reduced the P available for human consumption. The loss terms estimated here include spoilage of feed crops (40 Gg P), uncollected manure on farms (232 Gg P), and by-product P flows from processing of food crops (243 Gg P). Two-fifths of the P lost during livestock slaughter and at the retail level was returned to livestock as by-product animal protein feed (121 Gg). Although actual biofuels contained little P (at most ~3 Gg), the P embedded in crops used for fuel refining (after excluding major co-products) further limited the amount of P consumed in human diets.

3.3.2. P contained in traded products

Phosphate rock and fertilizer trade resulted in a large P flux directly between the US and other countries. A total of 667 Gg P was imported to the US, approximately half of which was phosphate rock from Morocco; 1560 Gg P was exported, of which 99% was processed P fertilizers where by far the largest destination country was India (41%).

The P contained in traded agricultural commodities was largely associated with food and feed crop exports (128 and 244 Gg, respectively, after adjusting for losses due to processing in the US). Only 17 Gg P was imported in livestock products (mostly in live animals) and 12 Gg P was exported (mostly in meat products).

3.3.3. Embodied P flows and fertilizer displacement via trade

Fig. 3 shows the magnitude of P inputs (fertilizer and manure applied) and P outputs (crop and grazing uptake) associated with production of exports from and imports to the US relative to P flows in the US for domestic consumption only. One quarter (25%) of domestic P fertilizer was allocated to production of exports (167 and 126 Gg P for food and feed crops, respectively). Much of the P fertilizer applied to exported food crops was ultimately contained in the traded product (84%; largely in wheat going to Japan, Egypt, Nigeria, Mexico, among other countries). More P was exported in feed crops than was applied to soils due to the high PUE of soybean nationally in 2007 (47% of soybean exports went to China) (Fig. 4). This was despite the lower PUE of corn (40% of which went to Japan and Mexico). P flows embodied in the production of exported crops considerably exceeded those associated with imports from other countries.

Of the total P fertilizer used to produce food consumed in the US in 2007 (≈1386 Gg), 16% was applied in other countries to produce imported commodities. P fertilizer use embodied in the production of imported livestock (144 Gg) was 10% of total P fertilizer required to produce food for domestic consumption, and considerably larger than the P embodied in the production of US livestock exports (62 Gg). Pig and beef imports accounted for 82% of the embodied P fertilizer used abroad for livestock imports, predominantly from Canada and Australia (73%).

In contrast to imports, poultry was the largest consumer of P fertilizer use embodied in US exports (43% of P fertilizer use for livestock) that went largely to Russia, Mexico, and China (52%). P fertilizer use embodied in pig exports accounted for another 24% (largely for Japan and Mexico), while beef accounted for the remaining 19% (for Mexico, Canada, and Japan). A relatively higher fraction of P intake by livestock produced in the US for domestic consumption was from pasture grazing (e.g., by cattle), which had proportionally lower fertilizer P use than feed crops (Fig. 3).

Cropland P balances associated with US imports and exports differed due to the dominance of red meat (beef, pork, and other ruminants) imports to the US versus the dominance of crop exports from the US. Of the total cropland P surplus in the entire US agricultural system (the US and its trading partners), 16% occurred in countries producing US imports (Fig. 5). The net P surplus in the US due to export production was a relatively smaller fraction of the total (~ 5%) given underlying differences between the P balances for exported food crops (surplus of 39 Gg P) relative to livestock and feed exports (deficit of 10 Gg P) (Table S2). This disparity reflects the influence of soybean exports from the US used for feed abroad. Unlike corn and wheat, soybean harvest relied on soil nutrient stocks considerably more than P fertilizer use in 2007 (Fig. 4). However, P uptake by soybean may have been supplemented by fertilization of corn in previous seasons in areas under corn-soy rotations.

3.3.4. Relative contribution of trade versus domestic P flows

The total P contained in traded crop and livestock products as well as the P fertilizer use embodied in their production (Fig. 6) was substantially larger for US exports (784 Gg P) than for US imports (298 Gg P). The US was therefore a substantial net exporter of domestic mineral P use that was ultimately embedded or embodied in traded goods in 2007. Additionally, direct export of P fertilizers exceeded these other trade-related flows combined. The P flux in exported fertilizer was even slightly greater than the entire mineral P used in the US for domestic consumption.

3.3.5. P requirements for US food and their embodied import shares

The quantity of P fertilizer used either domestically or abroad to produce food consumed in the US differed greatly among commodities (Fig. 7). Production of livestock products accounted for 57% of total P fertilizer demands for food consumed in the US in 2007. In particular, red meat had the largest overall P use with by far the greatest reliance on embodied P fertilizer use abroad (37%) in order to produce US imports. Poultry had a relatively lower P demands in terms of protein despite comparable caloric returns to beef, as well as a much lower reliance on foreign P fertilizer use for imports (7%). Fruit and other cereals had relatively low contributions to total P fertilizer use and relatively small calorie footprints, but had higher reliance on embodied P fertilizer imports (24-28%).

The P fertilizer used per unit of P consumed in different foods in the US is presented in the supplemental text (Table S3). These reveal strong differences in P fertilizer use and food system losses between major food crops. Corn products had a dietary P requirements comparable to poultry (>20 kg P / kg P consumed) and much higher than wheat (6 kg P / kg P consumed). This reflects the products derived from corn (e.g., high-fructose corn syrup) and the larger domestic P surplus for corn (12 kg P ha^{-1}) versus wheat (4 kg P ha^{-1}).

3.3.6. Scenarios of national fertilizer reduction

Changing farm management and minimizing food waste could have large overall implications for reducing P fertilizer use in the US agricultural system, by as much as half if these were tackled together. Balancing P inputs and outputs on a crop-by-crop basis nationally (S1) would result in a 23% decrease in P fertilizer use, largely from reducing fertilizer P for corn, wheat, cotton, and potato. Similarly, a 20% decrease in P fertilizer use could be achieved if consumer food waste was dramatically reduced (S3), and, as a result, overall food demand was lowered.

The greatest single reduction in fertilizer use (44% decrease) would occur under a shift to lacto-ovo vegetarian diets given the P fertilizer demands of meat products (Table 1; Scenario 5); however, P fertilizer use for food crops would need to increase in order to meet protein requirements (offsetting ~126 Gg P of the P savings). Replacing 20% of beef protein by poultry protein (S4) would reduce total P fertilizer by ~5% (ranging from 52 to 65 Gg P depending on whether feed supplement use for poultry is included). Improved recycling of manure P as an organic fertilizer source via utilization of on-farm losses could therefore have a greater impact on reducing national P fertilizer use (by 11%) than a partial shift from beef protein to poultry (S2).

3.4. Discussion

Mineral P use was distributed disproportionately to the production of a relatively small number of commodities in the US agricultural system in 2007, with important implications for potential P losses. More than half of the mineral P used in the US was devoted to feed and livestock production, largely to produce meat for domestic consumption and corn or soybean exports. Meat had substantially higher P use and associated losses per kilo-calorie consumed compared to all other foods (e.g., Foley *et al.* 2011). Corn ethanol entailed a further 11% of total domestic P fertilizer use in 2007 (compared to 2% for first-generation biofuels globally (Hein & Leemans 2012)), exceeding the P fertilizer used in the US to produce any major food crop except wheat (13%). Much of the mineral P used nationally contributed to soil P surpluses for key crops (e.g., corn and wheat). In turn, products derived from some crops were linked to relatively high post-harvest P losses, with a similar amount of P entering waste streams after harvest as that which was retained in agricultural soils (Fig. 1).

Agricultural trade accounted for one quarter of domestic P fertilizer use and facilitated strong linkages in P fertilizer applications between the US and its trading partners. In total, 16% of the P fertilizer used to produce food consumed in the US was applied in other countries for foreign imports. Unlike trade in animal products, which contains relatively little P, crops exported from the US created a large P flux out of the country, reinforcing it as a major net exporter of mineral P. The magnitude of P flows embodied in the production of traded commodities stresses the need to explicitly incorporate trade into national P budgets (Matsubae *et al.* 2011), particularly for countries with export-oriented agriculture such as the US (Schipanski & Bennett 2012).

Continued depletion of US phosphate rock reserves provides further incentive to use mineral P more efficiently. The US was the third largest phosphate rock producer globally and the leading exporter of phosphate fertilizers (~25% of the global total) in 2010 (Jasinski 2012a). Current forecasts suggest that depletion of domestic phosphate rock reserves could occur within decades (Vaccari 2009; Childers *et al.* 2011; Jasinski 2012b), although projecting non-renewable resource depletion is confounded by many factors. Opportunities to reduce P fertilizer use and mitigate P losses from the farm to consumer level could collectively help to slow the pace of domestic phosphate rock depletion (Table 1), possibly lessening dependence on fertilizer imports (Childers *et al.* 2011). We explore the importance of such changes in the context of growth in US agricultural production, trade, and potential environmental externalities.

3.4.1. Achieving more sustainable P use in US agriculture

Excessive P use is a concern in US agriculture given regional water quality problems and long legacies of past P fertilization on elevated soil P in some areas (Sharpley 2003; Dodds *et al.* 2008; Jacobson *et al.* 2011; Kleinman *et al.* 2011). Cropland P surpluses for corn and wheat disproportionately influenced the fate of mineral P introduced to the domestic agricultural system after accounting for manure recycling. Recuperation of P in future crop harvests could diminish total P fertilizer inputs needed in subsequent years (Sattari *et al.* 2012); for example, corn-soybean rotations are common agronomic practice in the US Corn Belt (Grassini *et al.* 2011), meaning that inter-annual coordination in P fertilizer and manure use for these crops may partially account for the P deficit for soybean nationally in 2007 (Macdonald *et al.* 2009; USDA-ERS 2010). However, P surpluses exacerbate P runoff to aquatic systems, particularly in areas susceptible to high P runoff due to other biophysical or management factors (Jacobson *et al.* 2011; Kleinman *et al.* 2011). Resolving P surpluses for major US crops will be central to reducing potential environmental externalities associated with feed, biofuel, and export crop production in the US (Foley *et al.* 2011).

Achieving more efficient use of mineral P may also depend on underlying demand for meat and corn ethanol. The USDA projects that US livestock production will increase despite stabilizing per capita meat consumption due to export growth, while corn used for ethanol doubled from 2007 to 2010 (2.3 to 5.1 million bushels) and is projected to rise slowly until 2020 (USDA-ERS 2011c). Assuming constant 2007 P fertilizer use-efficiency in our model, P fertilizer use for ethanol could increase to as much as 377 Gg P under 2020 projections. The quantity of P fertilizer used for ethanol may be increasingly viewed as a trade-off between US energy policy and consumption of non-renewable phosphate rock (Hein & Leemans 2012) that could also pose challenges for recycling feedlot waste back to croplands. Co-products from the distilling process for ethanol are increasingly used as livestock feed, comprising roughly 105 Gg P of livestock P intake in 2006/2007 (Hoffman & Baker 2011). These feeds typically contain more than double the P (0.8-0.9%) of corn grain, potentially increasing manure P concentrations (Simpson *et al.* 2008). Measures to offset probable increases in P use could therefore include feeding poultry less phosphate supplements, wider adoption of more P-efficient biofuel crops (e.g., soybean biodiesel) (Hill *et al.* 2006), and further livestock feed modification to minimize local manure P imbalances (Maguire *et al.* 2007).

Our finding that just 8% of mineral P inputs were consumed in US diets reflects the influence of high meat consumption, processing of crop products, and potential consumer food waste typical of some developed countries (e.g., France (Senthilkumar *et al.* 2012)). We found that reducing consumer food waste could play a key role in diminishing P losses from the US food system, but this may depend on changes in consumer behaviour. Alternatively, household food losses could be recycled to agricultural lands as an organic fertilizer source in greater quantities, which is currently done for ~2.5% of US household waste (Suh & Yee 2011). While our calculations include by-products from major crops used as animal feed (USDA-ERS 2011a), other national by-product uses of crops are highly uncertain (e.g., some P-containing effluent from biofuel refining may be recycled (NRC 2011)). National accounting of P flows in waste streams will be important for understanding the role of new technologies to recuperate these P losses for reuse in the food system (Cordell *et al.* 2012).

3.4.2. Accounting for trade

Trade results in a large flux of P from the US to other countries as well as displacement of fertilizer use both into and away from US soils. The US imported 5% of the animal products and 10% of the crop products in the food supply in 2001 (Jerardo 2003), yet our results show an opposing pattern in terms of P fertilizer use abroad to produce US imports (11% of total P fertilizer used for animal products and 6% for crop products). The P fertilizer used to produce these imports was associated with P surpluses abroad largely because more P-intensive products were imported (e.g., cattle and pigs). However, the actual quantity of P fertilizer used was much greater in the US due to concentration of crop production for export (e.g., for feed going to China and Japan), with highly uneven implications for domestic P balances due to variability in crop PUEs (Fig. 4).

Continued globalization of agriculture systems, changing consumption patterns, and concentration of phosphate rock reserves (Grote *et al.* 2005; Bouwman *et al.* 2011) mean that trade will be increasingly important for managing the modern P cycle. Schipanski and Bennett (2012) found that trade has the potential to increase P-use efficiency globally as countries tend to import crops from trading partners with higher PUE. However, exporting large amounts of crops, as in the case of the US, exacerbates the difficulty to recycle P on farms nationally, while contributing to P losses in importing countries due limited ability to recycle manure P from imported feed (Schipanski & Bennett 2012) or waste from human populations (Senthilkumar *et al.* 2012). On the other hand, direct export of P fertilizers from the US bypasses domestic agriculture altogether and limits the possibility to recycle this P. Accounting for both the production- and consumption-side responsibility for P fertilizer use via trade, such as discussed for embodied carbon emission offsets (Zaks *et al.* 2009; Davis & Caldeira 2010), could have relevance for more integrated and equitable assessment of P fertilizer use globally.

3.5. Conclusions

More effective distribution of P use for major crops on the national scale and greater recycling of all agricultural wastes will be important for utilizing remaining US phosphate rock reserves as efficiently as possible while also maintaining export-oriented agriculture. Practical challenges that exist for recycling manure and other waste (Kellogg *et al.* 2000; Bateman *et al.* 2011b) may become viable as new technologies are embraced in response to depletion of domestic phosphate rock reserves (Rittmann *et al.* 2011). Our quantitative scenarios suggest that altering domestic farm management and consumption patterns could help offset future increases in P fertilizer use due to expanding livestock and biofuel production. Trade also has important implications for the lifespan of US phosphate reserves given that the US acts as both a key P fertilizer and crop exporter. As global economic integration increases and domestic P supplies become scarcer, greater attention to full-system accounting of mineral P use may be required.

3.6. Acknowledgements

We thank Geneviève Metson and Meagan Schipanski for helpful comments and discussions on various aspects of this research. The University of Wisconsin-Madison Center for Limnology hosted GKM as a visiting researcher during the early stages of this work. Funding was provided by the Natural Sciences and Engineering Research Council of Canada (NSERC) and the Fonds québécois de la recherche sur la nature et les technologies (FQRNT) to EMB and GKM and National Science Foundation (NSF) grants (NTL-LTER and WSC) to SRC.

3.7. REFERENCES

ASAE (2003) American Society of Agricultural and Engineers standards: Manure production and characteristics. (American Society of Agricultural and Engineers, Joseph, MI.

Bateman, A., van der Horst, D., Boardman, D., Kansal, A. & Carliell-Marquet, C. (2011) Closing the phosphorus loop in England: The spatio-temporal balance of phosphorus capture from manure versus crop demand for fertiliser. *Resources Conservation and Recycling,* **55,** 1146-1153.

Beaton JD, R. T., Halstead EH, Cowell LE (1995) Global transfers of P in fertilizer materials and agricultural commodities. *Phosphorus in the global environment: transfers, cycles and management SCOPE 54* (editor H. Tiessen). Wiley, Chichester.

Bennett, E. M. & Balvanera, P. (2007) The future of production systems in a globalized world. *Frontiers in Ecology and the Environment,* **5,** 191-198.

Bouwman, L., Goldewijk, K. K., Van Der Hoek, K. W., Beusen, A. H. W., Van Vuuren, D. P., Willems, J., Rufino, M. C. & Stehfest, E. (2011) Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period. *Proceedings of the National Academy of Sciences,* **Early Edition,** 1-6.

Childers, D. L., Corman, J., Edwards, M. & Elser, J. J. (2011) Sustainability Challenges of Phosphorus and Food: Solutions from Closing the Human Phosphorus Cycle. *Bioscience,* **61,** 117-124.

Ciais, P., Bousquet, P., Freibauer, A. & Naegler, T. (2007) Horizontal displacement of carbon associated with agriculture and its impacts on atmospheric CO2. *Global Biogeochem. Cycles,* **21,** GB2014.

Cordell, D., Drangert, J. O. & White, S. (2009) The story of phosphorus: Global food security and food for thought. *Global Environmental Change,* **19,** 292-305.

Cordell, D., Neset, T. S. S. & Prior, T. (2012) The phosphorus mass balance: identifying 'hotspots' in the food system as a roadmap to phosphorus security. *Current Opinion in Biotechnology*.

Davis, S. J. & Caldeira, K. (2010) Consumption-based accounting of CO2 emissions. *Proceedings of the National Academy of Sciences,* **107,** 5687-5692.

Dodds, W. K., Bouska, W. W., Eitzmann, J. L., Pilger, T. J., Pitts, K. L., Riley, A. J., Schloesser, J. T. & Thornbrugh, D. J. (2008) Eutrophication of U.S. Freshwaters: Analysis of Potential Economic Damages. *Environmental Science & Technology,* **43,** 12-19.

Erb, K.-H., Krausmann, F., Lucht, W. & Haberl, H. (2009) Embodied HANPP: Mapping the spatial disconnect between global biomass production and consumption. *Ecological Economics,* **69,** 328-334.

FAO (2012) FAOSTAT: FAO Statistical Databases. (Food and Agriculture Organization of the United Nations, Rome, Italy).

FAOSTAT (2011) Technical Conversion Factors for Agricultural Commodities. (Food and Agriculture Organization of the United nations, Rome, Italy)

Foley, J. A., Ramankutty, N., Brauman, K. A., Cassidy, E. S., Gerber, J. S., Johnston, M., Mueller, N. D., O'Connell, C., Ray, D. K., West, P. C., Balzer, C., Bennett, E. M., Carpenter, S. R., Hill, J., Monfreda, C., Polasky, S., Rockstrom, J., Sheehan, J., Siebert, S., Tilman, D. & Zaks, D. P. M. (2011) Solutions for a cultivated planet. *Nature,* **478,** 337-342.

Gallagher, P., Dikeman, M., Fritz, J., Wailes, E., Gauther, W. & Shapouri, H. (2006) Biomass from Crop Residues: Cost and Supply Estimates. (U.S. Department of Agriculture, Washington, DC.

Galloway, J. N., Burke, M., Bradford, G. E., Naylor, R., Falcon, W., Chapagain, A. K., Gaskell, J. C., McCullough, E., Mooney, H. A., Oleson, K. L. L., Steinfeld, H., Wassenaar, T. & Smil, V. (2007) International Trade in Meat: The Tip of the Pork Chop. *AMBIO: A Journal of the Human Environment,* **36,** 622-629.

Grassini, P., Thorburn, J., Burr, C. & Cassman, K. G. (2011) High-yield irrigated maize in the Western U.S. Corn Belt: I. On-farm yield, yield potential, and impact of agronomic practices. *Field Crops Research,* **120,** 142-150.

Grote, U., Craswell, E. & Vlek, P. (2005) Nutrient flows in international trade: Ecology and policy issues. *Environmental Science & Policy,* **8,** 439-451.

Gustavsson, J., Cederberg, C., Sonesson, U., van Otterdijk, R. & Meybeck, R. (2009) Global food losses and food waste: causes and prevention. (eds. Food and Agriculture Organization of the United Nations, Rome, Italy.

Heffer, P. (2009) Assessment of Fertilizer Use by Crop at the Global Level 2006/07 – 2007/08. International Fertilizers Industry Association, Paris, France, pp. 1-12.

Hein, L. & Leemans, R. (2012) The Impact of First-Generation Biofuels on the Depletion of the Global Phosphorus Reserve. *AMBIO: A Journal of the Human Environment***,** 1-9.

Hill, J., Nelson, E., Tilman, D., Polasky, S. & Tiffany, D. (2006) Environmental, economic, and energetic costs and benefits of biodiesel and ethanol biofuels. *Proceedings of the National Academy of Sciences,* **103,** 11206-11210.

Hoekstra, A. Y. & Mekonnen, M. M. (2012) The water footprint of humanity. *Proceedings of the National Academy of Sciences,* **109,** 3232–3237.

Hoffman, L. & Baker, A. (2011) Estimating the Substitution of Distillers' Grains for Corn and Soybean Meal in the U.S. Feed Complex. (US Department of Agriculture, Economic Research Service.

IFA/IFDC/IPI/PPI/FAO (2002) Fertilizer Use by Crops. *(*International Fertilizer Industry Association, Rome, Italy*),* **5th Ed.**

Jacobson, L. M., David, M. B. & Drinkwater, L. E. (2011) A Spatial Analysis of Phosphorus in the Mississippi River Basin. *J. Environ. Qual.,* **40,** 931-941.

Jasinski, S. (2012a) 2010 Minerals Yearbook: Phosphate Rock [Advance Release] (US Geological Survey.

Jasinski, S. (2012b) Phosphate rock: Mineral commodity summaries (2011). (US Geological Survey, Reston, VA)

Jerardo, A. (2003) Import Share of U.S. Food Consumption Stable at 11 Percent. (pp. 6. USDA Economic Research Service, Washington, DC)

Kellogg, R. L., Lander, C. H., Moffitt, D. C. & Gollehon, N. (2000) Manure Nutrients Relative to the Capacity of Cropland and Pastureland to Assimilate Nutrients: Spatial and Temporal Trends for the United States. (US Department of Agriculture, Natural Resources Conservation Service/Economic Research Service, Washington, DC)

Kleinman, P. J. A., Sharpley, A. N., McDowell, R. W., Flaten, D. N., Buda, A. R., Tao, L., Bergstrom, L. & Zhu, Q. (2011) Managing agricultural phosphorus for water quality protection: principles for progress. *Plant and Soil,* **349,** 169-182.

Krausmann, F., Erb, K.-H., Gingrich, S., Lauk, C. & Haberl, H. (2008) Global patterns of socioeconomic biomass flows in the year 2000: A comprehensive assessment of supply, consumption and constraints. *Ecological Economics,* **65,** 471-487.

Lander, C. H., Moffitt, D. C. & Alt, K. (1998) Nutrients Available from Livestock Manure Relative to Crop Growth Requirements. (U.S. Department of Agriculture NRCS, Wasington, DC)

Macdonald, J. M., Ribaudo, M. O., Livingston, M. J., Beckman, J. & Huang, W. (2009) Manure Use for Fertilizer and for Energy: Report to Congress. (pp. 53. US Department of Agriculture, Economic Research Service, Washington, DC)

Maguire, R. O., Crouse, D. A. & Hodges, S. C. (2007) Diet Modification to Reduce Phosphorus Surpluses: A Mass Balance Approach. *J. Environ. Qual.,* **36,** 1235-1240.

Matsubae, K., Kajiyama, J., Hiraki, T. & Nagasaka, T. (2011) Virtual phosphorus ore requirement of Japanese economy. *Chemosphere,* **84,** 767-772.

Milbrandt, A. (2005) A Geographic Perspective on the Current Biomass Resource Availability in the United States. (National Renewable Energy Laboratory, Golden, CO.

Naylor, R., Steinfeld, H., Falcon, W., Galloway, J., Smil, V., Bradford, E., Alder, J. & Mooney, H. (2005) Losing the Links Between Livestock and Land. *Science,* **310,** 1621-1622.

NRC (2011) Renewable Fuel Standard: Potential Economic and Environmental Effects of U.S. Biofuel Policy. (US National Research Council, The National Academies Press, Washington, DC.

OECD (2008) Environmental Performance of Agriculture in OECD countries since 1990: 2. Nutrients (nitrogen and phosphorus balances). (pp. 575. Organisation for Economic Co-operation and Development, Paris, France)

PotashCorp (2006) 2006 overview of Potash Corp and Its Industry. (PotashCorp, Northbrook, IL)

Rittmann, B. E., Mayer, B., Westerhoff, P. & Edwards, M. (2011) Capturing the lost phosphorus. *Chemosphere,* **84,** 846-853.

Sattari, S. Z., Bouwman, A. F., Giller, K. E. & van Ittersum, M. K. (2012) Residual soil phosphorus as the missing piece in the global phosphorus crisis puzzle. *Proceedings of the National Academy of Sciences,* **109,** 6348-6353.

Schipanski, M. & Bennett, E. (2012) The Influence of Agricultural Trade and Livestock Production on the Global Phosphorus Cycle. *Ecosystems,* **15,** 256-268.

Schussler, J., Baker, L. A. & Chester-Jones, H. (2007) Whole-system phosphorus balances as a practical tool for lake management. *Ecological Engineering,* **29,** 294-304.

Senthilkumar, K., Nesme, T., Mollier, A. & Pellerin, S. (2012) Conceptual design and quantification of phosphorus flows and balances at the country scale: The case of France. *Global Biogeochem. Cycles,* **26,** GB2008.

Sharpley, A. N., T. Daniel, T. Sims, J. Lemunyon, R. Stevens, and R. Parry. (2003) Agricultural Phosphorus and Eutrophication, 2nd ed. U.S. Department of Agriculture, Agricultural Research Service, pp. 4.

Simpson, T. W., Sharpley, A. N., Howarth, R. W., Paerl, H. W. & Mankin, K. R. (2008) The New Gold Rush: Fueling Ethanol Production while Protecting Water Quality. *J. Environ. Qual.,* **37,** 318-324.

Smil, V. (2000) Phosphorus in the environment: Natural flows and human interferences. *Annual Review of Energy and the Environment,* **25,** 53-88.

Statistics Canada (2003) Livestock Feed Requirements Study: Canada and provinces 1999, 2000 and 2001. (Statistics Canada, Agriculture Division, Ottawa, Canada)

Suh, S. & Yee, S. (2011) Phosphorus use-efficiency of agriculture and food system in the US. *Chemosphere,* **84,** 806-813.

Tilman, D., Cassman, K. G., Matson, P. A., Naylor, R. & Polasky, S. (2002) Agricultural sustainability and intensive production practices. *Nature,* **418,** 671-677.

USDA-ERS (2010) Fertilizer Use and Price. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2011a) Feed Grains Database. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2011b) Food Availability (Per Capita) Data System. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2011c) USDA Agricultural Projections to 2020. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2012a) Fertilizer Imports/Exports. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2012b) Livestock, Dairy, and Poultry Outlook: Tables. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-FAS (2012a) Livestock and Meat Trade Data. (United States Department of Agriculture, Foreign Agricultural Service, Washington, DC)
USDA-FAS (2012b) Production, Supply, and Distribution Database. (United States Department of Agriculture, Foreign Agricultural Service, Washington, DC)
USDA-NASS (2008a) Cattle and Calf Inventory for 2007. (United States Department of Agriculture National Agricultural Statistics Service, Washington, DC)
USDA-NASS (2008b) Livestock Slaughter (summary tables). (United States Department of Agriculture National Agricultural Statistics Service, Washington, DC)
USDA-NASS (2009) US Census of Agriculture: 2007 Census Report. (United States Department of Agriculture National Agricultural Statistics Service, Washington, DC)
USDA-NRCS (2009) Crop Nutrient Tool: Nutrient Content of Crops. (United States Department of Agriculture, Natural Resource Conservation Service, Washington, DC)
Vaccari, D. A. (2009) Phosphorus: A Looming Crisis. *Scientific American,* **300,** 54-59.
Van Vuuren, D. P., Bouwman, A. F. & Beusen, A. H. W. (2010) Phosphorus demand for the 1970-2100 period: A scenario analysis of resource depletion. *Global Environmental Change,* **20,** 428-439.
Wang, F., Sims, J. T., Ma, L., Ma, W., Dou, Z. & Zhang, F. (2011) The Phosphorus Footprint of China's Food Chain: Implications for Food Security, Natural Resource Management, and Environmental Quality. *J. Environ. Qual.,* **40,** 1081-1089.
Zaks, D. P. M., Barford, C. C., Ramankutty, N. & Foley, J. A. (2009) Producer and consumer responsibility for greenhouse gas emissions from agricultural production—a perspective from the Brazilian Amazon. *Environmental Research Letters,* **4,** 044010.

3.8. TABLES AND FIGURES

Table 1. Hypothetical scenarios illustrating the effects of different changes on overall P fertilizer applications to food and feed crops consumed in US diets in 2007. The fertilizer use changes (+/- Gg P) include those embodied in imports as well as US domestic use, but exclude the fraction of P fertilizer applied to US exports. The total mineral P fertilizer ultimately recovered in diets (kg P fertilizer used / kg P consumed in food) is indicated for the 2007 baseline and each alternative scenario holding all other variables constant.

Scenario	Food crop P fertilizer applied	Feed crop P fertilizer applied	P fertilizer consumed in food (kg P kg^{-1})	Description
2007 baseline	489	897	9.4	
Scenario 1 (S1) – Balanced P nationally	-146	-173	7.2	National P fertilizer application is reduced for crops with overall surpluses and increased for crops with deficits until all crops have net zero P balances; assumption of no change in manure P.
Scenario 2 (S2) – Complete recycling of manure	-28	-124	8.6	100% capture of manure on farms and no losses during transportation. Additional manure P is applied to feed and food crops in proportion with 2007 applications, with an equal reduction in P fertilizer use based on assumption of equivalent P-fertilizer value to inorganic fertilizers.
Scenario 3 (S3) – Substantial reduction in food waste	-53	-225	7.5	Total per capita demand for food is reduced via 20% reduction in food crop waste (no change in kilo-calories consumed) and 32% reduction in livestock product waste (no change in protein consumed). These are common USDA consumer-level food loss estimates for grains and meats.
Scenario 4 (S4) – More poultry, less beef	No change	-65	9.0	20% of beef protein is replaced by poultry protein (Donner 2007). Change in mineral P feed supplements not included (~14 Gg net P increase needed for poultry).
Scenario 5 (S5) – Lacto-ovo diets	+126	-742	5.2	All meat consumption replaced with protein from mix of grain, dairy, eggs, legumes/nuts, fruits and vegetables; protein replacement is proportional to current per capita protein sources (USDA) from these foods. Accounts for conservative life-cycle fertilizer use for beef, but assumes no net P fertilizer loss to food crops due to a decrease in manure applications of ~520 Gg P yr^{-1}.

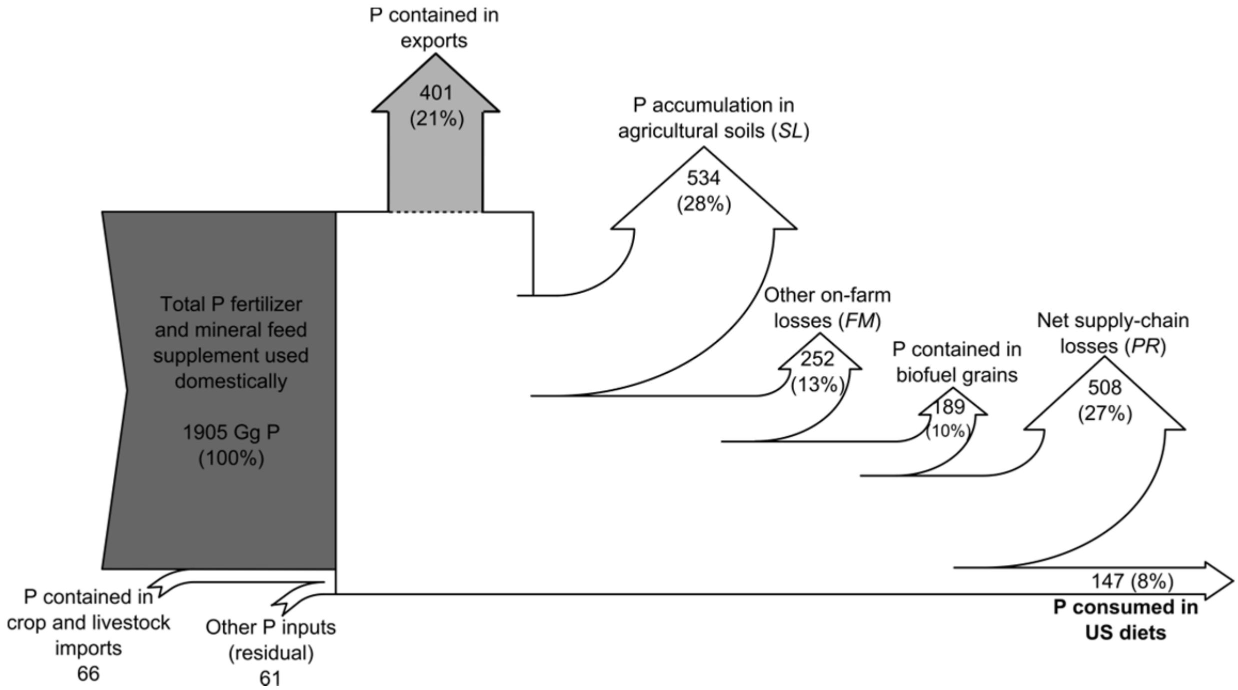


Fig. 1. The fate of domestic mineral P inputs (fertilizer and feed supplements) in the US agricultural system in 2007. All flows are in Gg of P and approximate percent of total mineral P inputs for the US domestically in 2007.

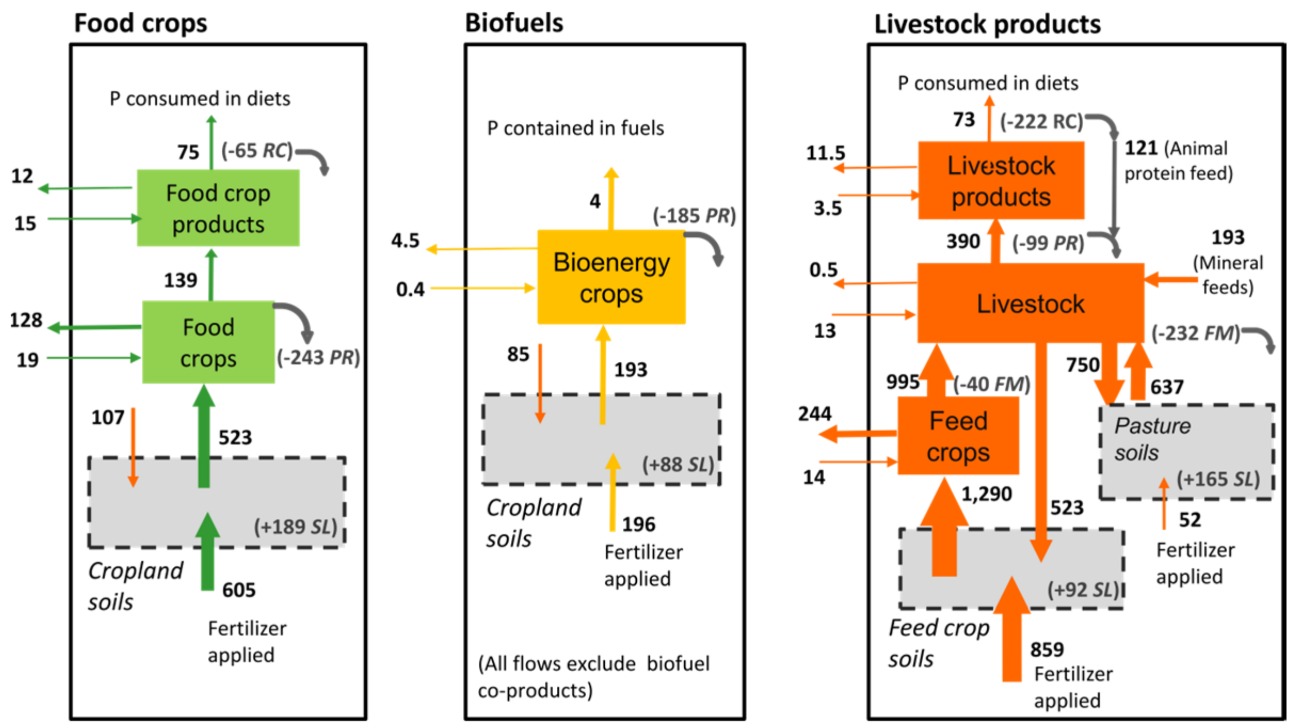


Fig. 2. Breakdown of major P flows (Gg of P) associated with domestic production and consumption of food, biofuels (excluding co-product uses), and livestock products in the US in 2007. Arrows crossing the solid rectangles represent the P contained in foreign trade. Apparent losses at each stage include soil balances (*SL*), other farm losses (*FM*), apparent processing losses (*PR*), and retail to consumer food loss estimates (*RC*). Feed crop P removal from soils includes estimates of crop residues used as livestock forage (~60 Gg P) (following Gallagher *et al.* 2006 removal estimates) and ethanol co-product distillers grains fed to livestock (~105 Gg P) (following Hoffman & Baker 2011).

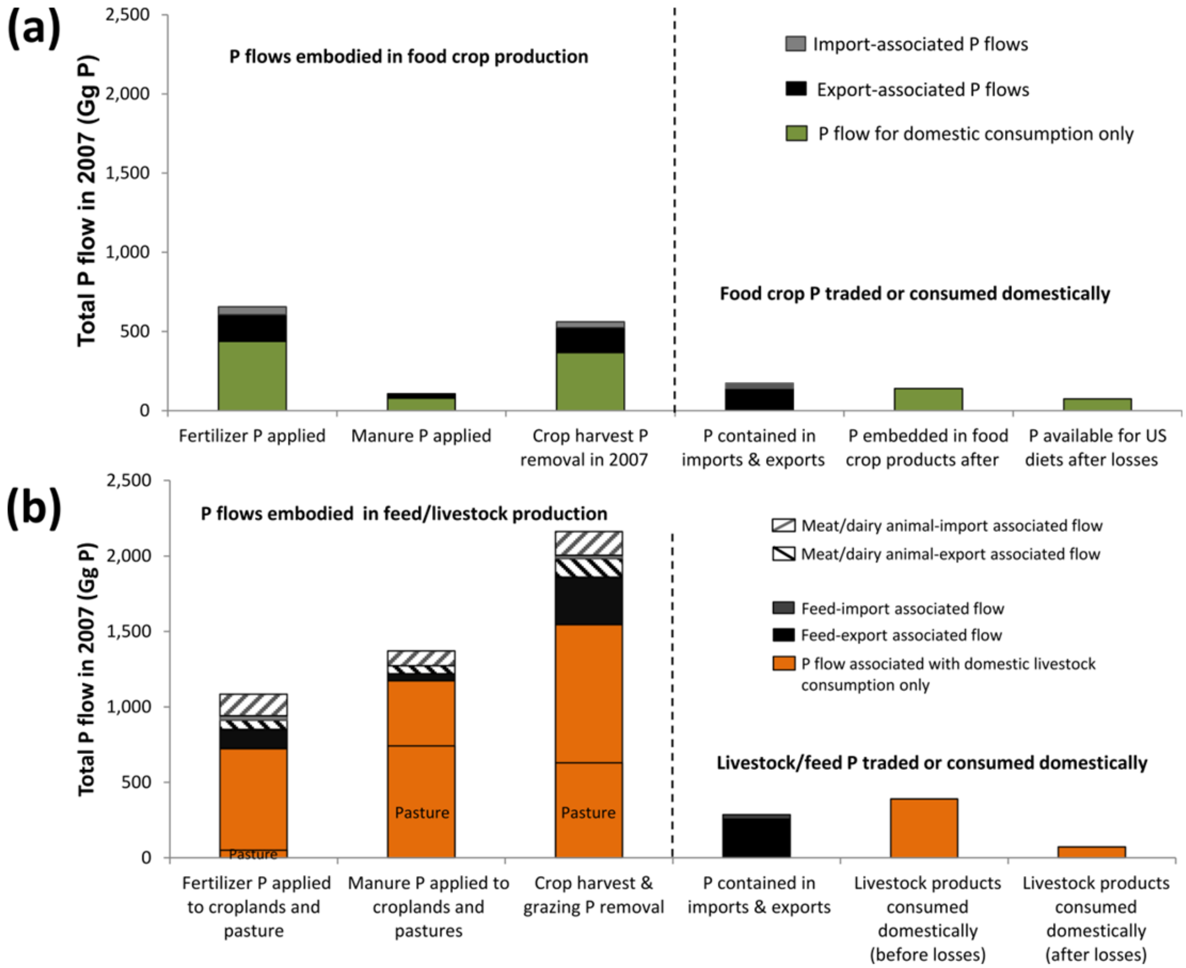


Fig. 3. Comparison of the P flows associated with (i.e., 'embodied' in) domestic production, consumption, and trade of food crop (a) and livestock commodities (b) in the US agricultural system in 2007. The left side of each plot shows the P flows associated with producing commodities and the right side shows the P contained in imports and exports or in domestically consumed foods. All flows for US domestic production and consumption are shown in green and orange for food crops and livestock commodities, respectively. P flows associated with imports and exports are shown in gray and black boxes, respectively.

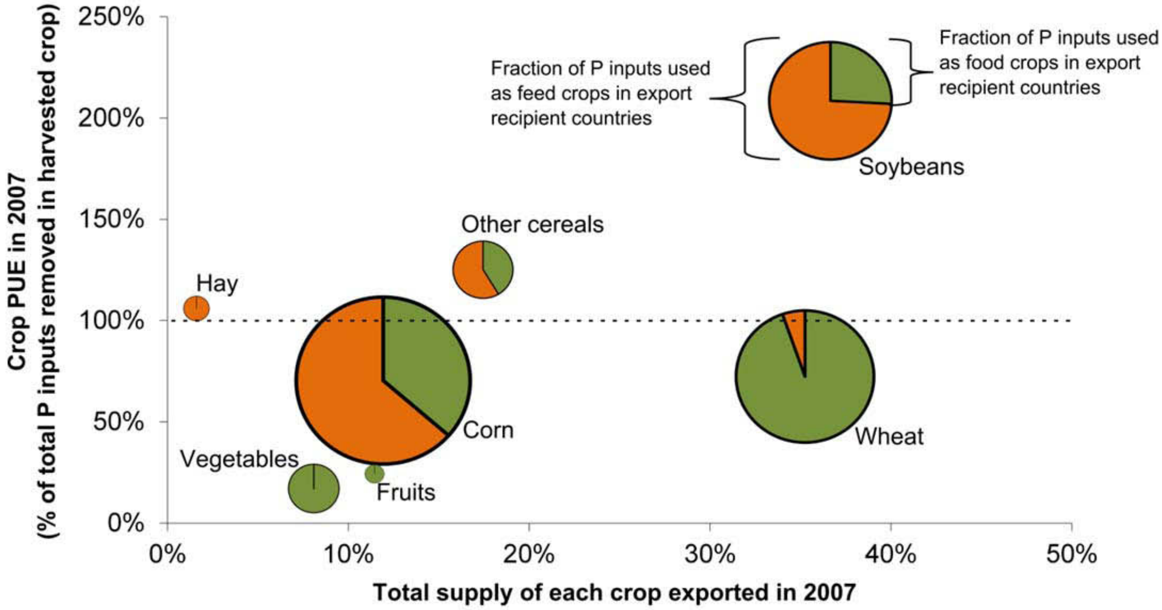


Fig. 4. Soil phosphorus-use efficiencies (PUEs) for food and feed crops produced in the US relative to the total supply of each crop exported in 2007. PUE is expressed as the percent of total P fertilizer and manure applied to exported crops that was removed from cropland soils during crop harvest. The size of the bubbles indicates the relative amount of P fertilizer and manure applied to each exported crop, with colors representing the fraction of each crop used for food or feed in major destination countries. Note that including embodied feed use for livestock exports increases the export fractions from 5-10% depending on the crop.

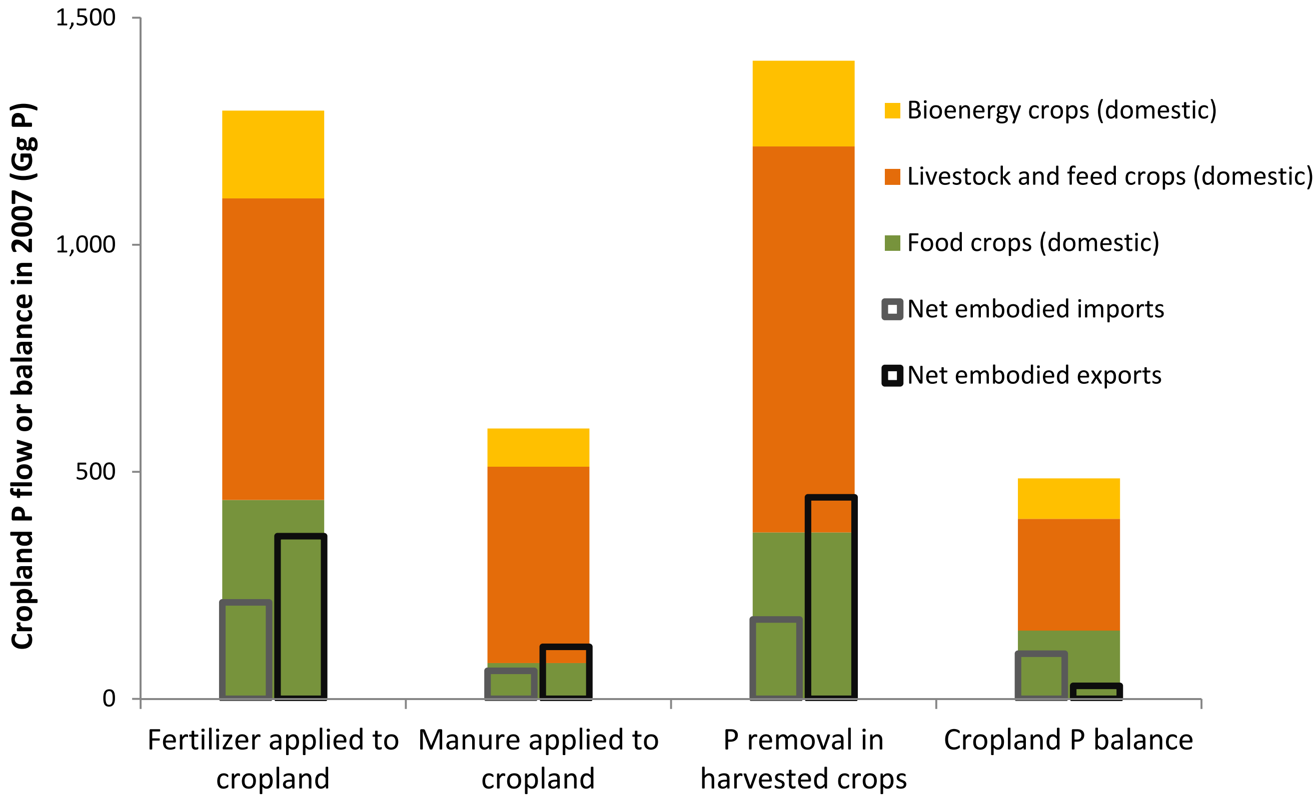


Fig. 5. Total P inputs and outputs to cropland soils associated with production of food, feed, and biofuel crops that were produced and consumed domestically in the United States in 2007 (green, orange, and yellow boxes, respectively), relative to those associated with overall imports (hollow gray boxes) and exports (hollow black boxes). See Table S2 for detailed breakdowns of the embodied P balance components.

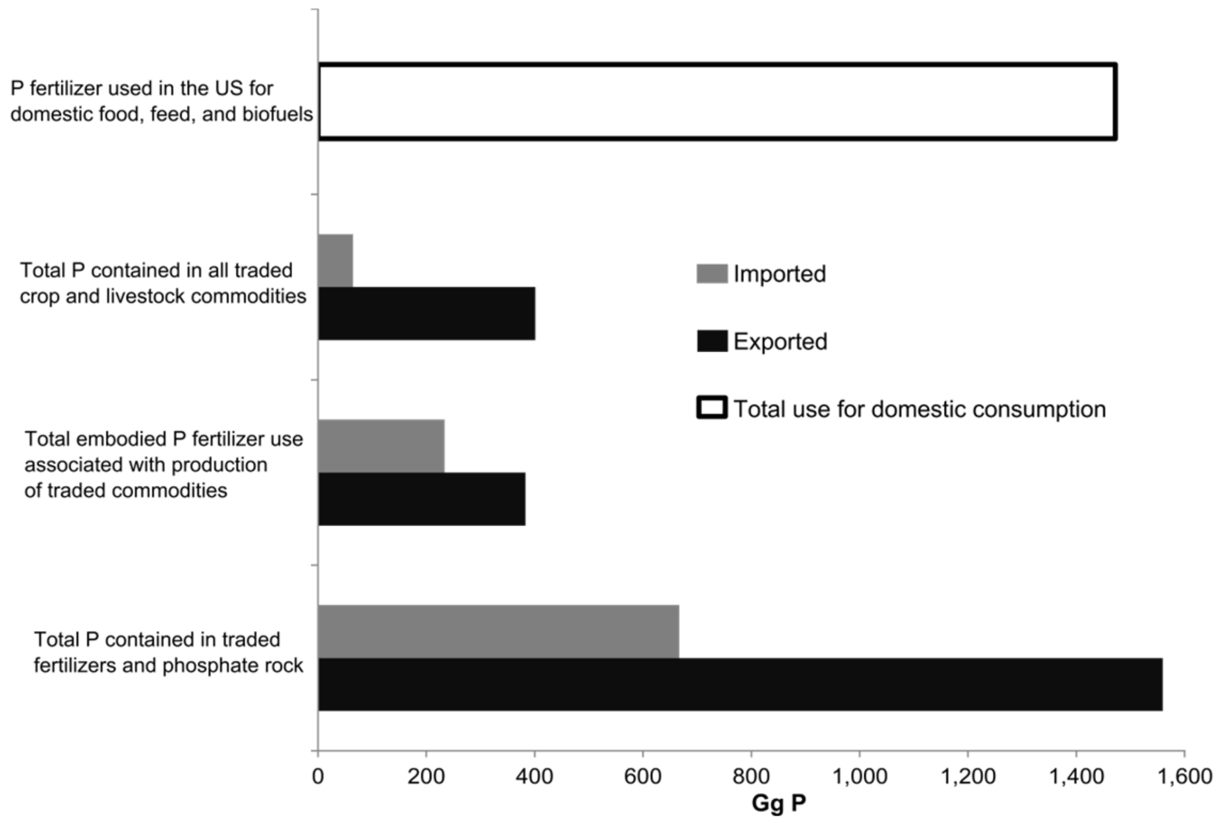


Fig. 6. Comparison of the overall magnitude of P flows associated with different components of international trade for the US agricultural system in 2007. The embodied P and domestic P fertilizer use totals include the contribution of mineral P supplements fed to livestock.

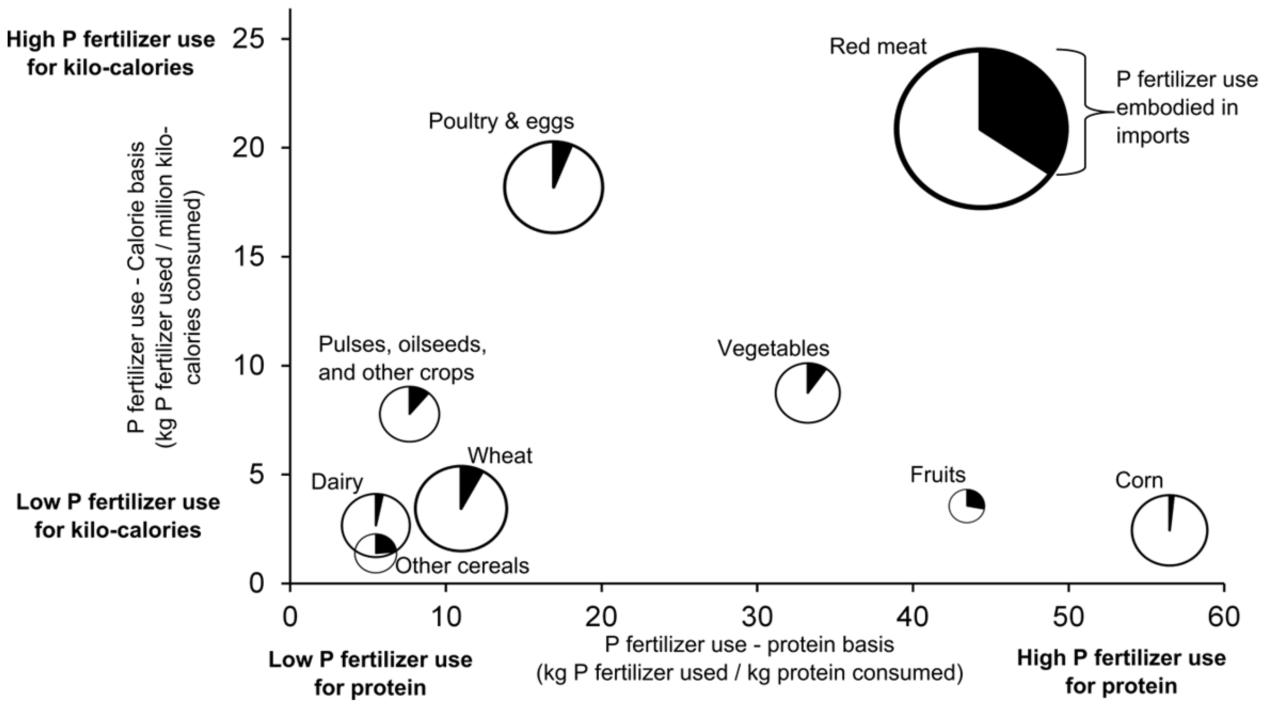


Fig. 7. Fertilizer P use associated with different foods consumed in the US in 2007 expressed in simple per unit nutritional returns. The relative share of the total P fertilizer from imports is shaded in grey (including life-cycle estimates for cattle and other ruminants). The size of the bubbles indicates the total P fertilizer used to produce each commodity. See Table S3 for P fertilizer use expressed in terms of the P eventually consumed in human diets.

3.8. SUPPORTING INFORMATION TEXT

Overview of primary data sources

Our model is primarily based on crop and livestock production data from the US Census of Agriculture for 2007 (USDA-NASS 2009), as well as data for some minor crops from FAO (2012). We calculated the percentage of total supply (sum of production, beginning stocks, and imports) for each crop that was used for food, feed, biofuel, seed or other purposes. We derived supply and use data from USDA 'Supply and Disappearance' tables for 47 crops (USDA-ERS 2011b; USDA-ERS 2011a; USDA-ERS 2012a), as well as from FAO 'Food Balance Sheets' (FAO 2012) for cotton and 4 minor crops. All P flows associated with crops used for seed or industrial purposes were excluded from our study (typically <5% except for cotton). Biofuel crops were limited to corn ethanol and soybean biodiesel (~95% of total biofuel use in 2007) (USDA-ERS 2012b). These percentages were subsequently applied to all Census of Agriculture data to allocate crop production and all associated P flows (fertilizer use, manure use, and crop harvest P removal) to different uses. Additionally, the percentage of the total supply of each crop that was imported or exported could be accounted for using these numbers. Crops imported to the US were assumed to be used in proportion to the same uses as crops that were produced in the US. Similarly, we included year-end domestic crop supplies in our annual use estimates by dividing them proportionally among each major use (food, feed, or biofuel) in 2007.

We distinguished between P flows associated with either whole crops or live animals and those associated with major processed crop or livestock products. The P contents of different products were compiled from various sources (NRC 1996; USDA-NRCS 2009; MacDonald *et al.* 2011; USDA-ARS 2011; IFA 2012; Schipanski & Bennett 2012) (for fuel ethanol, we assumed the same P content as food-grade ethyl alcohol). In cases where crop products were not reported on an equivalent whole crop or farm basis (e.g., 'soybean meal' instead of soybean, or 'orange juice' instead of oranges), we used conversion factors (USDA-ARS 1992; FAOSTAT 2011; USDA-ERS 2011b) to adjust between whole-crop and product weights if necessary. These product fractions also allowed us to account for the fractions of certain crops used for specific purposes. For example, we excluded the estimated portion of total cotton supply (47%) used for textile/industrial purposes from all P flows based on the fraction of seed cotton weight that becomes 'linters' (FAOSTAT 2011). Product fractions were also used to estimate P flows associated with co-products of some crops that had different uses (e.g., soybean meal for feed and soybean oil for food are both derived from soybean crushing). Our model therefore accounts for major processed food and biofuel uses of corn (excluding biofuel co-products, which are predominantly grouped with residual feed use by the USDA (USDA-ERS 2011a)), soybean oil uses for food or biodiesel (i.e., methyl esters) and soybean meal (assuming that 98% was used for feed (Donner 2007)). For fruit and vegetables, we were able to distinguish between major fresh and processed crop uses (e.g., fresh oranges and oranges processed as orange juice) for imports, exports and domestic food consumption using USDA data (USDA-ERS 2011b).

As with crop products, we had to translate some raw data for livestock between live animal, carcass-equivalent, and meat weights (USDA-NASS 2008b; FAOSTAT 2011; USDA-FAS 2012b). To estimate the live animals associated with beef, pork, chicken, turkey and lamb, we used conversion factors for each type of meat to adjust the mass of meat products first to a dressed carcass weight-equivalence and then to live animal weights.

Estimation of US fertilizer use

The USDA reports total P fertilizer use in the US, but only provides annual breakdowns for 4 major crops (corn, soybean, cotton, and wheat). As a result, we used a disaggregation approach by taking total P fertilizer applied to groups of crops (e.g., ‘fruits and vegetables’) reported by IFA (Heffer 2009) for 2006/2007 and used this to estimate the relative application rate for each crop. This estimate was based on the fraction of the crop group total that was used for each individual crop in 1998, the most recent year with detailed crop-specific national P fertilizer data for the US (IFA/IFDC/IPI/PPI/FAO 2002). Additionally, the USDA (USDA-ERS 2010) P fertilizer use data for corn is reported for grain corn only, whereas corn used for silage in the US is a sizeable fraction of total cropland area (USDA-NASS 2009). We therefore estimated P fertilizer use for corn silage using the same application rate per harvested area as corn (adjusted for 81% of area fertilized)(USDA-ERS 2010), in conjunction with Census of Agriculture production data. P fertilizer application to hay crops is also unrecorded at the national level. We therefore included estimates of P fertilizer application to hay based on areas planted to alfalfa or grass hay in 2007 (USDA-NASS 2009) and assumed the same P application rates as from a USDA national nutrient management model (EPIC) (USDA-NRCS 2006). No estimates of P fertilizer application to pasture were found after a detailed internet search, so we approximated this using the area of pastureland that was reported to be amended with commercial fertilizers in the agricultural census (USDA-NASS 2009), and assuming the same P application rate as that of 'grass hay'.

Estimation of US livestock P flows

We compiled livestock inventory data for 2007 from the US Census of Agriculture. Additional breakdowns were made to group animals into classes for either 'breeding', 'milk/eggs', or 'meat', relying on more detailed inventory data for cattle as well as our own interpretations of what animals comprised certain census categories. We differentiated between feedlot and grazing cattle based on detailed inventory data for 2007 (USDA-NASS 2008a). In total, we estimated P flows for 5 classes of cows (beef fattening-stage, beef non-fattening stage, dairy, heifers, and other), 2 pig (breeding and other pigs), and 4 poultry (layers, broilers, pullet chicks, ducks, and turkeys), as well as goats, sheep, and horses. We then used annual P excretion factors (adjusted for multiple annual cycles of pigs and chickens) from Maguire et al. (2007), with a simple adjustment for beef cattle using P excretion factors from Kellogg et al. (2000). For sheep, horses, goats, and ducks, we obtained P excretion factors from ASAE (2003). Average recoverability of manure P for cropland application was taken from Kellogg et al. (2000) for all states and for each of the major livestock classes (assuming beef recoverability for sheep, goats, and horses). A 15% loss estimate was also made to account for potential collection, transportation, and runoff losses of recovered manure P on farms (Kellogg *et al.* 2000).

To estimate subsequent manure application to specific crops, we used 2003 USDA national farm survey data (Macdonald *et al.* 2009). Because some crops with large sub-classes were grouped together (e.g., corn for grain and corn for silage), we used the fraction of total crop P removal from each sub-class to allocate manure to that particular crop sub-class. A similar approach was used to estimate P fertilizer use for different hay crops (e.g., small grain hay versus hay used for greenchop).

To estimate other non-feed sources of P intake by livestock, we used industry data on mineral P feed supplements used for poultry, pigs, and cattle in the US (PotashCorp 2006) and USDA data for animal protein feeds (i.e., by-products of livestock processing), each multiplied by their typical P contents (NRC 1996; IFA 2012). Our mass-balance method for estimating P grazing off-take gave results comparable in magnitude to other national estimates for the US (Kellogg *et al.* 2000; OECD 2008).

Calculating quantities of crops traded and P contained in traded goods

Our calculations of P directly contained in traded commodities were based on product-specific P contents and the quantities of major unprocessed, semi-processed, or highly-processed crop production traded. The summary data used to calculate import and export quantities for major crops (USDA-ERS 2011a) do not include some major processed forms of traded crops. Accordingly, we incorporated imports and exports of key processed food and feed crop products (for corn, soybean, oats, barley, canola, peanut, wheat, grapes, and alfalfa) into the traded fractions using supplemental data (USDA-ERS 2011a; USDA-FAS 2012a). In the case of imported and exported live animals, we used average slaughter weights and P contents for US live animal exports, and for OECD countries for US live animal imports (USDA-NASS 2008b; FAOSTAT 2011).

It was also necessary to account for the uses of crops exported from the US in destination countries. To do so, we applied FAO 'Supply and Utilization' of data (FAO 2012) to the exports, weighted according to the fraction of exports for each crop going to each major destination country (with ≥5% of the total exports for each crop). However, we assumed that all processed fruits and vegetables were used for food, that soybean cake was used for feed, and that 'other utilization' of soybean oil and corn grain reported by FAO (FAO 2012) was indicative of biofuel use (this was always <5% of uses for all major destination countries).

Overview of P flows associated with US imports

In order to calculate P fertilizer use embodied in the production of imported crops and the crops consumed by imported livestock, we used crop-specific P fertilizer application rate data from IFA (IFA/IFDC/IPI/PPI/FAO 2002). A detailed overview of the basic assumptions and methods for estimating P rates for certain countries is given in Potter et al. (Potter *et al.* 2010). For some crops in some of the import source countries, P fertilizer rate data was unavailable. We therefore used world or major trading partner average P fertilizer rates (particularly for import source countries in Asia or South America) or US rates (particularly for imports from Canada). However, no manure P application data was available for application to any imported crops.

We followed a method similar to that described for the US to calculate manure P produced by imported livestock and the application of this manure to agricultural soils. However, we accounted for regional variation in livestock production and manure management. We calculated manure P production using average manure factors and live weights for OECD countries (OECD 2008; FAOSTAT 2011) to account for some of these broad differences. To estimate manure application to either cropland or pasture embodied in cattle imports, we followed the basic approach of Schipanski & Bennett (2012). For example, we assumed that P from grazed animals was deposited on pastures only, and that grain- or other forage-fed animals were produced in mixed/landless systems where approximately 50% of manure from cattle and ruminants in developed and 75% application for transitioning countries would be available for cropland application (*sensu* Bouwman *et al.* (2005)). For imported pigs and poultry, we used US recoverable manure P fractions and farm-level losses, reflecting the fact that most pig imports were from Canada. In order to calculate P removal by pasture-grazed animals, we assumed that each hectare of pasture grazed in order to provide P intake was associated with ~12 kg P ha^{-1} yr^{-1} of P uptake from soils (Kellogg *et al.* 2000).

Calculating US domestic consumption of food, processing losses, and P footprints

All estimates of food availability were derived from USDA food use estimates for each major food crop (USDA-ERS 2011b). We then adjusted the total food availability to account for imports, exports, and non-food uses (e.g., feed, seed, industrial or biofuels). For dairy and eggs, we excluded small fractions used for agricultural feeding and hatching from the total available for human use (USDA-ERS 2011b), but otherwise assumed that all domestic dairy and egg production (adjusted for traded quantities) was available for consumption in the US in 2007.

Apparent processing losses of food crops was primarily due to changes in the weights and P contents from farm to product levels for each crop (e.g., from oranges harvested in orchard to processed orange juice). Although some food crops losses may be used as by-product feed for livestock (e.g., citrus pulp, molasses, and sugar beets) (Ash 1993), this is not tracked by USDA. Similarly, apparent processing losses of livestock during slaughter were estimated as the difference between average national weights at slaughter and typical processed weights and P contents, using the factors described above to convert between live animal and meat weights. Different P contents were used to reflect the P content at each stage of livestock processing; for example, live animals contain considerably more high-P content materials such as bone than boneless meats (USDA-ARS 2011).

For the calculation of P fertilizer used for livestock produced and consumed in the US, we included estimates of P flows for younger cows, sheep, and goats that were raised but not slaughtered in 2007. For imports, we multiplied the P fertilizer requirements by a factor of 2.3 for cows (the average lifespans of beef, heifers and calves) and 1.75 (average for sheep and goats) to better reflect the lifespan of different animals and different stages of red meat production (Chapagain & Hoekstra 2004). Although these are conservative estimates of the actual lifespans of cows, we used these to ensure greater comparability with US domestic P flows for these red meats, which have considerably different feed and pasture diet compositions. However, beef cows in industrial systems may be reared for as long as 36 months before being slaughtered (Chapagain & Hoekstra 2004).

SI References

ASAE (2003) American Society of Agricultural and Engineers standards: Manure production and characteristics. (American Society of Agricultural and Engineers, Joseph, MI).

Ash, M. (1993) Animal Feeds Compendium. (U.S. Department of Agriculture, Economic Research Service, Washington DC).

Bouwman, A. F., Van der Hoek, K. W., Eickhout, B. & Soenario, I. (2005) Exploring changes in world ruminant production systems. *Agricultural Systems,* **84,** 121-153.

Chapagain, A. K. & Hoekstra, A. Y. (2004) Water footprints of nations, value of water research Report Series No. 16. UNESCO-IHE, Delft, The Netherlands.

Donner, S. D. (2007) Surf or turf: A shift from feed to food cultivation could reduce nutrient flux to the Gulf of Mexico. *Global Environmental Change-Human and Policy Dimensions,* **17,** 105-113.

FAO (2012) FAOSTAT: FAO Statistical Databases. (Food and Agriculture Organization of the United Nations, Rome, Italy)

FAOSTAT (2011) Technical Conversion Factors for Agricultural Commodities. (Food and Agriculture Organization of the United nations, Rome, Italy)

Heffer, P. (2009) Assessment of Fertilizer Use by Crop at the Global Level 2006/07 – 2007/08. (pp. 1-12. International Fertilizers Industry Association, Paris, France)

IFA (2012) Conversion factors - Nutrients as % of product. (International Fertilizer Industry Association, Paris, France).

IFA/IFDC/IPI/PPI/FAO (2002) Fertilizer Use by Crops. (International Fertilizer Industry Association, Rome, Italy), **5th Ed.**

Kellogg, R. L., Lander, C. H., Moffitt, D. C. & Gollehon, N. (2000) Manure Nutrients Relative to the Capacity of Cropland and Pastureland to Assimilate Nutrients: Spatial and Temporal Trends for the United States. (US Department of Agriculture, Natural Resources Conservation Service/Economic Research Service, Washington, DC)

MacDonald, G. K., Bennett, E. M., Potter, P. A. & Ramankutty, N. (2011) Agronomic phosphorus imbalances across the world's croplands. *Proceedings of the National Academy of Sciences,* **108,** 3086-3091.

Macdonald, J. M., Ribaudo, M. O., Livingston, M. J., Beckman, J. & Huang, W. (2009) Manure Use for Fertilizer and for Energy: Report to Congress. (US Department of Agriculture, Economic Research Service, Washington, DC), pp. 53.

Maguire, R. O., Crouse, D. A. & Hodges, S. C. (2007) Diet Modification to Reduce Phosphorus Surpluses: A Mass Balance Approach. *J. Environ. Qual.,* **36,** 1235-1240.

NRC (1996) Nutrient Requirements of Beef Cattle: Seventh Revised Edition: Update 2000. (National Research Council, The National Academies Press, Washington, DC)

OECD (2008) Environmental Performance of Agriculture in OECD countries since 1990: 2. Nutrients (nitrogen and phosphorus balances). (pp. 575. Organisation for Economic Co-operation and Development, Paris, France)

PotashCorp (2006) 2006 overview of Potash Corp and Its Industry. (Potash Corporation, Northbrook, Illinois)

Potter, P., Ramankutty, N., Bennett, E. M. & Donner, S. D. (2010) Characterizing the Spatial Patterns of Global Fertilizer Application and Manure Production. *Earth Interactions,* **14,** 1-22.

Schipanski, M. & Bennett, E. (2012) The Influence of Agricultural Trade and Livestock Production on the Global Phosphorus Cycle. *Ecosystems,* **15,** 256-268.

USDA-ARS (1992) Weights, Measures, and Conversion Factors for Agricultural Commodities and Their Products, Washington, DC)
USDA-ARS (2011) USDA National Nutrient Database for Standard Reference, Release 24. (U.S. Department of Agriculture, Agricultural Research Service, Washington, DC)
USDA-ERS (2010) Fertilizer Use and Price. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2011a) Feed Grains Database. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2011b) Food Availability (Per Capita) Data System. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2012a) Agricultural Baseline Projections: Baseline Related Historical Data. (United States Department of Agriculture, Economic Research Service, Washington, DC)
USDA-ERS (2012b) Bioenergy: Biofuel, Biofuel Feedstock, and Coproduct Market Data. (United States Depatment of Agriculture, Economic Research Service, Washington, DC)
USDA-FAS (2012a) Global Agricultural Trade System (GATS). (United States Department of Agriculture, Foreign Agriculture Service, Washington, DC)
USDA-FAS (2012b) Livestock and Meat Trade Data. (United States Department of Agriculture, Foreign Agricultural Service, Washington, DC)
USDA-NASS (2008a) Cattle and Calf Inventory for 2007. (United States Department of Agriculture National Agricultural Statistics Service, Washington, DC)
USDA-NASS (2008b) Livestock Slaughter (summary tables). (United States Department of Agriculture National Agricultural Statistics Service, Washington, DC)
USDA-NASS (2009) US Census of Agriculture: 2007 Census Report. (United States Department of Agriculture National Agricultural Statistics Service, Washington, DC)
USDA-NRCS (2006) Model Simulation of Soil Loss, Nutrient Loss, and Change in Soil Organic Carbon Associated with Crop Production. (United States Department of Agriculture, Natural Resource Conservation Service, Washington, DC)
USDA-NRCS (2009) Crop Nutrient Tool: Nutrient Content of Crops. (United States Department of Agriculture, Natural Resource Conservation Service, Washington, DC)

3.9. SUPPORTING INFORMATION TABLES

Table S1. Total P inputs and outputs (Gg of P) to US soils for the 52 crops in this study, as well as the fractions of each crop exported and used for food or feed domestically. Some crop use fractions do not add to 1.0 because of excluding seed or industrial uses (e.g., cotton). Biofuel use for corn was 21.7% of the total grain use (not shown) and <1% of total soybean use in 2007. P balances are expressed in terms of the 2007 harvested area of each crop.

Crop	Fertilizer P applied	P in harvested crop	Manure P applied	Fraction of crop exported	Fraction for food (domestic)	Fraction for feed (domestic)	P balance (kg P ha^{-1})
Alfalfa - hay	21.6	165.4	133.5	0.01	0.00	0.99	-1.2
Apples	1.1	0.4	<0.1	0.10	0.90	0.00	5.3
Apricots	<0.1	<0.1	<0.1	0.10	0.90	0.00	14.5
Artichokes	<0.1	<0.1	<0.1	0.01	0.99	0.00	14.5
Asparagus	0.2	<0.1	<0.1	0.04	0.96	0.00	8.0
Avocados	0.6	<0.1	<0.1	0.01	0.99	0.00	16.4
Barley	10.1	15.2	4.4	0.14	0.62	0.23	-0.4
Cabbages & brassicas	1.4	0.3	<0.1	0.03	0.96	0.00	38.7
Canola	23.5	3.7	<0.1	0.12	0.15	0.72	42.8
Carrots & turnips	2.6	0.5	<0.1	0.19	0.81	0.00	55.0
Cauliflowers & broccoli	1.0	0.2	<0.1	0.21	0.79	0.00	52.5
Corn - grain	901.6	874.7	392.6	0.12	0.13	0.54	12.0
Corn - silage	55.6	69.1	31.0	0.00	0.00	1.00	7.2
Cottonseed (no linters)	42.9	9.8	11.6	0.11	0.03	0.72	10.5
Dates	<0.1	<0.1	<0.1	0.13	0.79	0.00	22.5
Dry beans	4.2	6.2	0.1	0.17	0.81	0.00	-3.2
Eggplants	0.2	<0.1	<0.1	0.08	0.92	0.00	70.3
Garlic	0.4	0.3	<0.1	0.05	0.91	0.00	8.1
Grapefruit	0.9	0.3	<0.1	0.26	0.74	0.00	17.6
Grapes	3.8	0.8	<0.1	0.17	0.83	0.00	8.0
Hay (non-alfalfa)	11.3	74.2	59.9	0.02	0.00	0.98	-1.0
Lemons & limes	0.2	0.1	<0.1	0.23	0.77	0.00	2.8
Lentils	2.8	0.6	<0.1	0.23	0.75	0.00	18.3
Lettuce & chicory	7.8	1.2	<0.1	0.08	0.92	0.00	54.5
Oats	3.2	4.2	17.6	0.01	0.31	0.61	27.2
Olives	<0.1	0.5	<0.1	0.02	0.93	0.00	-39.4
Onions, dry	4.7	1.1	<0.1	0.06	0.87	0.00	54.8
Oranges	3.5	1.3	<0.1	0.09	0.91	0.00	7.9
Peaches & nectarines	0.6	0.2	<0.1	0.11	0.89	0.00	6.6
Peanuts	8.5	5.5	2.2	0.15	0.69	0.07	10.8
Pears	0.2	<0.1	<0.1	0.19	0.81	0.00	3.7
Peas	7.2	0.2	<0.1	0.63	0.34	0.00	20.3
Pineapples	0.2	<0.1	<0.1	0.01	0.99	0.00	34.1
Pistachios	1.7	0.9	<0.1	0.48	0.52	0.00	17.5
Potatoes	49.4	11.4	0.2	0.12	0.88	0.00	83.5
Rice	16.1	22.9	0.4	0.40	0.52	0.00	-5.7
Rye	0.3	0.5	<0.1	0.02	0.25	0.29	-2.1
Sorghum	27.8	36.2	2.0	0.30	0.13	0.57	-2.3
Soybean	160.2	425.0	45.6	0.37	0.13	0.48	-8.5
Spinach	0.7	0.1	<0.1	0.06	0.94	0.00	35.1
Strawberries	1.2	0.2	<0.1	0.11	0.89	0.00	45.6
Sugarbeets	11.8	12.4	0.2	0.04	0.94	0.00	-0.8
Sugarcane	8.3	13.3	0.2	0.04	0.94	0.00	-14.0
Sunflower seeds	3.6	7.4	0.1	0.11	0.24	0.22	-4.5
Tangerines & mandarins	0.2	<0.1	<0.1	0.04	0.96	0.00	10.4
Tomatoes	15.1	4.0	<0.1	0.06	0.94	0.00	65.4
Watermelons	4.6	0.2	<0.1	0.06	0.94	0.00	85.1
Wheat	262.3	196.9	18.4	0.35	0.57	0.06	4.1

Table S2. Cropland soil P balances (Gg of P) associated with production of US imports abroad and US exports domestically detailed for each major crop system. Total livestock flows include both direct feed crop imports and exports as well as livestock imports and exports. Feed crop exports are shown separately because of the large influence of soybean on the cropland P balance.

	Fertilizer P applied	Manure P applied	Crop P removal	Cropland surplus	Cropland deficit
Food crops – Embodied in US imports	51	—	37	14	—
Food crops – Embodied in US exports	167	28	156	39	—
Feed crops only – Embodied in exports	126	46	198	—	-26
Total livestock – Embodied in imports	158	62	136	22	—
Total livestock – Embodied in exports	188	85	283	—	-10
Biofuels – Embodied in imports	3	—	2	2	—
Biofuels – Embodied in exports	4	1	5	<1	—

Table S3. Total P fertilizer used for US food consumption (either domestically or in other countries to produce imports) as well as P fertilizer use expressed per unit of consumption based on the three metrics considered in this study.

Category	**Total P fertilizer for domestic consumption** (embodied imports + domestic production - embodied exports)	**% of total P fertilizer use from imports**	**P fertilizer recovered in diets** (kg P fertilizer / kg P consumed)	**P fertilizer use for protein** (kg P fertilizer / kg protein consumed)	**P fertilizer use for Calories** (kg P fertilizer / million kilo-calories consumed)
Other cereals	37	24	2.2	5.4	1.4
Dairy	98	4	2.0	5.3	2.7
Pulses	12	27	3.1	5.7	3.8
Nuts & sugars	29	6	4.3	5.5	0.2
Wheat	176	8	6.2	10.7	3.5
Vegetables	87	10	12.2	33.1	8.8
Oilseeds	34	10	12.4	9.9	2.4
Fruits	27	28	14.3	43.4	3.6
Corn	121	2	22.1	56.3	2.5
Poultry & eggs	203	7	20.9	16.7	18.2
Red meat	607	37	42.7	43.9	20.9

CONNECTING STATEMENT

Chapters 2 and 3 provided new insight on how farm nutrient management, agricultural trade, and demand for particular agricultural commodities act as drivers of P fertilizer and manure use that could have large-scale influences on soil P levels. It is clear that agriculture typically involves a dramatic acceleration of P cycling through soils over background levels, given the magnitudes of soil amendments that could elevate soil P or P removal via crop harvest that can gradually deplete soil P. But what happens to soil P once agriculture ceases?

In Chapter 4, I present the first quantitative synthesis addressing this question using data compiled from studies around the world. I examine evidence of past agriculture on contemporary soil P pools following land-use change away from agriculture, as well as some of the key factors that may contribute to the direction and magnitude of the 'land-use legacy' effect. This chapter closes with a discussion of what these soil P legacies could mean for long-term ecosystem management following termination of agriculture.

CHAPTER 4: THE INFLUENCE OF TIME, SOIL CHARACTERISTICS AND LAND-USE HISTORY ON SOIL PHOSPHORUS LEGACIES: A GLOBAL META-ANALYSIS

Graham K. MacDonald, Elena M. Bennett, Zofia E. Taranu

Published in *Global Change Biology*

4.0. ABSTRACT

Agriculturally-driven changes in soil phosphorus (P) are known to have persistent effects on local ecosystem structure and function, but regional patterns of soil P recovery following cessation of agriculture are less well understood. We synthesized data from 94 published studies to assess evidence of these land-use legacies throughout the world by comparing soil labile and total P content in abandoned agricultural areas to that of reference ecosystems or sites remaining in agriculture. Our meta-analysis shows that soil P content was typically elevated after abandonment compared to reference levels, but reduced compared to soils that remained under agriculture. There were more pronounced differences in the legacies of past agriculture on soil P across regions than between the types of land use practiced prior to abandonment (cropland, pasture, or forage grassland). However, consistent patterns of soil P enrichment or depletion according to soil order and types of post-agricultural vegetation suggest that these factors may mediate agricultural legacies on soil P. We also used mixed effects models to examine the role of multiple variables on soil P recovery following agriculture. Time since cessation of agriculture was highly influential on soil P legacies, with clear reductions in the degree of labile and total P enrichment relative to reference ecosystems over time. Soil characteristics (clay content and pH) were strongly related to changes in labile P compared to reference sites, but these were relatively unimportant for total P. The duration of past agricultural use and climate were weakly related to changes in total P only. Our finding of reductions in the degree of soil P alteration over time relative to reference conditions reveals the potential to mitigate these land-use legacies in some soils. Better ability to predict dynamics of soil nutrient recovery after termination of agricultural use is essential to ecosystem management following land-use change.

4.1. INTRODUCTION

Conversion of undisturbed soils to agriculture can greatly influence soil phosphorus (P) pools (Sharpley & Smith 1983; Ellert & Gregorich 1996; Negassa & Leinweber 2009). Soil amendments, such as fertilizer and manure, typically raise cropland soil P concentrations when they exceed crop requirements (Hansen *et al.* 2002). At the same time, biomass removal through crop harvest can deplete soil P in low-input or P-limited systems (Newman 1997). Tillage mixes topsoil horizons and can accelerate transfer of nutrients from lower soil depths as well as from organic matter (Sharpley & Smith 1983; Jobbágy & Jackson 2001). Burning of vegetation in fields, which is common during shifting-cultivation in the tropics, also releases pulses of biologically available P into soils (Ewel 1986).

Permanent abandonment and temporary fallow of agricultural land is a global phenomenon that has implications for nutrient cycling in post-agricultural ecosystems (Hurtt *et al.* 2006; McLauchlan 2006). As much as 200 million hectares, or about 13% of the current global extent of cropland, have undergone agricultural abandonment over the past three centuries (Hobbs & Cramer 2007; Ramankutty *et al.* 2008b). A history of agricultural use can have important abiotic legacies on contemporary non-agricultural ecosystems by changing the pool of available P in soils as well as patterns of P distribution across the landscape (Fraterrigo *et al.* 2005; Standish *et al.* 2006; Baeten *et al.* 2010). These land-use legacies are known to have persistent impacts on soil P even decades to millennia after agricultural areas have been abandoned (Dupouey *et al.* 2002; Falkengren-Grerup *et al.* 2006; Hartshorn *et al.* 2006). Although a large portion of the Earth's surface may be subject to potential impacts of past agriculture on soil nutrient pools, large-scale understanding of the nature and longevity of these effects is limited.

Variability in the specific legacies of past agriculture on current soil P pools across studies has limited our ability to quantify trajectories of soil nutrients following agricultural abandonment (McLauchlan 2006). Historic cultivation has been shown to influence soil P content more than historic pasturing in temperate secondary forests (Koerner *et al.* 1997; Compton & Boone 2000), but these effects appear to differ within and between temperate regions (e.g., Europe (Wilson *et al.* 1997; de Keersmaeker *et al.* 2004) and North America (Fraterrigo *et al.* 2005; Matlack 2009)). Several authors have suggested that the duration of agriculture may control the magnitude of soil P legacies (Verheyen *et al.* 1999; Flinn *et al.* 2005), which could explain weaker legacy effects on plant available P in some secondary forests of North America compared to European forests with longer agricultural histories (Abrams & Hayes 2008; Matlack 2009). However, this hypothesis has not been tested at larger scales. Shifting-cultivation characteristic of the tropics often depletes total P relative to primary forests (McGrath *et al.* 2001), but effects of past agriculture on labile P also appear to vary between tropical regions based on soil types and agricultural practices (Ramakrishnan & Toky 1981; Lawrence & Schlesinger 2001; Johnson *et al.* 2003).

Phosphorus is integral to ecosystem management because it limits primary production in most freshwater and some terrestrial ecosystems (Elser *et al.* 2007). If agriculture elevates soil P, increased rates of P delivery to surface waters can cause prolonged eutrophication of aquatic ecosystems (Carpenter 2005). For example, Scott *et al.* (2001) found that dissolved P concentrations in runoff from a formerly agricultural forest with a history of manure applications were significantly higher than a nearby reference forest in central New York. Elevated soil P in temperate terrestrial ecosystems can contribute to altered vegetative community composition after agriculture, for example, by affecting the ability of certain species to colonize (Baeten *et al.* 2010). At the same time, agricultural depletion of bioavailable P in areas with highly-weathered and low-fertility soils could reduce future productivity (Markewitz *et al.* 2004). Such P-limitation has long been considered as critical to agricultural sustainability in the tropics (Ewel 1986).

The total stock of P in soils is distributed across distinct fractions with differing availability to plants (e.g., readily bioavailable or ‘labile’) and degrees of incorporation with mineral or organic soil constituents (Johnson *et al.* 2003). Labile P that is most accessible to plants typically represents a small but variable fraction of total P (Yang & Post 2011), while slowly-cycling organic and ‘occluded’ pools constitute a much larger share and regulate ecosystem P availability over decadal scales (Cross & Schlesinger 1995; Richter *et al.* 2006). Complex biological and geological processes within ecosystems determine the actual availability of P to plants and the total supply of P to the ecosystem, which can make measurement of distinct labile and occluded pools challenging (Yang & Post 2011). Weathering rates control total soil P availability in the long-term, but agricultural management can introduce P inputs and removal rates that are orders of magnitude greater than weathering rates (Newman 1997). Changes in total P supply and composition occur over geologic time scales, with declining P supply over pedogenesis as soils become increasingly weathered (Walker & Syers 1976). Land-use change and agricultural management can drastically accelerate or reverse soil P changes driven by long-term pedogenesis and may therefore have important ecological implications for P bioavailability (Garcia-Montiel *et al.* 2000; Townsend *et al.* 2002).

Better understanding of the intensity and duration of soil nutrient changes resulting from past agriculture could inform management of successional ecosystems following agricultural abandonment (Foster *et al.* 2003). Yet there has been little synthesis to understand broad patterns of soil P after cessation of agriculture or the factors that affect this (but see, e.g., McGrath *et al.* (2001)), and we are aware of no prior studies that have quantitatively addressed global patterns of post-agricultural soil P dynamics or comparisons across regions. We conducted a comprehensive global meta-analysis to assess potential legacies of past agriculture on current soil P pools and to investigate several hypotheses identified in the literature. We address the following specific questions for labile and total P: (1) Do broad categories of past agricultural land use result in any generalizable legacies for current soil P across regions? (2) What factors contribute most to soil P dynamics following cessation of agriculture? (3) Do land-use legacies on soil P differ over time since cessation of agriculture, and are these patterns consistent across regions? Based on the findings of previous studies, we also considered the unique effects of different vegetation transition types on post-agricultural soil P, particularly afforestation (Guo & Gifford 2002; Berthrong *et al.* 2009; Liao *et al. In press*), as well as potential variation in soil P legacies due to inherent differences in soils across soil orders and weathering status (Cross & Schlesinger 1995; Johnson *et al.* 2003; Negassa & Leinweber 2009).

4.2. Methods

We analyzed data from 94 studies to understand the degree to which soil P differs in areas with histories of cropland, pasture, or forage grassland following agricultural abandonment relative to pre-agricultural or current agriculture levels. Agricultural abandonment is broadly defined here as any change to non-agricultural use, whether permanent or temporary. We based these analyses on comparison of abandoned agricultural areas to nearby reference sites with no known history of agricultural use, as well as nearby sites that remained under agricultural use. Standard meta-analysis techniques based on resampling tests of mean effect size (Adams *et al.* 1997; Gurevitch *et al.* 2001) were used to assess changes in soil P attributable to the type of former land use and broad categories of post-agricultural vegetation, as well as the overall soil P legacies across regions and soil types. In a more in-depth meta-analysis using a subset of studies that reported more detailed information, we applied multifactor mixed effects models to assess the contribution of key variables that we hypothesized might determine post-agricultural soil P legacies.

We searched the peer-reviewed literature using common keywords related to soil phosphorus, agriculture, and land-use history in ISI Web of Science, Scopus, and Google Scholar up to March 2011. Studies included in our meta-analysis had to report labile or total P measurements for non-agricultural sites previously used for arable cropping, grazing, or forage grass production (at least 1 year since cessation of use), as well as a comparable reference site that best represented pre-agriculture soil or late-succession ecosystem conditions for a given location (e.g., 'primary forest') and had no reported agricultural history or minimal disturbance (57 and 32 studies for labile and total P, respectively). We also collected data from studies that reported soil P measurements for both contemporary and former agricultural sites that had comparable land use and management histories (37 and 23 for labile and total P). This approach provided two perspectives for assessing the temporal patterns of soil P following cessation of agriculture, with studies representative of most regions globally (Fig. S1; Appendix 1).

All studies had to contain the following methodological information: soil sampling depths and P measurement methods that were equivalent between the reference/current agriculture and abandoned agriculture sites; the dominant type of agriculture practiced prior to abandonment; the approximate time since cessation and vegetation type established after abandonment; as well as the number of study sites, replicates, and soil samples collected. Where available, we collected information on soil type to determine the dominant soil order (USDA taxonomy) for sites within each study as well as information on soil amendments (i.e., known or assumed histories of manure or fertilizer application). If data were unavailable from a given study, we cross-referenced studies from the same location to search for the missing information. Studies reporting fertilization, experimental manipulation of soils or extensive forest harvest after cessation of agriculture were omitted.

Although different methods were used to measure soil labile P across studies that may extract varying degrees of bioavailable P, use of consistent methods within individual studies ensured within-study comparability. The most common methods used to assess labile P (e.g., Olsen, Mehlich-3, Mehlich-1, Enger, and Bray-1; see Table S1) typically extract inorganic readily available P in soils, but several studies used Hedley fractionation (sum of three labile pools), which includes some exchangeable organic P (Johnson *et al.* 2003). Total P measurements were based on differing forms of acid digestion with the exception of one study that used spectroscopy (Table S2).

Soil bulk density (g soil cm^{-3}) may be altered as a result of land use, which confounds interpretation of soil nutrient changes based on concentration data for sites with differing land-use histories (Guo & Gifford 2002). We therefore focused on comparing changes in P content (expressed as kg ha^{-1}) rather than concentrations in order to control for possible bulk density effects. Many studies reported soil P content or provided bulk density data necessary to convert soil P concentrations to a mass per unit area. If no bulk density data were reported, we used available organic matter or carbon content to estimate bulk density based on a commonly used transfer function (Guo & Gifford 2002; Berthrong *et al.* 2009), or excluded studies altogether if this information was also missing.

4.2.1. Calculation of land-use legacy effects on soil phosphorus

We calculated the mean effect size of past agriculture on soil P pools following transition to non-agricultural land use using the natural logarithm of the response ratio (lnRR), X_{ab}/X_{ref} and X_{ab}/X_{ag}, where X_{ab} is the mean soil P content of the abandoned agricultural site, X_{ref} is the mean of the reference site and X_{ag} is the mean of the current agricultural site (Hedges *et al.* 1999). We used a nonparametric bootstrap resampling approach (e.g., Guo & Gifford 2002; Berthrong *et al.* 2009; Powers *et al.* 2011) to test for legacy effects across studies. The overall mean effect size (mean of all within-group observations) was calculated for each former land use as well as separate means for each vegetation transition category. Bootstrap bias-corrected accelerated 95% confidence intervals based on 10,000 random simulations were used to determine significance at the α=0.05 level (significant when intervals do not include zero or do not overlap between categories) (Adams *et al.* 1997; Canty & Ripley 2011). When more than one site or replicate was reported for a given study, we pooled observations according to time since cessation for sites with similar edaphic characteristics, management histories, or soil types, but observations from different depths were considered independent (*sensu* Powers *et al.* 2011).

We repeated the mean effect size analyses to test for overall legacy effects on soil P relative to reference sites for several other factors, including regions/continents, soil orders, and soil amendment histories. To test if our main results varied by sampling depth, we also assessed former land use effect sizes according to depth intervals (classified as surface soil (0-10cm), topsoil (10 to 30cm), subsoil (>30 cm), or full profile (up to 150 cm)). Measures of uncertainty (e.g., standard error) were underreported, so we judged uncertainty based on the inverse of sample size (Hoeksema *et al.* 2010). Because there was no systematic publication bias according to sample size (Fig. S2), we gave equal weight to all effect size observations in each of our meta-analyses. All statistical analyses were performed in R v.2.13.1 (R Development Core Team 2011), using the 'boot' package for bootstrapping (Canty & Ripley 2011).

4.2.2. Multifactor meta-analysis

We conducted a more detailed meta-analysis using additive mixed models (AMM) with a Gaussian (normal distribution) link function (Wood 2006). Available information was compiled from studies for additional variables that we hypothesized could influence post-agricultural soil P dynamics relative to reference sites (e.g., changes in soil pH, the approximate length of the agricultural period, and clay content; Table 1) for a total of 34 and 20 studies for labile and total P, respectively. We assessed variable relationships with AMM spline smoothers because several predictors were nonlinearly related to lnRR (estimated degrees of freedom (edf) >1), but standard linear mixed models were used where appropriate (Zuur *et al.* 2009). All response variables had approximately normal distributions after a total of four outliers were removed.

Use of mixed models accounted for the nested structure of the data (e.g., nested by study) in our multifactor analysis (Wood 2006). We tested four random effects structures and determined the best one using likelihood ratio tests of nested models or the Akaike Information Criterion (AIC): (1) a study-level effect; (2) the effect of observations nested within study; (3) a multilevel structure, with soil depth intervals nested within observations nested within studies; and (4) a study-level effect crossed with soil P measurement method (labile P methods were grouped into seven categories based on the principle extractant to test for any potential systematic effects). In all cases, the multilevel structure provided by far the lowest AIC and was used throughout.

We first examined the bivariate effect of each continuous predictor with AMMs and linear mixed models for categorical variables, and then conducted model selection using a subset of uncorrelated variables. This approach allowed for a comprehensive assessment of both the independent effects of each *a priori* variable regardless of the degree of correlation with other predictors (multicollinearity), as well as the additive effects of different combinations of terms. We used an information theoretic approach to model selection based on AIC-ranking and multi-model inference (Burnham & Anderson 2002) due to uncertainty with forward or backward model selection procedures (Zuur *et al.* 2009). Multicollinear predictors with high relative AICs were excluded from model selection until each term in the full model had a variance inflation factor <3 (determined with the 'AED' package), which is a fairly conservative cut-off (Zuur *et al.* 2009). Given the exploratory nature of our study, model ranking was based on an all-subsets model comparison, considering all possible combinations of terms (Grueber *et al.* 2011). For each model in the set of possible models, we calculated AIC_c (AIC corrected for small sample size), the ΔAIC_c (difference in AIC_c compared to the top-ranked model), as well as the Akaike weights (AI_w; the likelihood of a particular model being the best in a given candidate set) using the 'MuMIn' package (Barton 2011). AMM analyses were conducted using the 'mgcv' package for R (Wood 2006) and linear mixed models with 'nlme' (Pinheiro *et al.* 2011).

Model averaging and inference was based on the top ranked models (within $\Delta 2$ AIC_c), as these models can be considered essentially comparable to the model with the lowest AIC_c (Burnham & Anderson 2002). To account for model uncertainty, we also present the relative importance (RI) of variables in a larger candidate set, where the model AI_w summed to 0.95, indicating a 95% likelihood of that set containing the best model (Burnham & Anderson 2002). Because AIC_c can be influenced by the number of model parameters, we iteratively removed the spline smoothers on each continuous term (from lowest to highest overall edf) and reran the all-subsets calculations with each term entered in a linear parametric form to reduce the model parameters. The subsequent model set was retained if this procedure decreased the AIC_c of the top ranking model for that term.

We also used the complete reference and current agriculture comparison data from all studies to test overall time since transition (TST) effects with simple bivariate AMMs. To assess possible regional variation, we then fit a model testing the interaction of TST by Region for all abandoned cropland observations relative to reference sites (pasture and grassland were omitted to control for any confounding effects of land use), based on separate spline smoothers for each region to predict region-specific temporal dynamics. It was not possible to test TST by Region interactions with the current agriculture data due to limited sample size.

4.3. Results

4.3.1. Overall land-use legacy effects

Comparison of soil P content in abandoned agricultural areas to that of reference sites with no farming history reveals several important legacies of past agriculture on current labile and total P pools. We found overall mean elevation of soil P relative to reference sites following abandonment, particularly in areas that were formerly used as cropland. Differences in the magnitude and direction of soil P legacies across studies were also associated with the type of post-agricultural vegetation, soil texture, and fertilization histories. The greatest elevation of both labile and total P content relative to reference sites was constrained to areas that received fertilizer or manure amendments during the agricultural period. However, abandoned agricultural areas had consistently lower labile and total P compared to sites that remained in agricultural use.

Labile and total P relative to reference sites

There were relatively minimal differences in soil P legacies attributable to broad categories of former land use following abandonment, especially between areas used as cropland or pasture. Abandoned croplands had significant ($P<0.05$) overall elevation of soil labile and total P relative to reference sites, whereas abandoned forage grasslands had a significant mean depletion of soil P (Fig. 1). Although abandoned pastures had mean elevation of both P forms relative to reference sites, the overall effect was barely significant ($P\approx0.05$) for labile and not significant for total P. Our results show roughly equal numbers of observations with enrichment or depletion of labile and total P when considering all three former land use types (Fig. S2). There were no significant within-group differences for labile or total P between depth intervals (Fig. S3).

The type of vegetation established after agriculture was abandoned accounted for some changes in soil P relative to reference sites. Transitions to plantation (afforestation) in particular were associated with greater overall mean elevation of labile and total P across all three former land uses (cropland, pasture and forage grassland) (Fig. 1). Plantations, shrublands and grasslands established on abandoned croplands had higher mean labile and total P relative to reference sites compared to mean depletion for secondary forests or fallows, but these means only differed significantly for the labile P 'plantation' category. For abandoned pastures and forage grassland, there were particularly strong differences in soil P legacies for areas that transitioned to either secondary forest or plantation.

There were several significant overall legacy effects on soil P when studies were grouped according to soil order. However, patterns in the direction and magnitude of effect sizes between soil orders are clearer based on soil texture than weathering status (Table 2). We generally observed an association between higher mean clay content and mean depletion of soil P across the orders. More highly-weathered, very high clay content Oxisols (~40% clay) and high clay content Alfisols (~26%) were the only soil orders with significant (P<0.05) mean reduction in total P following abandonment relative to reference sites (Table 2). These soils also had generally reduced or no change for labile P. Conversely, medium clay content but highly-weathered Ultisols, and less-weathered Andisols and Entisols (12-15% clay), had significant mean elevation of labile P. Low clay content and moderately-weathered Spodosols (~6% clay) had strong and significant elevation of total P relative to reference sites. An additive mixed model testing the overall relationship between lnRR and clay content across all studies supports the important role of soil texture for labile P legacies (P=0.0007) but less so for total P (P=0.5910).

Information on historical soil nutrient amendments was reported in fewer than half of studies, but this had clear influence on the degree of soil P enrichment for areas with histories of cropland use. Soils that received fertilizer or manure applications at some point prior to abandonment had strong elevation of labile and total P contents relative to reference sites. In contrast, soils with no history of amendment had significant depletion of labile P and relatively no effect on total P (Fig. 2). All but three and two observations were from abandoned croplands for labile P and total P, respectively.

Labile and total P relative to current agricultural sites

Abandoned agricultural sites typically had lower mean labile and total P compared to sites still used for agriculture (Fig. 3). There was depletion or no change in soil P relative to current agricultural areas for every post-agricultural vegetation type except for labile P where agricultural areas became fallows. In the case of croplands, this relative depletion was only significant for both soil P measures where croplands became non-agricultural grassland or forest (P<0.05). After cessation of pasture use, there were minimal changes in labile and total P relative to current pastures for those areas that transitioned to secondary forest or plantation; however, a single study showed a more negative effect on labile P relative to current pasture following transition to non-agricultural grassland.

4.3.2. Multi-factor relationships and model selection

Our meta-analysis of studies with more detailed predictor data indicated strong bivariate relationships between several predictors and the response ratios for labile and total P (Table 3). Overall, five of the variables were significant for explaining differences in labile P between the abandoned agriculture and reference sites, including time since transition (TST), change in pH from the reference site (pHratio), whether or not the site was afforested (Plantation), clay content (Clay), and former land use (FLU). Six variables were significant for explaining differences in total P, including TST, length of the agricultural period (LOA), Region, maximum sampling depth (Depth), FLU, and mean annual temperature (MAT). In both cases, these variables had the lowest respective AIC_c values in the same order.

Model selection resulted in additive models with fairly strong explanatory power (~50-60% of variation in lnRR explained as judged by R^2 of the model with the lowest AIC_c; Table 3), and moderate levels of confidence based on the sum of Akaike weights (Δ2 AIC_c models: n=3 and $\sum AI_w$=0.32 for labile P; n=2 and $\sum AI_w$=0.36 for total P). TST was highly important for labile and total P, as it was included in each of the Δ2 AIC_c models as well as the entire 95% candidate set of models (RI=1.0; Fig. 6). Region was also important (RI>0.7 for both labile and total P) and there was considerable variation in the model-averaged parameters between regions (Figs 4 and 5). However, the RI values of soil characteristics (Clay and pHratio) differed greatly between labile P and total P (Fig. 6). Clay and pHratio had relatively strong negative and nonlinear effects for labile P, respectively, but neither was important across candidate models for total P (RI<0.5). The full model selection output for the 95% candidate sets of labile and total P models are presented in Appendices 2 and 3.

Several variables that had significant independent relationships with labile and total P effects had low importance when combined with the other model terms. In particular, the former land use (FLU) and the duration of the agricultural period (LOA) were retained in only a small proportion of the candidate models (RI<0.25) despite having significant bivariate relationships with the labile and total P responses ratios. However, Plantation appeared in all top AIC_c models for labile P, with a strong positive effect and relatively high importance (RI>0.5), which is consistent with the significant bivariate relationship.

We found only minimal evidence for climate influences on soil P legacies. MAT had a nonlinear and significant bivariate effect for total P (edf = 3.0; approximate P=0.005). However, MAT and mean annual precipitation (MAP) were multicollinear with Region (i.e., variance inflation factors >5) and were thus excluded from model selection. The weak bivariate relationship for MAP and AridityIndex for both P forms suggests that any climate relationship is subject to variability (Table 3).

4.3.4. Time since transition effects

Using data from all studies confirmed the strong temporal effects on soil P recovery following cessation of agriculture. TST had a significant effect on labile and total P in each of our meta-analysis comparisons, but this relationship was more variable for labile P relative to current agriculture sites (Fig 7). Labile P in abandoned agricultural areas relative to reference sites declined gradually over time (edf = 2.6; P<0.0001), but there was a weaker negative effect compared to current agricultural sites that disappeared after ~30 years (edf = 3.6; P=0.0133) (Fig. 7a,c). The negative and more linear TST effects for total P were similar for both reference and current agriculture comparisons (Fig. 7b,d). Some variation can be attributed to the former land uses represented at each TST value, particularly the distinction between former croplands and forage grasslands.

Abandoned croplands had overall declines in labile and total P relative to reference levels over time, but with distinct regional patterns (Fig. 8). We generally observed increases in labile P over time following depletion due to agricultural land-use history in tropical regions (Africa, South America, and South Asia) but declines in labile P over time following enrichment in temperate regions (Europe and North America). The TST by Region predicted curves for labile P decreased for Europe and North America (P<0.0001), but increased or changed little over time for other regions (not significant). Mean effect sizes for labile P differed significantly for some regions (e.g., South Asia/Africa/North Asia and Europe/North America; P<0.05). Almost all regional mean cropland effect sizes differed significantly from each other for total P, yet only Europe (P<0.0001) and North Asia (P=0.003) had significant temporal trends.

4.4. Discussion

Our global meta-analysis shows potentially large and enduring legacies of past agriculture on soil labile and total P across regions and soil types, but with some reduction in the magnitude of these effects over time since abandonment. Archaeological research in Europe and North America has indicated persistent elevation (Dupouey *et al.* 2002) and depletion (Sandor *et al.* 1986) of labile P even millennia after abandonment of early agricultural settlements. Although we found overall enrichment of soil P in abandoned agricultural sites compared to sites that were never used for agriculture, the most intense elevation of soil P pools occurred no more than 30-50 years since cessation of agriculture. In contrast, several areas had depleted soil P compared to reference ecosystems up to 100 years after abandonment, and the vast majority of abandoned agricultural sites had lower P content compared to sites still under agriculture. Time since cessation of agriculture is therefore essential for understanding how land-use history affects the supply of bioavailable and total P to post-agricultural ecosystems.

Our meta-analysis confirms that a history of cropland use had typically greater overall influence on soil P than pasturing (Koerner *et al.* 1997; Compton & Boone 2000; Fraterrigo *et al.* 2005), but there were stronger regional differences in the direction and temporal patterns of soil P legacies following abandonment. Taken as a whole, our findings suggest that vegetation, soil type, broad differences in management regimes across regions (especially shifting versus continuous cultivation), and to some degree climate may have an integrative effect on soil P fertility in the post-agricultural period. McGrath *et al.* (2001) attributed increases in labile inorganic P concentrations under secondary forests of the Amazon in the period after abandonment of shifting cultivation to rapid P cycling in younger forests before eventual incorporation of P in forest biomass. Our study suggests that this pattern possibly extends for a longer period in some tropical areas where soil P was depleted during the agricultural period, whereas some recovering temperate ecosystems act as a sink for agricultural soil P inputs over time.

We found some evidence that the type and duration of agriculture contributed to soil P legacies (e.g., Verheyen *et al.* 1999; McLauchlan 2006), but these factors had less influence than expected relative to other variables. The strong positive overall effects of past soil amendments indicates that differences in management and land-use intensity account for some inconsistencies in soil P legacies across studies. The form and duration of nutrient application can also distinctly influence different soil P fractions (Motavalli & Miles 2002; Negassa & Leinweber 2009), but detailed data on this was rare based on our literature search. Reduced labile P following abandonment compared to sites still under agriculture could be related to losses of P inputs from the agricultural period to more recalcitrant forms (Richter *et al.* 2006), greater mineralization of organic P in soils under current tillage (Sharpley & Smith 1983), or simply because some agricultural sites were still receiving fertilizer P inputs.

Interpretation of land-use legacies on soil P is also influenced by conditions in the post-agricultural period, especially the vegetation established following land-use change. Soil P in abandoned agricultural lands was enriched across all vegetation transition categories except secondary forests and fallows, which could be attributed to greater P demands of successional forests relative to other ecosystem types (Johnson *et al.* 2003; Markewitz *et al.* 2004). The magnitude of cultivation legacies on soil P have been shown to also differ between temperate successional coniferous and hardwood forests (Magid 1993; Compton & Boone 2000), possibly as a result of the greater ability of coniferous tree roots (e.g., under *Pinus* afforestation) to access mineralizable organic P that can supplement labile inorganic P pools (Chen *et al.* 2008a). Although afforestation often results in soil acidification (Jobbágy & Jackson 2001; Berthrong *et al.* 2009) that could reduce P availability due to binding of P with Al and Fe phosphates under more acidic conditions (Sollins *et al.* 1988), we found that most afforested sites still had elevated labile P pools despite commonly lower pH than references sites (almost all abandoned sites with pH ratios <1.0 in Fig. 4 were plantations). In other cases, P availability could be influenced by liming during the agricultural period (reported in a small number of studies) that persistently raises soil pH relative to undisturbed sites (Pywell *et al.* 1994), or the effects of varying species composition on soil pH in post-agricultural forests (Finzi *et al.* 1998).

Our meta-analysis indicates some consistent overall land-use legacies on soil P according to soil orders, but that soil texture may explain post-agricultural soil P dynamics more than weathering status. Clay content strongly influenced labile P, with much lower degrees of alteration compared to reference sites as clay content increased. Clay content and mineralogy directly affect the P sorption capacity of soils (Hansen *et al.* 2002), so prolonged elevation of labile P after agriculture is unlikely in clayey soils with higher P-fixation capacities where agricultural P inputs can precipitate to less-available forms (McCollum 1991). Our findings therefore support the role of soil texture as a mediating factor for how land-use history impacts soil P pools following abandonment in both relatively low-input (Lawrence & Schlesinger 2001) and high-input (Markewitz *et al.* 2002) systems.

Analyzing post-agricultural soil P dynamics is confounded by differences in initial soil fertility between sites with varying land-use histories (Flinn *et al.* 2005; McLauchlan 2006). Recurring sampling of the same location after agricultural abandonment enables direct conclusions about changes in soil P fractions (Richter *et al.* 2006), but retrospective designs were only used in three studies in our meta-analysis. Space-for-time substitutions (chronosequences) are instead commonly used to infer soil nutrient changes over a temporal gradient, but factors beyond just controlling for parent material (e.g., differing successional pathways across sites) could influence findings (Walker *et al.* 2010). Roughly half of the studies in our meta-analysis used stringent criteria to limit potential confounding factors (e.g., by choosing directly adjacent sites or comparing only sites with similar soil textures, topography, management histories, or landscape positions), whereas <10% reported no explicit efforts to minimize inherent site differences. Some of our results could therefore be influenced by limited comparability of reference and abandoned agricultural sites.

Although highly influential, the temporal trends in our meta-analysis are subject to some uncertainty. Manure has been used as a soil amendment for centuries in regions such as Western Europe (Verheyen *et al.* 1999), but limited use of mineral P fertilizers prior to the mid-20th century in most regions could explain the lesser soil P legacies in areas where agriculture ended more than a century ago. Marginal agricultural lands are also often the first to be abandoned due to inherently poor soil suitability, and least productive areas may be the only ones to never experience cultivation (Lawrence & Schlesinger 2001; Matlack 2009; Foote & Grogan 2010).

Changes in the labile P pools following termination of agriculture could have important consequences for both terrestrial and nearby aquatic ecosystems, whereas alteration of total P has long-term implications for the supply of bioavailable P to aggrading ecosystems (Richter *et al.* 2006). Meta-analysis of soil P dynamics following cessation of agriculture revealed some large and enduring land-use legacies on soil P, yet our temporal findings of decreased soil P enrichment over time suggest that mitigation of some impacts of past agriculture on soil P may be possible over shorter timescales than has been reported in individual studies (e.g., Dupouey *et al.* 2002). The type of successional vegetation established following agricultural abandonment is likely to play an important role in remediation of soil P levels. Our synthetic findings therefore indicate that accounting for the influence of land-use history on soil P should incorporate the roles of soil texture, alteration of pH, and successional vegetation (especially afforestation).

Trajectories of soil P content after agriculture are essential to better understanding how these land-use legacies affect the structure and function of contemporary ecosystems.

4.5. Acknowledgements

Robyn (Foote) Reudink and Paul Grogan kindly provided soil samples from their study (Ecosystems, 2010), on which Helene Higgins conducted laboratory analyses for soil P. We appreciate feedback on this manuscript from Alexander MacDonald, Matthew Mitchell, Kate Liss, as well as an anonymous reviewer. We thank Kamil Barton for assistance with the MuMIn package. Funding was provided to GKM and EMB from the Natural Sciences and Engineering Research Council of Canada (NSERC) and to GKM from the Fonds québécois de la recherche sur la nature et les technologies (FQRNT).

4.6. References

Abrams, M. & Hayes, V. (2008) Impacts of contrasting land-use history on composition, soils, and development of mixed-oak, coastal plain forests on Shelter Island, New York. *The Journal of the Torrey Botanical Society,* **135,** 37-52.

Adams, D. C., Gurevitch, J. & Rosenberg, M. S. (1997) Resampling test for meta-analysis of ecological data. *Ecology,* **78,** 1277-1283.

Baeten, L., Vanhellemont, M., De Frenne, P., De Schrijver, A., Hermy, M. & Verheyen, K. (2010) Plasticity in response to phosphorus and light availability in four forest herbs. *Oecologia,* **163,** 1021-1032.

Barton, K. (2011) *MuMIn: multi-model inference 1.5.2 for R version 2.13.1.* http://r-forge.r-project.org/projects/mumin/

Berthrong, S. T., Jobbágy, E. & Jackson, R. B. (2009) A global meta-analysis of soil exchangeable cations, pH, carbon, and nitrogen with afforestation. *Ecological Applications,* **19,** 2228-2241.

Burnham, K. P. & Anderson, D. R. (2002) *Model Selection and Multimodel Inference: A Practical Information-Theoretic Approach*. Springer Verlag, New York, NY.

Canty, A. & Ripley, B. (2011) boot: Bootstrap R (S-Plus) Functions. R package version 1.3-2.

Carpenter, S. R. (2005) Eutrophication of aquatic ecosystems: Bistability and soil phosphorus. *Proceedings of the National Academy of Sciences of the United States of America,* **102,** 10002-10005.

Chen, C. R., Condron, L. M. & Xu, Z. H. (2008) Impacts of grassland afforestation with coniferous trees on soil phosphorus dynamics and associated microbial processes: A review. *Forest Ecology and Management,* **255,** 396-409.

Compton, J. & Boone, R. (2000) Long-term impacts of agriculture on soil carbon and nitrogen in New England forests. *Ecology,* **81,** 2314-2330.

Cross, A. F. & Schlesinger, W. H. (1995) A literature review and evaluation of the. Hedley fractionation: Applications to the biogeochemical cycle of soil phosphorus in natural ecosystems. *Geoderma,* **64,** 197-214.

de Keersmaeker, L., Martens, L., Verheyen, K., Hermy, M., De Schrijver, A. & Lust, N. (2004) Impact of soil fertility and insolation on diversity of herbaceous woodland species colonizing afforestations in Muizen forest (Belgium). *Forest Ecology and Management,* **188,** 291-304.

Dupouey, J., Dambrine, E., Laffite, J. & Moares, C. (2002) Irreversible impact of past land use on forest soils and biodiversity. *Ecology,* **83,** 2978-2984.

Ellert, B. H. & Gregorich, E. G. (1996) Storage of carbon, nitrogen and phosphorus in cultivated and adjacent forested soils of Ontario. *Soil Science,* **161,** 587-603.

Elser, J. J., Bracken, M. E. S., Cleland, E. E., Gruner, D. S., Harpole, W. S., Hillebrand, H., Ngai, J. T., Seabloom, E. W., Shurin, J. B. & Smith, J. E. (2007) Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecology Letters,* **10,** 1135-1142.

Ewel, J. J. (1986) Designing agricultural ecosystems for the humid tropics. *Annual Review of Ecology and Systematics,* **17,** 245-271.

Falkengren-Grerup, U., Brink, D. & Brunet, J. (2006) Land use effects on soil N, P, C and pH persist over 40–80 years of forest growth on agricultural soils. *Forest Ecology and Management,* **225,** 74-81.

Finzi, A. C., Breemen, N. V. & Canham, C. D. (1998) Canopy tree-soil interactions within temperate forests: Species effects on soil carbon and nitrogen. *Ecological Applications,* **8,** 440-446.

Flinn, K. M., Vellend, M. & Marks, P. L. (2005) Environmental causes and consequences of forest clearance and agricultural abandonment in central New York, USA. *Journal of Biogeography,* **32,** 439-452.

Foote, R. & Grogan, P. (2010) Soil carbon accumulation during temperate forest succession on abandoned low productivity agricultural lands. *Ecosystems,* **13,** 795-812.

Foster, D., Swanson, F., Aber, J., Burke, I., Brokaw, N., Tilman, D. & Knapp, A. (2003) The importance of land-use legacies to ecology and conservation. *BioScience,* **53,** 77-88.

Fraterrigo, J., Turner, M., Pearson, S. & Dixon, P. (2005) Effects of past land use on spatial heterogeneity of soil nutrients in southern Appalachian forests. *Ecological Monographs,* **75,** 215-230.

Garcia-Montiel, D. C., Neill, C., Melillo, J., Thomas, S., Steudler, P. A. & Cerri, C. C. (2000) Soil phosphorus transformations following forest clearing for pasture in the Brazilian Amazon. *Soil Science Society of America Journal,* **64,** 1792-1804.

Grueber, C. E., Nakagawa, S., Laws, R. J. & Jamieson, I. G. (2011) Multimodel inference in ecology and evolution: challenges and solutions. *Journal of Evolutionary Biology,* **24,** 699-711.

Guo, L. B. & Gifford, R. M. (2002) Soil carbon stocks and land use change: a meta analysis. *Global Change Biology,* **8,** 345-360.

Gurevitch, J., Curtis, P. S. & Jones, M. H. (2001) Meta-analysis in ecology. *Advances in Ecological Research,* **32,** 199-247.

Hansen, N. C., Daniel, T. C., Sharpley, A. N. & Lemunyon, J. L. (2002) The fate and transport of phosphorus in agricultural systems. *Journal of Soil and Water Conservation,* **57,** 408-417.

Hartshorn, A. S., Chadwick, O. A., Vitousek, P. M. & Kirch, P. V. (2006) Prehistoric agricultural depletion of soil nutrients in Hawai'i. *Proceedings of the National Academy of Sciences,* **103,** 11092-11097.

Hijmans, R. J., Cameron, S. E., Parra, J. L., Jones, P. G. & Jarvis, A. (2005) Very high resolution interpolated climate surfaces for global land areas. International Journal of Climatology, 25, 1965-1978.

Hedges, L. V., Gurevitch, J. & Curtis, P. S. (1999) The meta-analysis of response ratios in experimental ecology. *Ecology,* **80,** 1150-1156.

Hobbs, R. & Cramer, V. (2007) Why Old Fields? Socioeconomic and ecological causes and consequences of land abandonment. *Old fields: dynamics and restoration of abandoned farmland* (eds. V. Cramer & R. Hobbs), pp. 1-15. Island press, Washington DC.

Hoeksema, J. D., Chaudhary, V. B., Gehring, C. A., Johnson, N. C., Karst, J., Koide, R. T., Pringle, A., Zabinski, C., Bever, J. D., Moore, J. C., Wilson, G. W. T., Klironomos, J. N. & Umbanhowar, J. (2010) A meta-analysis of context-dependency in plant response to inoculation with mycorrhizal fungi. *Ecology Letters,* **13,** 394-407.

Hurtt, G. C., Frolking, S., Fearon, M. G., Moore, B., Shevliakova, E., Malyshev, S., Pacala, S. W. & Houghton, R. A. (2006) The underpinnings of land-use history: three centuries of global gridded land-use transitions, wood-harvest activity, and resulting secondary lands. *Global Change Biology,* **12,** 1208-1229.

Jobbágy, E. & Jackson, R. (2001) The distribution of soil nutrients with depth: global patterns and the imprint of plants. *Biogeochemistry,* **53,** 51-77.

Johnson, A. H., Frizano, J. & Vann, D. R. (2003) Biogeochemical implications of labile phosphorus in forest soils determined by the Hedley fractionation procedure. *Oecologia,* **135,** 487-499.

Koerner, W., Dupouey, J., Dambrine, E. & Benoit, M. (1997) Influence of past land use on the vegetation and soils of present day forest in the Vosges mountains, France. *Journal of Ecology,* **85,** 351-358.

Lawrence, D. & Schlesinger, W. H. (2001) Changes in soil phosphorus during 200 years of shifting cultivation in Indonesia. *Ecology,* **82,** 2769-2780.

Liao, C., Luo, Y., Fang, C., Chen, J. & Li, B. (*In press*) The effects of plantation practice on soil properties based on the comparison between natural and planted forests: a meta-analysis. *Global Ecology and Biogeography*.

Magid, J. (1993) Vegetation effects on phosphorus fractions in set-aside soils. *Plant and Soil,* **149,** 111-119.

Markewitz, D., Davidson, E., Moutinho, P. & Nepstad, D. (2004) Nutrient loss and redistribution after forest clearing on a highly weathered soil in amazonia. *Ecological Applications,* **14,** 177-199.

Markewitz, D., Sartori, F. & Craft, C. (2002) Soil change and carbon storage in longleaf pine stands planted on marginal agricultural lands. *Ecological Applications,* **12,** 1276-1285.

Matlack, G. (2009) Long-term changes in soils of second-growth forest following abandonment from agriculture. *Journal of Biogeography,* **36,** 2066-2075.

McCollum, R. E. (1991) Buildup and Decline in Soil Phosphorus: 30-Year Trends on a Typic Umprabuult. *Agronomy Journal,* **83,** 77-85.

McGrath, D. A., Smith, C. K., Gholz, H. L. & Oliveira, F. d. A. (2001) Effects of land-use change on soil nutrient dynamics in Amazonia. *Ecosystems,* **4,** 625-645.

McLauchlan, K. (2006) The nature and longevity of agricultural impacts on soil carbon and nutrients: A review. *Ecosystems,* **9,** 1364-1382.

Motavalli, P. M. & Miles, R. M. (2002) Soil phosphorus fractions after 111 years of animal manure and fertilizer applications. *Biology and Fertility of Soils,* **36,** 35-42.

Negassa, W. & Leinweber, P. (2009) How does the Hedley sequential phosphorus fractionation reflect impacts of land use and management on soil phosphorus: A review. *Journal of Plant Nutrition and Soil Science,* **172,** 305-325.

Newman, E. I. (1997) Phosphorus balance of contrasting farming systems, past and present: Can food production be sustainable? *Journal of Applied Ecology,* **34,** 1334-1347.

Pinheiro, J., Bates, D., DebRoy, S., Sarkar, D. & R Development Core Team (2011) *nlme: Linear and Nonlinear Mixed Effects Models. R package version 3.1-102*

Powers, J. S., Corre, M. D., Twine, T. E. & Veldkamp, E. (2011) Geographic bias of field observations of soil carbon stocks with tropical land-use changes precludes spatial extrapolation. *Proceedings of the National Academy of Sciences,* **108,** 6318-6322.

Pywell, R. F., Webb, N. R. & Putwain, P. D. (1994) Soil fertility and its implications for the restoration of heathland on farmland in Southern Britain. *Biological Conservation,* **70,** 169-181.

R Development Core Team (2011) *R: A language and environment for statistical computing. R Foundation for Statistical Computing.* http://www.R-project.org

Ramakrishnan, P. & Toky, O. (1981) Soil nutrient status of hill agro-ecosystems and recovery pattern after slash and burn agriculture (Jhum) in north-eastern India. *Plant and Soil,* **60,** 41-64.

Ramankutty, N., Evan, A. T., Monfreda, C. & Foley, J. A. (2008) Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000. *Global biogeochemical cycles,* **22,** GB1003.

Richter, D., Allen, H., Li, J., Markewitz, D. & Raikes, J. (2006) Bioavailability of slowly cycling soil phosphorus: major restructuring of soil P fractions over four decades in an aggrading forest. *Oecologia,* **150,** 259-271.

Sandor, J., Gersper, P. & Hawley, J. (1986) Soils at prehistoric agricultural terracing sites in New Mexico: iii. Phosphorus, selected micronutrients, and pH. *Soil Science Society of America Journal,* **50,** 177.

Scott, C., Walter, M., Nagle, G., Walter, M., Sierra, N. & Brooks, E. (2001) Residual phosphorus in runoff from successional forest on abandoned agricultural land: 1. Biogeochemical and hydrological processes. *Biogeochemistry,* **55,** 293-310.

Sharpley, A. N. & Smith, S. J. (1983) Distribution of phosphorus forms in virgin and cultivated soils and potential erosion losses. *Soil Science Society of America Journal,* **47,** 581-586.

Sollins, P., Robertson, G. & Uehara, G. (1988) Nutrient mobility in variable- and permanent-charge soils. *Biogeochemistry,* **6,** 181-199.

Standish, R., Cramer, V., Hobbs, R. & Kobryn, H. (2006) Legacy of land-use evident in soils of Western Australia's wheatbelt. *Plant and Soil,* **280,** 189-207.

Townsend, A. R., Asner, G. P., Cleveland, C. C., Lefer, M. E. & Bustamante, M. M. C. (2002) Unexpected changes in soil phosphorus dynamics along pasture chronosequences in the humid tropics. *Journal of Geophysical Research,* **107,** 8067.

Verheyen, K., Bossuyt, B., Hermy, M. & Tack, G. (1999) The land use history (1278-1990) of a mixed hardwood forest in western Belgium and its relationship with chemical soil characteristics. *Journal of Biogeography,* **26,** 1115-1128.

Walker, L. R., Wardle, D. A., Bardgett, R. D. & Clarkson, B. D. (2010) The use of chronosequences in studies of ecological succession and soil development. *Journal of Ecology,* **98,** 725-736.

Walker, T. W. & Syers, J. K. (1976) The fate of phosphorus during pedogenesis. *Geoderma,* **15,** 1-19.

Wilson, B. R., Moffat, A. J. & Nortcliff, S. (1997) The nature of three ancient woodland soils in southern England. *Journal of Biogeography,* **24,** 633-646.

Wood, S. (2006) *Generalized Additive Models: An Introduction with R*. Chapman and Hall/CRC, Boca Raton, Florida.

Yang, X. & Post, W. M. (2011) Phosphorus transformations as a function of pedogenesis: A synthesis of soil phosphorus data using Hedley fractionation method. *Biogeosciences,* **8,** 2907-2916.

Zuur, A. F., Ieno, E. N., Walker, N., Saveliev, A. A. & Smith, G. M. (2009) *Mixed effects models and extensions in ecology with R*. Springer, New York, NY.

4.6. Tables and figures

Table 1. Variables used for model selection and multi-model inference.

Variable	Explanation
	Response variable
lnRR (soil P legacy effect size)	Natural log-transformed mean response ratio.
	Explanatory predictors (continuous)
Time since transition (TST)	Approximate years since abandonment (transition to non-agricultural use), taken as the mean of two dates when a range was reported; data was pooled by TST across similar sites for some studies.
Clay content (Clay)	Mean % clay of former agriculture and reference sites; mean clay content for soil texture classes from Miller & White (1998) was used if % clay was not reported directly.
Mean annual temperature (MAT)	Extracted from the WorldClim (Hijmans *et al.* 2005) spatial dataset (C); mean of 1961-1990 values.
Mean annual precipitation (MAP)	Extracted from the WorldClim (Hijmans *et al.* 2005) spatial dataset (mm); mean of 1961-1990 values.
Aridity Index	Simple climate metric calculated as the ratio of precipitation to temperature (MAP+10/MAT) (de Martonne 1926).
pH ratio	Ratio of soil pH of former agriculture site and reference site.
Maximum Depth	Maximum reported sampling depth (cm) for observation.
	Explanatory variables (categorical)
Former land use (FLU)	3 levels (cropland, pasture, forage grassland).
Region	5 to 6 levels for labile and total P (Africa, Europe, North America, Oceania/Pacific Islands, South America, South Asia).
Plantation	2 levels (non-plantation, plantation/afforestation).
Length of agriculture (LOA)	4 levels (short: <10 years; medium-short: 10-50 years; medium-long: 50-100 years; long: >100 years).

Table 2. Mean effect sizes for abandoned agricultural sites according to soil order (positive: weak (+) 0 to 0.5; strong (++) 0.5 to 1.0; very strong (+++) >1.0; negative: weak (-) 0 to -0.5; strong (--) -0.5 to -1.0). Significant effects (P<0.05) based on bootstrapped bias-corrected 95% confidence intervals are shown in bold. Soil orders are sorted from lowest to highest weathering status following Cross & Schlesinger (1995) and mean clay content is shown (as in Table 1) pooled between the labile and total P observations.

	Labile P		Total P		Approximate mean clay content (%)
Soil order	*# studies/obs.*	*effect*	*# studies/obs.*	*effect*	
Andisols	3/8	**+++**	2/4	+	15
Entisols	13/65	**++**	6/32	**+**	13
Inceptisols	10/35	-	6/43	+	26
Mollisols	3/5	++	5/16	--	26
Spodosols	6/59	+	5/34	**++**	6
Alfisols	10/40	**--**	3/10	**-**	26
Ultisols	5/34	**++**	4/27	+	12
Oxisols	7/41	+/-	5/32	**-**	40

Table 3. Bivariate relationships for lnRR and explanatory predictors for labile and total P. *F* values and estimated degrees of freedom (edf) for continuous (spline smoother; s(x)) terms from AMM models and denominator degrees of freedom (df) for parametric terms tested with linear mixed effects models are shown. Significant variable relationships are indicated in bold (****P* < 0.001, ** *P* < 0.01, * *P* < 0.05); however, *P* values for smooth terms are approximate at values 0.2 < *P* < 0.001 (Wood 2006; Zuur *et al.* 2009).

	Labile P					Total P				
Model terms	F	edf / df	approx. *P* value	R^2	AIC_c	F	edf / df	approx. *P* value	R^2	AIC_c
Smooth terms										
s(TST)	**11.7*****	**2.4**	**<0.001**	**0.16**	**487**	**17.6*****	**1.5**	**<0.001**	**0.08**	**98**
s(Clay)	**3.6***	**2.6**	**0.020**	**0.10**	**501**	0.7	1.0	0.406	0.03	119
s(MAP)	0.3	1.0	0.586	0.00	508	1.3	1.7	0.277	0.06	118
s(MAT)	2.1	2.4	0.114	0.11	508	**4.5****	**3.0**	**0.005**	**0.39**	**114**
s(AridityIndex)	0.6	1.0	0.452	0.04	507	0.2	1.0	0.678	0.02	119
s(pH ratio)	**5.1*****	**4.4**	**<0.001**	**0.09**	**497**	3.9	1.0	0.050	0.01	116
s(Max. depth)	3.2	1.0	0.076	0.02	505	**4.5****	**2.6**	**0.007**	**0.03**	**112**
Parametric terms										
FLU	**4.3***	**100**	**0.016**	**-**	**501**	**3.4***	**88**	**0.038**	**-**	**113**
Region	1.3	29	0.295	-	507	**4.1***	**14**	**0.017**	**-**	**108**
Plantation	**7.4****	**101**	**0.008**	**-**	**500**	1.9	89	0.176	-	115
LOA	1.4	99	0.245	-	506	**4.5****	**87**	**0.006**	**-**	**108**
Null model				-	504				-	115
Full model				0.45	477				0.49	96
Model with lowest AIC_c				0.49	469				0.58	81

FIGURES

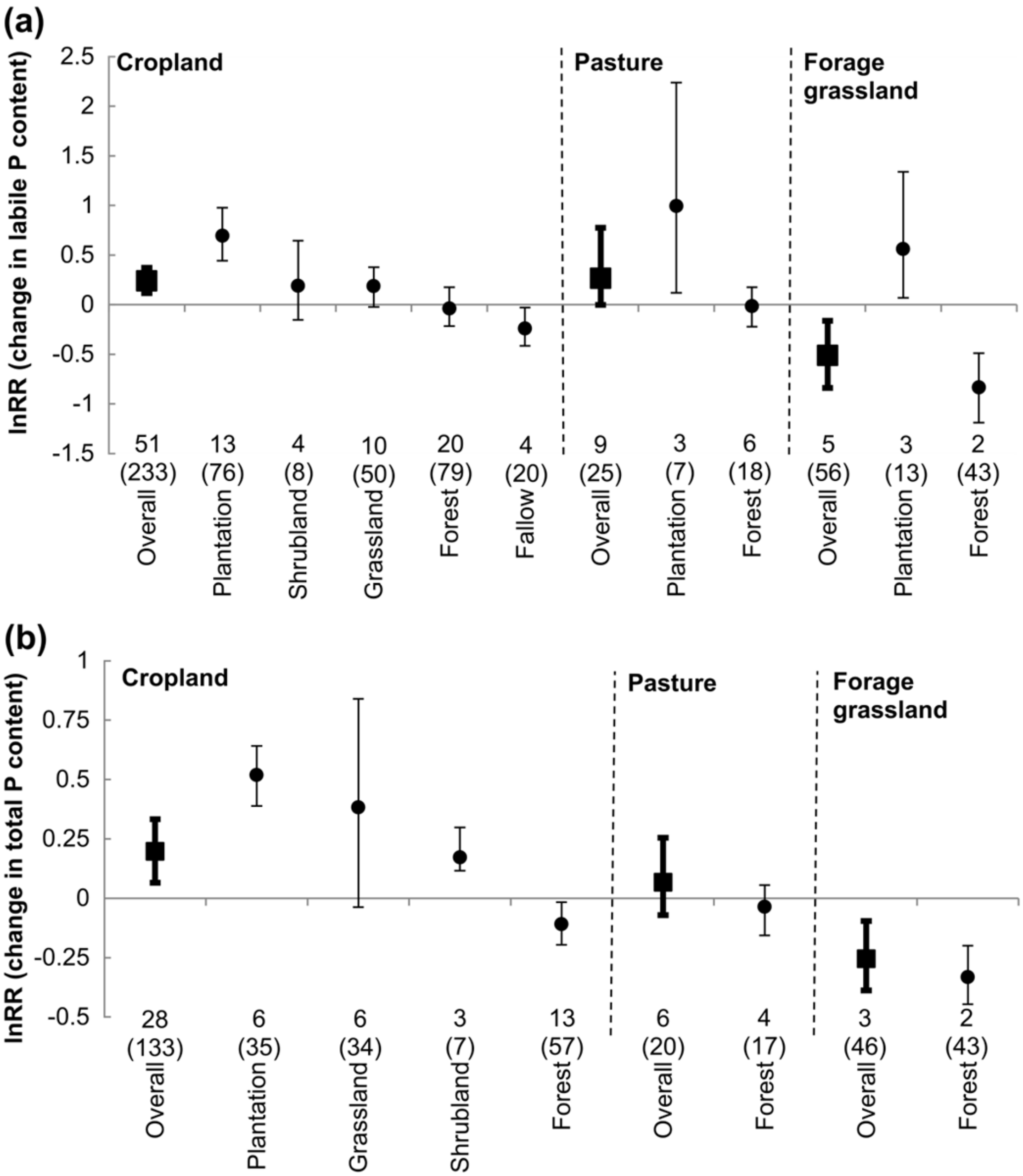


Fig. 1. Mean effect sizes (lnRR) for labile (a) and total P (b) with bootstrap bias-corrected 95% confidence intervals based on comparison of abandoned agricultural areas with reference sites. The mean for each post-agricultural vegetation transition type is shown, as well as the overall mean of all observations within the corresponding former land use category. The number of studies and observations (in parentheses) are presented below each mean. The total P overall mean effect sizes for pasture and forage grassland include 'plantation' observations (not shown separately because <4 observations each).

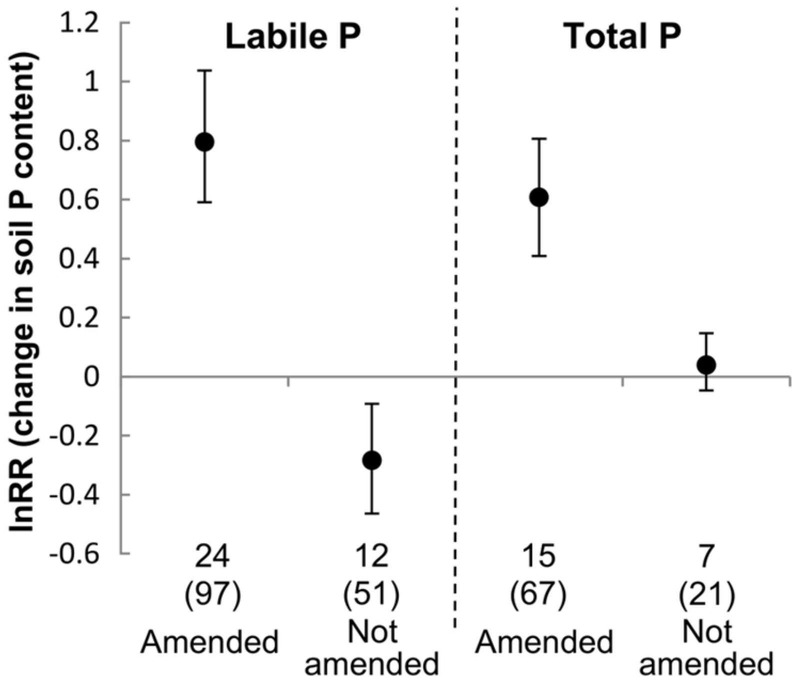


Fig. 2. Means and bootstrap bias-corrected confidence intervals for abandoned agricultural sites with reported manure or fertilizer amendment histories (Amended), and sites that did not receive historic amendments (Not amended), relative to the P content of reference sites. The number of studies and observations (in parentheses) is given.

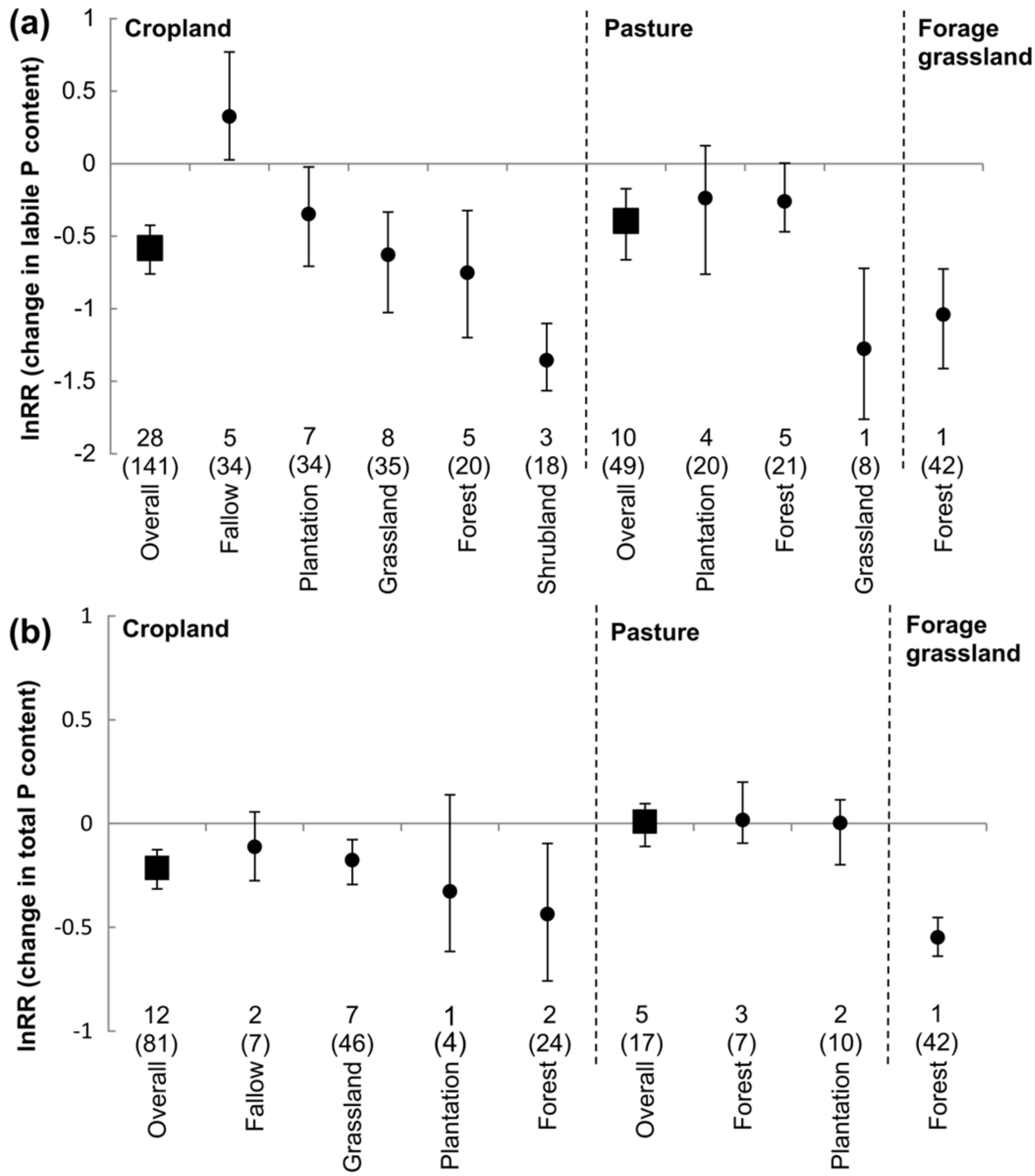


Fig. 3. Mean effect sizes and bias-corrected confidence intervals for labile (a) and total P (b) based on comparison of abandoned agricultural areas with current agricultural sites. The mean for each vegetation transition type is shown, as well as the overall mean of all observations within the corresponding former land use category. The number of studies and observations (in parentheses) are reported below each mean.

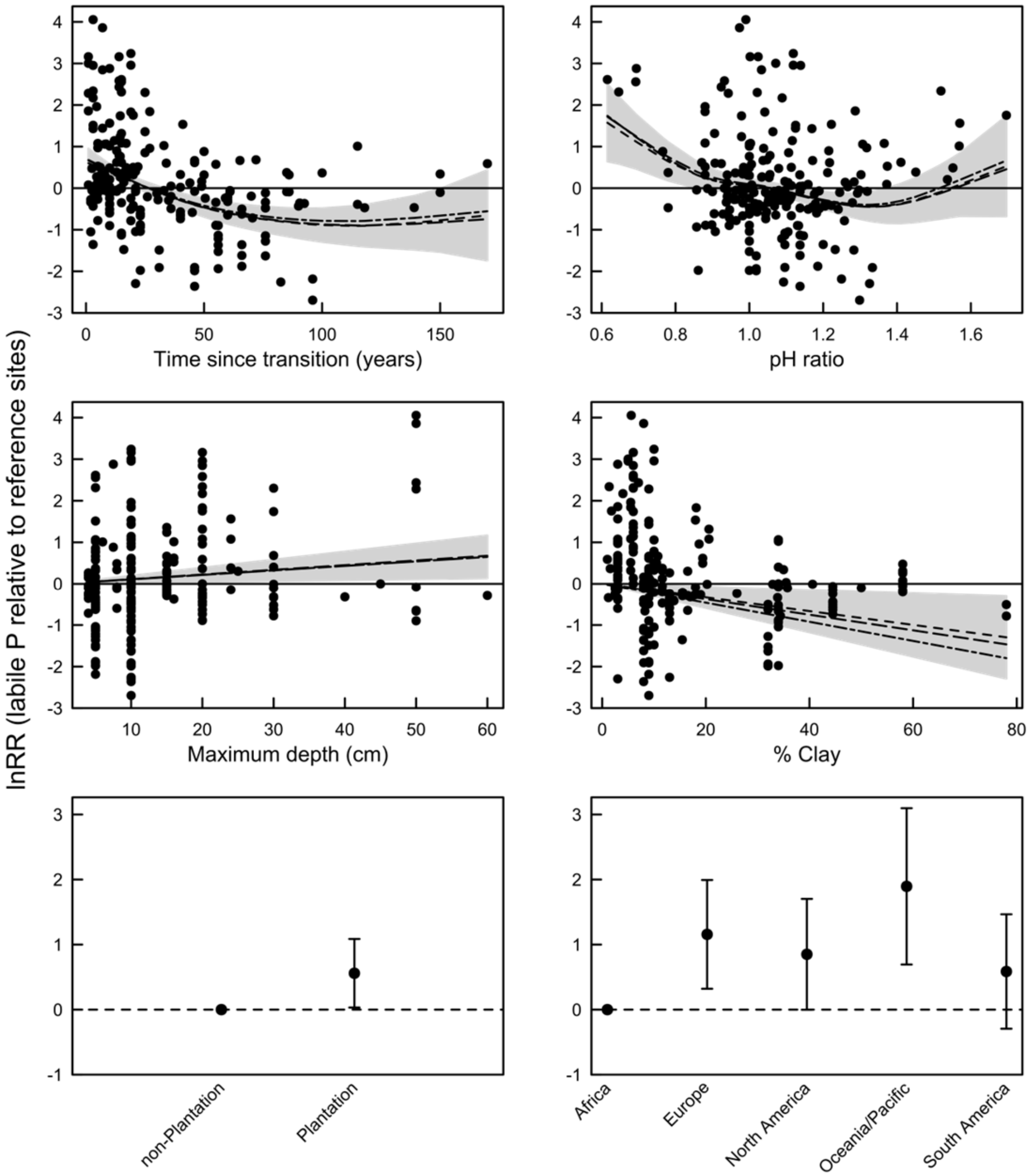


Fig. 4. Labile P model selection results for top-ranked models (Δ2 AIC_c). Plots show fitted AMM lines and data points for each model (*n*=3; dashed lines) representing the predictors retained in the set. Confidence intervals were similar for each of the fitted lines, so only those for the lowest AIC_c model are displayed (shaded regions). Model-averaged parameter estimates and 95% confidence intervals are shown for each level of the categorical variables, relative to reference levels (reference level=0).

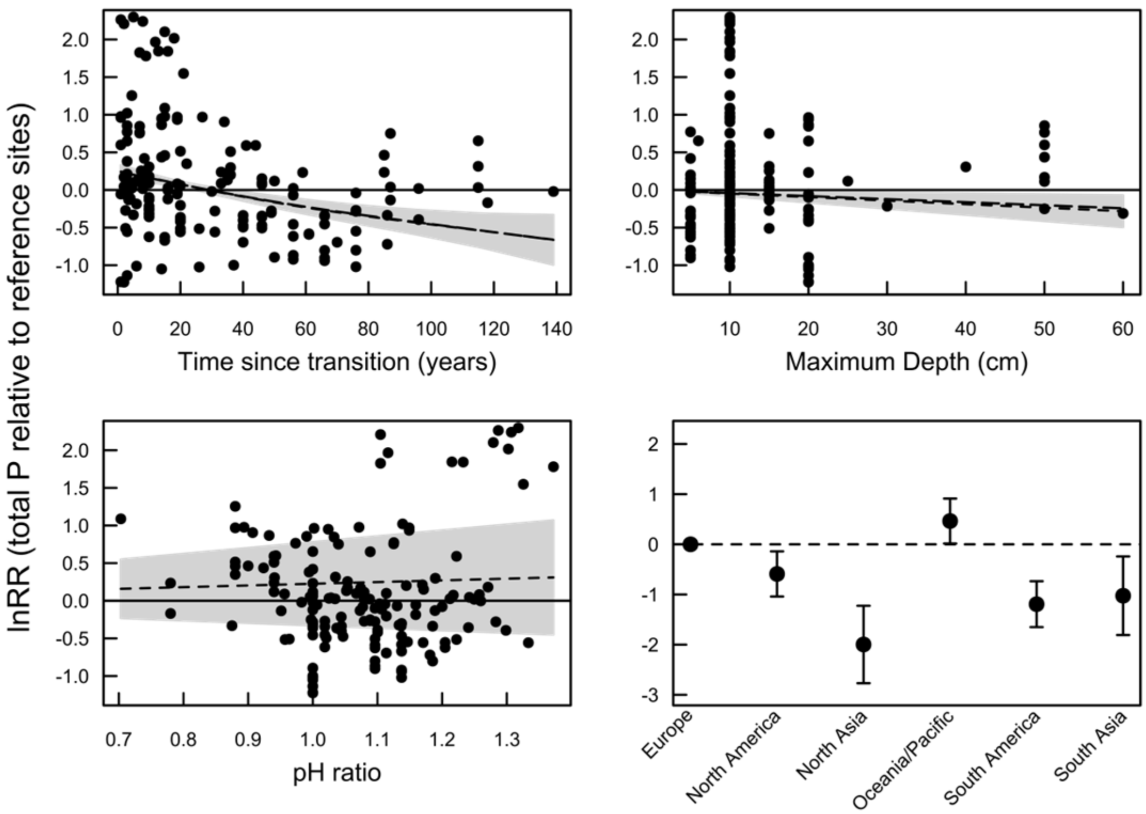


Fig. 5. Total P model selection results for the top-ranked model (n=2) in the Δ2 AIC_c set. Plots show the fitted AMM lines and data points for the explanatory variables retained. Model-averaged parameter estimates and 95% confidence intervals are shown for each level of the categorical variable relative to the reference level for Region ('Europe'=0).

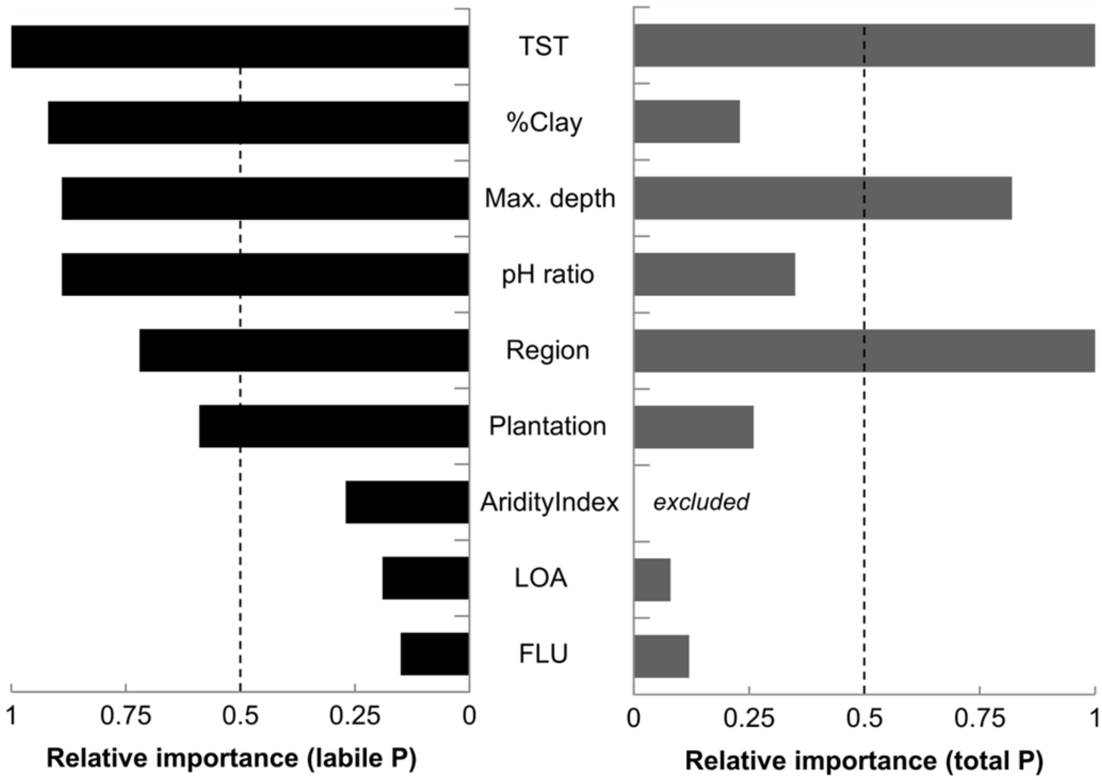


Fig. 6. Relative importance scores for labile and total P models, defined as the sum of the Akaike weights ($\sum AIw_i$) for each predictor across all models in the 95% confidence candidate set ($\sum AIw_i = 0.95$). Models retained in the set were *n*=77 (out of 512 possible models) and *n*=30 (out of 256 possible models) for labile and total P, respectively. An RI >0.5 indicates a relatively important variable (Burnham & Anderson 2002).

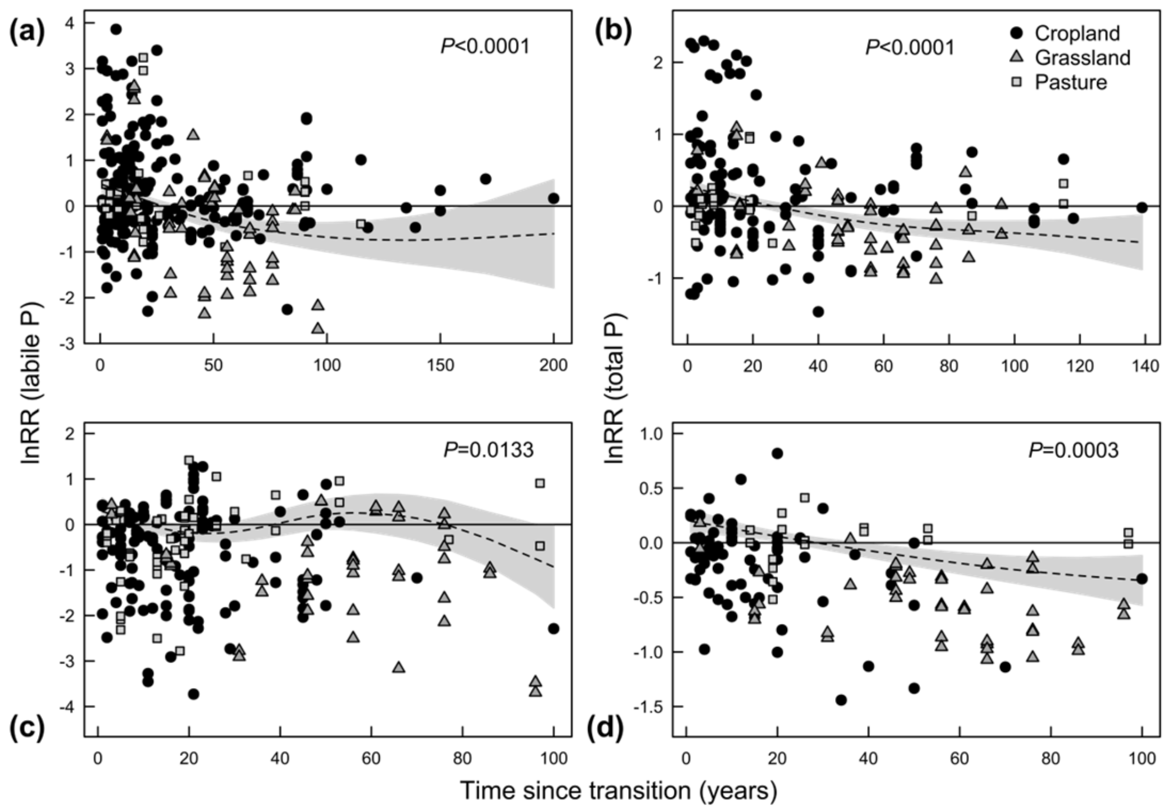


Fig. 7. Results of AMM models testing the effect of TST on lnRR for labile (a) and total P (b) based on all studies with reference site data, compared with results for labile (c) and total P (d) using the data from all studies with current agriculture sites. Former land uses are indicated for context.

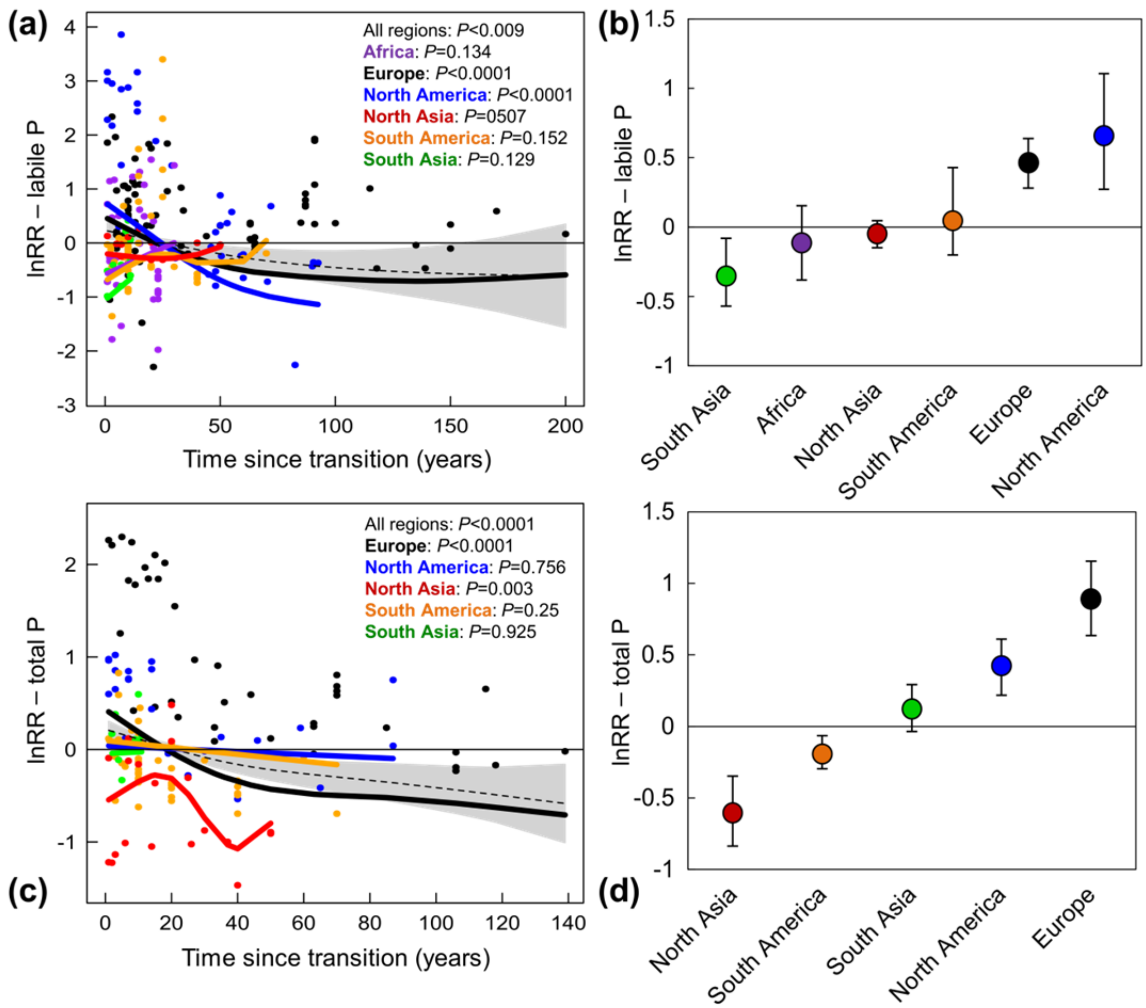


Fig. 8. Temporal trends in labile (a) and total P (c) effect size over time since transition for all abandoned croplands relative to references sites (dashed lines with confidence intervals) and corresponding interactive effects of TST by Region as predicted AMM lines. Plots (b) and (d) show mean lnRR for abandoned cropland sites in each region with bias-corrected 95% confidence intervals for labile and total P, respectively.

4.7. SUPPORTING INFORMATION

SI TABLES

Table S1. Methods used for measuring labile P, including broader grouping of methods based on similar chemical extractants. The specific methods in italics were most common across studies in the meta-analysis.

Method groupings	Specific methods
H_2SO_4	*Mehlich 1* (also includes some HCl methods) H_2SO_4 (weak) Duchafours method Truog method
NH_4F (Ammonium fluoride)	*Bray and Kurtz* Ammonium fluoride
$NaHCO_3$	*Hedley labile* (Resin + $NaHCO_3$-Pi + $NaHCO_3$-Po) *Olsen* Modified Olsen methods Dabin Olsen Colwell Olsen
Lactate or acetic acids (CH_3COOH):	Lactate methods Enger method Enger-Riehm method
Other acetates / Acetate combinations	Lancaster method Morgan's method (Ammonium acetate) *Mehlich 3*
Other acids and HCl	Kirsanov method (HCl) Olsen and Sommers (Na_2SO4 + NaF)
Calcium Chloride	$CaCl_2$

Table S2. Methods used for measuring total P, including broader grouping of methods based on similar approaches used. The specific methods in italics were most common across studies in the meta-analysis.

Method groupings	Specific methods
Acid extractants and digestions	*Nitric acid: HNO_3 (and under pressure)*
	Perchloric acid: $HClO_4$
	Hydrofluoric acid
Other acid digestion	Aqua regia digestion
	Kjeldahl TP method
	Hydrofluoric acid digestion
	Modified Kjeldahl method (Parkinson and Allen, 1975)
	Other sulfuric acid/hydrogen peroxide methods
	Hedley (and modified Hedley) sequential extraction (sum)
Fusion methods	NaOH fusion
	Sodium carbonate fusion
Spectroscopy	X-ray fluorescence spectrometry

SI FIGURES

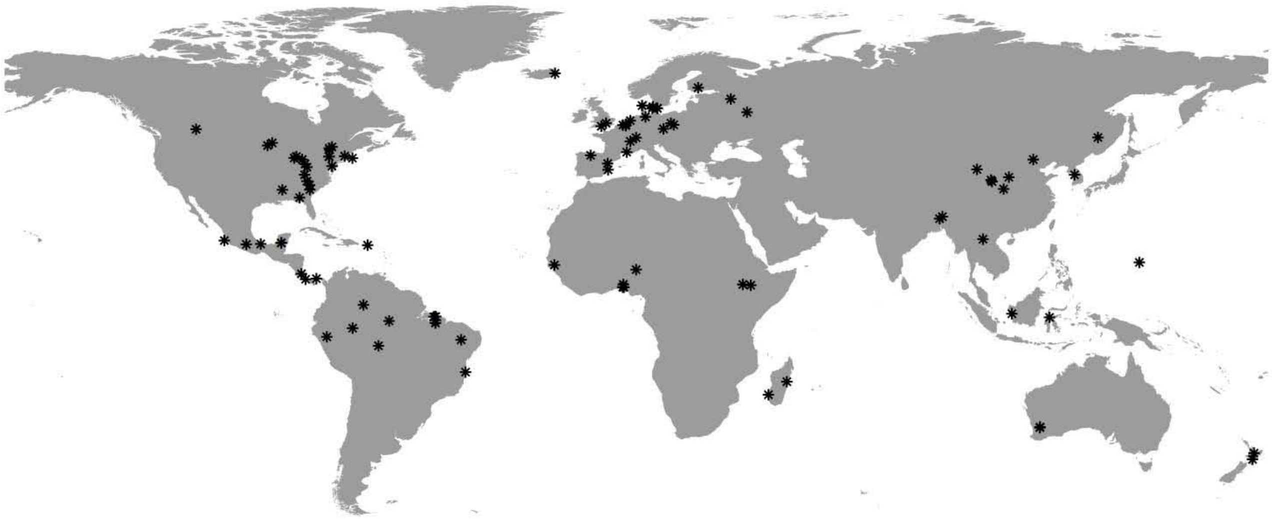


Fig. S1. Map of approximate study locations for the complete set of 94 studies included in the meta-analysis. Locations are based on coordinates provided or study area descriptions (e.g., place names). Where more than one location was included for a given publication, each broad study area has been indicated. If two studies considered the same general area but used different sampling locations, only one point has been marked on the map.

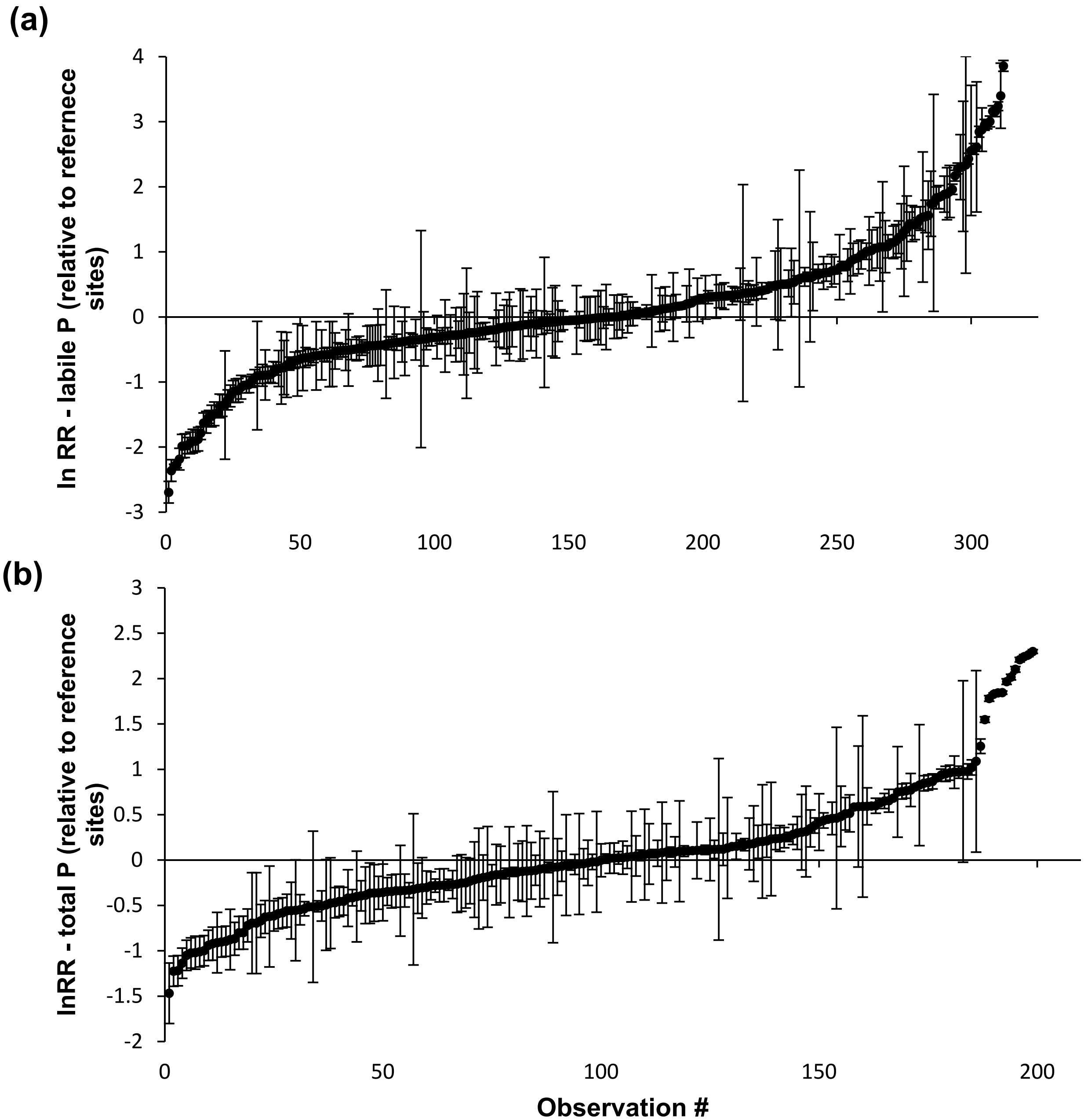
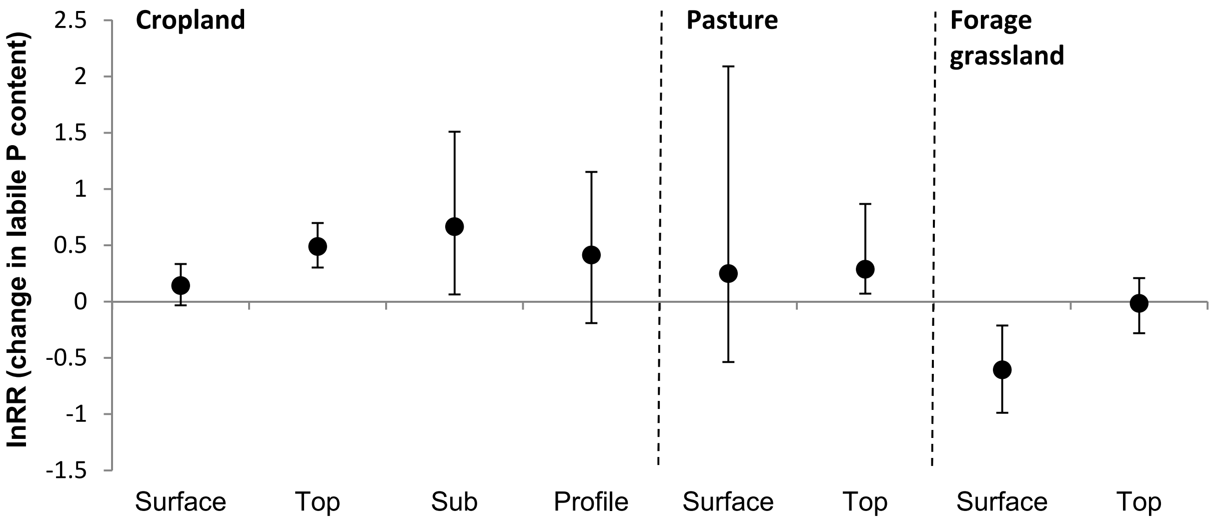


Fig. S2. Distribution of the response ratios (lnRR) for all studies included in the labile P (a) and total P (b) meta-analyses for the abandoned agriculture relative to reference ecosystem comparison (including former cropland, pasture, and forage grassland). Observations are sorted from smallest to largest effect size. Error bars show the inverse of sample size for each observation, which has been normalized using a factor of ten for ease of interpretation (10/*n*), as an approximate indicator of precision (*sensu* Hoeksema *et al.* 2010). We inferred that there was no systematic publication bias in these results attributable to sample size because similar effects occurred regardless of the degree of precision.



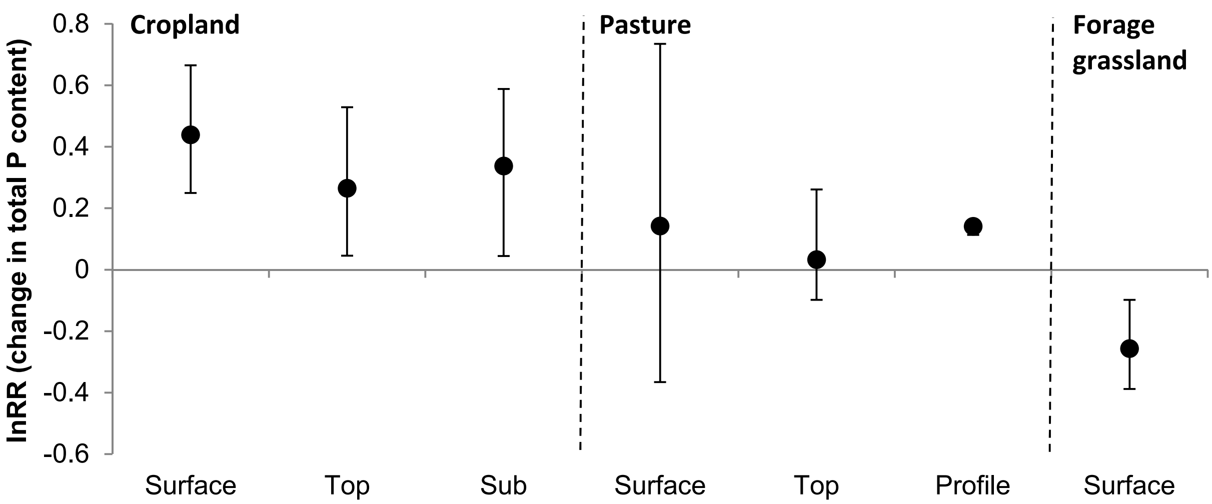


Fig. S3. Mean response ratios (lnRR) by soil depth interval for labile (top) and total P (bottom) based on comparison of abandoned agricultural areas with non-agricultural reference sites. Bootstrap bias-corrected 95% confidence intervals for each former land use type are shown. Depth intervals were classified as surface soil (0-10cm), topsoil (10 to 30cm), subsoil (>30 cm), or full profile (all depth intervals from 0 up to 150 cm).

SI References

Hoeksema, J. D., Chaudhary, V. B., Gehring, C. A., Johnson, N. C., Karst, J., Koide, R. T., Pringle, A., Zabinski, C., Bever, J. D., Moore, J. C., Wilson, G. W. T., Klironomos, J. N. & Umbanhowar, J. (2010) A meta-analysis of context-dependency in plant response to inoculation with mycorrhizal fungi. *Ecology Letters,* **13,** 394-407.

CONNECTING STATEMENT

Each of the previous chapters has explored how agriculture drives changes in P flows that could ultimately influence soil P over space, time, and across different regions. One of the main reasons to be concerned about this is the potential repercussions that altered soil P concentrations could have for water quality. While we have a solid understanding of the mechanisms that drive P loading from watersheds, complexities in how different lakes might respond to this makes the development of globally applicable models challenging.

In Chapter 5, I applied a multi-faceted statistical approach for predicting lake total P concentrations for a diverse set of lakes from unique biophysical regions across the globe. The emphasis of this study was to assess the role of anthropogenic drivers of P loading, such as agricultural land use, land management, and population density. I demonstrate how relatively coarse global estimates of various watershed characteristics could be useful for modelling eutrophication risk at the more local scale for individual lakes.

The results of Chapters 4 and 5 provide empirical understanding of how human alteration of the global P cycle can impact soil P and water quality over space and time, as well as the broader implications of these changes for ecosystem management.

CHAPTER 5: THE ROLE OF WATERSHED DRIVERS IN GLOBAL PREDICTIVE MODELS OF LAKE TOTAL PHOSPHORUS

Graham K. MacDonald, Günther Grill, Elena M. Bennett, Bernhard Lehner, Jan Kowalewski

5.0. ABSTRACT

Large-scale models of lake total phosphorus (TP) are important for understanding regional trends in freshwater quality and their management implications. However, complex processes within lakes and their watersheds that determine lake TP concentrations make global application of models derived from smaller regions challenging. We used global spatial data for a wide variety of watershed variables to predict lake TP across a set of >1000 lakes worldwide with three distinct statistical approaches (classification and regression trees (CART) with linear regression; generalized additive models (GAM); and random forest). Our multivariate models explained from 50% to as much as 79% of the variation in observed annual or seasonal TP concentrations for these lakes, with prediction error rates low enough to accurately infer lake trophic status based on common TP classifications (e.g., elevated eutrophication risk at TP >24 µg L^{-1}). Two key anthropogenic drivers (the proportion of watershed area in agriculture and watershed population density) were typically associated with elevated TP across our study lakes, but we found some disparities in the degree to which these influenced TP for lakes with different watershed biophysical characteristics. The explanatory power of global models based on these two anthropogenic predictors alone was also considerably lower (explaining 11 to 44% of the variation in lake TP). However, we found that controlling for simple metrics of climate, soil properties, and watershed morphometry (e.g., mean annual temperature, soil order, and mean watershed slope) greatly improved global model fits. Accounting for complex interdependencies among watershed biophysical characteristics and anthropogenic drivers is therefore essential for large-scale models of lake TP, and for understanding human influences on P loading across heterogeneous lakes. Our study also demonstrates the applications of global agricultural land use and population density maps that can be used in conjunction with high-resolution global hydrological, climate, and soils data to assess water quality trends at more local scales. The ability to predict TP concentrations for individual lakes across different regions using watershed

land use and biophysical characteristics derived from global data offers new avenues for water quality modelling, such as seamless global maps of eutrophication risk.

5.1. INTRODUCTION

Many lakes worldwide now face persistent eutrophication due to elevated phosphorus (P) inputs associated with agriculture and urbanization (Smith 2003; Keatley *et al.* 2011). P commonly limits freshwater primary production and is thus a key driver of algal growth rates that determine lake trophic status (Nürnberg 1996; Elser *et al.* 2007; Kosten *et al.* 2009; Wagner *et al.* 2011). There is evidence that we have passed a global tipping point in terms of freshwater eutrophication due to dramatic elevation of anthropogenic P inputs to the biosphere (Carpenter & Bennett 2011). Yet, our insight on the spatial patterns of lake eutrophication risk remains limited, in part, given lack of a broader understanding of how heterogeneous lake-watershed systems in different regions may respond to particular drivers of P loading.

Lakes are intricately connected to their drainage areas, meaning that processes occurring within watersheds are key components for both understanding and managing water quality (Soranno *et al.* 2010). At the same time, because eutrophication affects lakes around the world, there is a clear need for a global understanding of this problem (Carpenter *et al.* 1998). For these reasons, it is important to bridge our local and global understanding of the causes of eutrophication risk and its management implications.

Lake total phosphorus (TP) concentrations are influenced by both watershed processes that affect P delivery as well as the factors determining internal P cycling within a lake (Kalff 2002; Heathwaite 2010). Some of the main biophysical determinants of watershed P loading include precipitation, geology, soil characteristics, and watershed morphometry, which collectively influence the magnitude of drainage into lakes and how much P it contains (Dillon & Kirchner 1975; Nürnberg 1984; Håkanson 1995; D'Arcy & Carignan 1997; Kosten *et al.* 2009; Nõges 2009). Because of these complex determinants, lakes within different settings or regions can vary considerably in reference TP concentrations (background levels in the absence of human influences) (Dodds & Oakes 2004; Soranno *et al.* 2011)

Anthropogenic P inputs originate from both non-point (diffuse) and point (direct) sources (Carpenter *et al.* 1998). P loading from watersheds relates to the nature and intensity of land use, as well as the effects of past management practices on current soil nutrients (Kleinman *et al.* 2011). Studies of temperate lakes typically show elevation of lake TP associated with greater proportions of agriculture within watersheds, likely due to increases in watershed soil P concentrations and accelerated erosion rates linked to some forms of agriculture (Taranu &

Gregory-Eaves 2008). Anthropogenic P enrichment is often persistent due to slow flux of P from watershed soils or recycling of P from lake sediments (Carpenter 2005; Keatley *et al.* 2011; Kleinman *et al.* 2011).

The spatial pattern and specific type of agricultural land use within watersheds, such as the location of cropland or pasture relative to surface waters, are also important in determining TP (Arbuckle & Downing 2001; Fraterrigo & Downing 2008). In fact, areas that contribute most to P runoff and eutrophication can occupy a relatively small portion of total watershed area (Soranno *et al.* 1996; Pionke *et al.* 2000; Gergel 2005; Kleinman *et al.* 2006). The confluence of both biophysical context and land use means that factors such as watershed morphometry and soil drainage capacity may interact with anthropogenic drivers, such as agriculture, in determining lake TP concentrations (Fraterrigo & Downing 2008; Taranu & Gregory-Eaves 2008).

Once P enters lakes, the degree of P retention depends largely on discharge, mean lake depth, and volume, as these factors influence lakewater residence times and the degree to which the water column is mixed (Nürnberg 1984; Ahlgren *et al.* 1988; Kalff 2002). Lake TP concentrations often vary intra-annually due to seasonal differences in runoff and temperature that alter lake mixing (Nürnberg 1998). Mean lake depth alone is strongly related to TP via its role in determining water residence time (Nõges 2009), wherein shallow stratified lakes typically have higher TP concentrations than deeper well-mixed lakes and reservoirs (Nürnberg 1996; Kalff 2002).

Previous studies have provided considerable insight on watershed drivers that influence lake TP status within specific regions, but few studies have considered whether the same factors are important for lakes across multiple regions (Taranu & Gregory-Eaves 2008). Extending models from individual regions is challenging because of heterogeneity in the determinants of lake TP across systems (Kosten *et al.* 2009; Wagner *et al.* 2011) as well as variation in factors that influence how P is cycled within lakes (Huszar *et al.* 2006). Methods suitable for predicting lake TP in relatively homogenous regions are therefore generally assumed to be less useful for larger scale studies with diverse lake characteristics, landscape features, and pre-disturbance TP levels (Lamon *et al.* 2008; Cheng *et al.* 2010). The resolution, detail, and accuracy of data used in generating global models is also likely to differ from studies using local biophysical measurements and land use information, making it difficult to control for confounding factors on P loading for specific lakes. Global spatial datasets have recently been applied to studying trends in P flux from very large river watersheds (e.g., Harrison *et al.* 2010; Mayorga *et al.* 2010), but we are unaware of previous studies that have applied these to assess more local trends in lake TP (but see Keatley *et al.* 2011).

Our study examines the roles of key anthropogenic drivers and watershed biophysical characteristics for predicting lake TP across a set of more than 1000 heterogeneous lakes worldwide. We used high-resolution global hydrological data from HydroSHEDS (Lehner *et al.* 2008) in conjunction with global land use, climate, and soils data to calculate defined watershed boundaries and various landscape predictors, then applied these to model lake-specific TP concentrations. We explored the following questions: (1) How do biophysical and human drivers interact to determine lake TP across a diverse set of watersheds globally?; (2) Can we use simple landscape drivers, such as watershed agricultural land use and population density, to accurately predict lake TP even in the absence of reference TP and lake measurement data (e.g., detailed water chemistry)?; and (3) Can global scale models be used to predict lake TP despite potential inconsistencies in the roles of key drivers across diverse lakes?

We applied three distinct methods to examine both the explanatory power of different drivers and the applications of distinct statistical approaches for predicting TP across these lakes. Each approach offered unique advantages for modelling TP (i.e., controlling for reference TP, testing for nonlinearity, and accounting for complex interactions among many predictors), which allowed us to compare the accuracy of TP predictions and the relative effects of different drivers on the observed TP trends. Because our input data are derived from vastly different spatial scales, our study also sheds insight on the strength of association between relatively coarse global data and more local trends in lake TP (e.g., Keatley *et al.* 2011), as well as possible interactions among agriculture and other drivers at regional versus local contexts.

5.2. Methods

We compiled a representative global sample of lake TP observations that spanned as many biophysical regions as possible, representing two distinct sampling regimes (TP averaged over multiple seasons for an annual basis versus TP collected during a single season in a given year). We then calculated watershed-specific indicator data for key biophysical and anthropogenic drivers of TP. Using each independent TP dataset, we applied three statistical approaches to understand the influence of key predictors on TP determined by unique methods, as well as to assess the ability of these different procedures to accurately predict TP.

In particular, we used multiple statistical approaches to determine whether separate models for particular groups of lakes (Approach 1) provide a better fit than global models allowing nonlinearity among key predictors (Approach 2) or complex interactions among all available predictors (Approach 3). For Approach 1, we used a simple technique to control for reference TP by sub-setting lakes with relatively homogeneous watershed characteristics (sensu Robertson *et al.* 2006; Lamon *et al.* 2008) with classification and regression trees (CART); we then applied ordinary linear regression models (LM) to examine the direction and influence of key drivers on lake TP within each group, rather than for all lakes globally. In Approach 2, we applied flexible generalized additive models (GAMs) to examine nonlinearity and additive effects among a set of *a priori* predictors for the global lake TP dataset. Finally, in Approach 3, we tested random forest models (bootstrap aggregated regression trees) containing a large set of available predictors to account for potential complex interactions among many variables in a global model of TP. Similar approaches have been used to predict lake water quality components in regional scale analyses, particularly a study by Catherine et al. (2010), which applied linear models, GAMs, and random forest to predict algal biomass in French lakes.

5.2.1. Lake TP data compilation and watershed delineation

Contemporary lake and freshwater reservoir TP concentrations (μg L^{-1}) were compiled from various sources spanning different regions and two main sampling regimes: TP samples representing a single season (seasonal samples) or aggregated over multiple seasons in a year (annual samples). These approximate groupings correspond to single-season sampling during the temperate summer (northern or austral) and tropical dry-season, as well as lakes that were sampled at least once during each season. The main data sources were national inventories for the United States (US-EPA 2009) and China (NBSC 2008), as well as a cross-national database for the European Union (EU-EEA 2011). We also obtained data from three previously published lake meta-analysis studies representing both temperate and tropical systems (Huszar *et al.* 2006; Taranu & Gregory-Eaves 2008; Kosten *et al.* 2009). We conducted keyword searches in ISI Web of Science, Scopus, and Google Scholar to identify additional studies reporting lake TP concentrations in tabular form with geographic identifiers and sampling information (Appendix

4). Additional data was obtained from colleagues for Canadian lakes (Vermaire *et al.* 2012; Taranu *et al. In press*).

Our data search resulted in independent TP observations for several thousand lakes. We attempted to locate each lake using spatial coordinates provided by the sources and matched these to a high-resolution global spatial lake database (NGA/NASA 2003). Approximately two-thirds of all lakes had to be omitted at this stage due to missing or inaccurate spatial coordinates, although some lakes were located using annotated satellite images (Google Maps). For each georeferenced lake TP observation, we delineated the lake's watersheds using a Geographic Information System (GIS; ESRI ArcGIS, Redlands, California) and hydrological data at 15 arc second spatial resolution (Lehner *et al.* 2008; Lehner & Grill *In preparation*). Many lakes were excluded at this stage because they were located north of 60°N latitude (beyond the limits of the lake database) or because the lakes were too small to georeference with our grid-based spatial data. Some lakes were also excluded when watershed delineations were highly uncertain, for example, as a result of complex hydrological patterns or inaccuracies in digital elevation data that could have confounded our landscape analysis.

The vast majority of the lakes retained in our analysis (Figure 1) were sampled between the years 1998 and 2008 (median year of 2005). Almost all sources indicated that samples were from the epilimnion or lake surface; the US-EPA (2009) database samples were collected at a mean depth of 1.5 m. We excluded hypolimnion and lower depth samples wherever this information was available. However, a small number of lakes that were sampled only at a single depth in the top 4 m were included in order to incorporate lakes from as many biophysical settings as possible while omitting overly deep samples (4 m corresponds to the 25th percentile mean depth in the 'Annual' lakes dataset). Sources that were missing key sampling information (e.g., the season in which lake TP sampling was conducted) were omitted. Lake TP was normally calculated as the mean of numerous individual samples within each period, but there was considerable variation in the sampling intensity among sources. For lakes in the European dataset (EU-EEA 2011) that were sampled on an annual basis over more than one year, we calculated mean TP for all years to account for potential inter-annual variation, as selecting a single year of data resulted in a highly skewed distribution of TP values across all of the lakes. However, we used the standard deviation of the mean inter-annual TP to screen some large positive outlier years from these aggregate samples for individual lakes. Finally, we natural log-transformed each

TP dataset, omitting 5 outliers from the annual samples and 8 outliers from the seasonal samples to help normalize the data according to quantile-quantile (QQ) plots. Both TP sample datasets had normal distributions with minimal skewing in the tails of the distributions.

5.2.2. Predictors of lake TP

Because of uncertainty in exactly which variables are most important at the global scale for predicting TP, we tested numerous predictors that encompassed watershed climate, hydrological, topographic, and edaphic characteristics ('biophysical' setting), as well as key land use and management characteristics ('anthropogenic' drivers) (Table 1). Although we included some basic in-lake factors where available (volume and mean depth), our focus was on testing how well landscape variables predict lake TP concentrations

Simple descriptors of watershed morphometry were calculated or obtained for all lakes, including lake area, watershed area to lake area ratio (WA:LA), watershed slope, and lake discharge, which all affect water residence time and lake volume (Kalff 2002; Nõges 2009). Lake areas and mean watershed slope were derived from HydroSHEDS (Lehner *et al.* 2008), NGA/NASA (2003), and Lehner and Döll (2004). Lake discharge was calculated as an annual average at the lake outflow using runoff estimates from the WaterGAP global hydrological model for 1961-1990 (Alcamo *et al.* 2003; Döll *et al.* 2003). More direct components of watershed residence time and mixing (lake volume and mean depth) were available from sources for roughly half of the lakes, so these predictors were only included for those analyses that allow missing predictor data (i.e., CART and random forest).

We calculated predictor data that we hypothesized could influence P loading using various global land use, climate, soils and population density datasets (Table 1). These datasets were selected based on ease of acquisition, fineness of spatial resolution, or previous use in global ecological models. Anthropogenic driver data differed slightly in the years represented, but most data are representative of circa year 2000 conditions. To account for variation in the years represented within the predictor and lake TP data, we also estimated watershed mean historical population densities and the proportion of watershed area under agriculture (cropland + pasture) for the period 1700 to 2007 using global data from Klein-Goldweijk et al. (2010) and Ramankutty and Foley (1999; updated by N. Ramankutty in 2011) as metrics of long-term land use trends (Table 1). Previous studies have shown that historical land use can be important for understanding contemporary lake TP (e.g., Keatley *et al.* 2011). All biophysical data were continuous except for three categorical soil characteristics (FAO soil order, USDA soil texture class, and soil drainage class) from the Harmonized World Soil Database (FAO/IIASA 2012).

We standardized all of the landscape data to match the spatial resolution of hydrological data from HydroSHEDs (15 arc seconds; ~0.5 x 0.5 km), without further resampling. For some of the coarser resolution predictors related to agricultural management (cropland P fertilizer use, livestock manure P production, and cropland P balance; all with kg P ha^{-1} units at 0.5 degree resolution; ~50 x 50 km) we used a simple downscaling procedure to adjust these for the agricultural or land areas in each watershed. Briefly, we multiplied the original grid cell fertilizer and P balance values by the higher spatial resolution cropland fraction grid from Ramankutty et al. (2008) to obtain P yields (kg) then divided by the agricultural area of each watershed to obtain approximate kg ha^{-1} watershed rates; for livestock manure, we simply multiplied the kg P ha^{-1} values by a high resolution land area grid to obtain approximate total manure P yields (kg) for each watershed. We also estimated P yields in domestic wastewater (human population only) using a simple approach where global population density grids were multiplied by national per capita P emission estimates from van Drecht et al. (2009).

Each biophysical or anthropogenic driver variable was summarized according to watershed limits using either simple means or distance-weighting metrics. We assumed that potential spatial heterogeneity in soil characteristics (e.g., soil order, sand and clay content, drainage classes) and climate (mean annual temperature and precipitation) across watersheds would be relatively less influential on P loading, and therefore calculated these as simple mean watershed values. To account for the basic spatial pattern of land use, particularly the proximity of land use relative to a given lake, we calculated distance-weighted means for each of the anthropogenic driver predictors in Table 1 based on a procedure similar to Fraterrigo and Downing (2005) and King et al. (2005). Within each watershed and for each anthropogenic predictor, we calculated distance-weighted watershed averages using the distance of any grid cell from the lake as measured linearly along the estimated hydrological flow path:

$$WeightedAverage_f = \frac{\sum_{i=1}^{n} f_i * w_i}{\sum_{i=1}^{n} f_i}$$

where f is the variable being averaged, f_i is the value at the watershed grid cell *i,* and w_i is the inverse distance weighting (IDW) at grid cell *i*. IDW therefore determines whether a grid cell is hydrologically 'close' (receiving a higher weighting) or hydrologically 'far' (lower weighting) from a lake. This approach also helped to account for potential uncertainty in the specific dimensions of each watershed based on delineations from global hydrological data by giving more influence to land uses closer to the lake.

5.2.3. Statistical analysis

We used three statistical approaches to predict lake TP concentrations that were each applied independently using the seasonal and annual lake TP observations as the response variable (Figure 2). Each procedure is detailed in a separate section below, but we begin with a broad overview of the distinct advantages of these approaches. All statistical analysis was conducted in R v.2.14.2 (R Core Development Team, 2012).

CART is a common nonparametric method for repeated partitioning of response data into more homogeneous clusters (De'ath & Fabricius 2000). Tree-based models have the benefit of being robust under conditions of multicollinearity and informative for situations where complex nonlinear interactions exist among predictors (De'ath 2002; Faraway 2009). Regression tree models are also able to deal with observations that have missing predictor data, by passing a predictor to a lower level in the tree hierarchy until there is a significant split despite missing values (Therneau & Atkinson 2011). In Approach 3, we use these tree-based models with bootstrapping directly for prediction of TP, given the strength of this approach for prediction with complex data (Cutler *et al.* 2007); in Approach 1, we used single regression trees simply to group watersheds into relatively more homogeneous subsets.

GAMs are a form of semi-parametric regression using flexible spline smoothers that relax the assumption of linearity under linear regression (Wood 2006; Zuur *et al.* 2009), and are commonly used in ecology. There was a large degree of heterogeneity between our response and predictor data, with no linear patterns when considered globally, so GAMs offered an efficient and unbiased method to assess the functional relationship among these variables (Keele 2008).

For the LM and GAM regressions, we also use multi-model inference techniques of model averaging with information criterion measures. Multi-model inference considers a set of plausible models for prediction and inference rather than a single 'best' model, thus reducing uncertainty in parameter estimates obtained from a single arbitrary model that may or may not contain all relevant terms (Burnham & Anderson 2002). Predictors with extreme values (e.g., population density) were log-transformed where necessary depending on the type of model being used (nonparametric or linear).

Approach 1: Lake grouping with CART and subset linear models

In order to group lakes with similar biophysical settings, we first used a simple procedure to control for the effects of two indicators of anthropogenic influence on TP, agricultural extent and population density (Dodds & Oakes 2004). We chose these two drivers because they were found to be most influential in initial models, representing a compromise for a large-scale analysis. We followed the basic approach of Robertson et al. (2006) to obtain Pearson's partial residuals of lake TP controlling for these human influences, where watershed total agricultural extent (the proportion of watershed area in cropland use plus the proportion of watershed area in pasture use) and watershed population density were regressed against lake TP; a GAM was used for this regression because there was no clear evidence of a linear relationship for agriculture and population density. This is conceptually similar to Cheng et al. (2010) and Lamon et al. (2008), but they classified lakes solely according to characteristics such as mean depth and morphometry. We then used residualized lake TP as the response variable in regression trees, with key biophysical data representing various watershed characteristics as the predictors (Table 1).

We used the R package 'rpart' (Therneau & Atkinson 2012) to fit regression trees with all plausible partitions based on the biophysical predictors. We limited the partitioning to those trees where all of the 'leaves' (terminal nodes) contained at least ~5% of the data from each TP subset (32 and 20 observations for the annual and seasonal data, respectively) in order to reduce the number of splits. The need for pruning to reduce the size of each tree was assessed based on multiple runs of cross-validation, which indicated some support for unpruned trees. Because this approach was intended to sort lakes into more homogeneous groups, and not directly for prediction, we did not prune the tree obtained for the annual TP data (which had a relatively larger sample size) but pruned three splits from the seasonal data (given the relatively lower sample size).

Following the grouping of each lake with CART models, we ran separate multiple linear regression models to assess the influence of a larger set of anthropogenic drivers that we hypothesized would encompass the most influence on watershed P loading (Table 1). For each terminal node group of lakes, we checked the degree of multicollinearity among predictors and excluded terms with the highest degree of correlation until all terms in the full model had variance inflation factors (VIF) <3 (determined with the 'aed' package from Zuur *et al.* 2009). For additional local context with respect to watershed characteristics, log-transformed watershed slope and discharge in these models were also included in each model as long as these were not multicollinear with other predictors. Regression models containing all possible subsets of predictors were then generated using the R package 'MuMIn' (Barton 2011), with the maximum number of terms allowed in any model limited to four to reduce the chance of over-fitting. Model ranking was based on the adjusted Akaike Information Criterion (AIC_c). Finally, we averaged the linear model parameters across all models falling into the 95% confidence set (where Akaike confidence weights sum to 0.95), indicating a 95% likelihood of that set containing the best possible explanatory model.

We extracted standardized regression coefficients (the slope of the regression line) for each term in the 95% confidence set and pooled the predictions across each model-average set. These regression slopes indicate the direction and relative effect of each predictor on log-transformed TP, with each predictor standardized to a common scale to account for differences in units. Variable importance measures based on the sum of Akaike weights for models containing a given predictor were used to assess importance, where values >0.50 indicate a relatively important variable (Burnham & Anderson 2002).

Because we found considerable heterogeneity at the global scale in the relationship between TP and our hypothesized key anthropogenic drivers (total agricultural extent and population density), we also assessed how grouping lakes based on watershed characteristics might improve the relationships. Accordingly, we used bivariate LMs to test each of the two predictors for lakes within each CART terminal node group. Analysis of covariance (ANCOVA) was also used to test for significant differences in the intercepts and slopes for total agricultural extent and population density among the different lake groups.

Approach 2: Generalized Additive Models (GAMs)

We applied GAMs to each of our full TP datasets as an explicit test of a global model rather than subset models for lakes, as in Approach 1. We used the 'mgcv' package (Wood 2006) in R to fit spline smoothers for each of the continuous predictors with a Gaussian identity link function for normally distributed data and the degree of smoothness determined through generalized cross-validation. To reduce the likelihood of over-fitting models, cubic regression splines with 'shrinkage' penalization were used, which reduce the degree of smoothing such that a less influential term can be penalized out of the model entirely (with estimated degrees of freedom for the smooth term set equal to zero) (Wood 2006). We also use a conservative significance level cut-off ($P \leq 0.001$) that is more appropriate for GAMs (Zuur *et al.* 2009), although significance is typically robust at values $P \leq 0.02$ (Wood 2006).

For GAM model inference, we first considered the degree of correlation among predictors in each of the biophysical and anthropogenic driver categories (based on a subset of predictors chosen *a priori* to reduce overlap within categories; Table 1). We excluded any predictors with variance inflation factors >3 and Spearman's correlation coefficients >0.7, indicating high multivariate correlation, both of which can be considered fairly conservative cut-offs (Zuur *et al.* 2009; Dormann *et al.* 2012). To help avoid overly complicated models with a large number of predictors (accounted for in Approach 3), we first tested separate GAMs for the anthropogenic or biophysical predictor categories containing all possible subsets of terms in each category. Only the predictors contained in the models with the lowest absolute AIC_c were retained. The retained predictors were then checked for multivariate correlation and combined for another all-subsets model selection. All models falling within the 95% confidence set of models ($\sum$Akaike weights = 0.95), were combined to obtain model-averaged predictions.

Finally, to test the influence of geography in these models, we extended the 'non-spatial' GAM, above, by adding a smoother term for the interaction between latitude and longitude ('spatial' GAM), and conducted another all-subsets model selection. This method incorporates spatial coordinates as a fixed term used directly for prediction, rather than as a component of the model error or variance structure (Wood 2006; Dormann *et al.* 2007). Globally, the latitude and longitude coordinates were not highly correlated with any of our other predictors for these lakes, including climate (all predictors had Spearman's rho <0.7).

Approach 3: Random forest

Although simple tree-based models are effective for prediction with some data, the degree of pruning necessary to obtain optimal predictions as well as the choice among correlated variables and their locations within the tree hierarchy can be difficult to judge from a single tree (Dormann *et al.* 2012). Methods that generate many trees and aggregate results with bootstrapping (i.e., 'bagging') have therefore been used in ecology and other disciplines (e.g., Cutler *et al.* 2007; Bennett *et al.* 2008; Carlisle *et al.* 2010; Catherine *et al.* 2010) to avoid potential bias associated with single-tree predictions. One form of bagging called random forest (Breiman 2001), implemented in R with the 'randomForest' package (Liaw & Wiener 2002), which uses a random subset of data with a specified randomly selected number of predictors to build a large number of trees over which predictions can be averaged for greater accuracy (Cutler et al. 2007).

Because this random forest is more robust to multicollinearity than CART or GAMs (Dormann *et al.* 2012), we included all predictors in these models. We generated 500 regression trees based on all of the predictor data with the number of variables selected for each model set at twelve based on optimal bootstrap ('out-of-bag') performance estimates. The default random forest procedure was used to impute missing lake volume and mean depth values based on resampling the median across the whole dataset. Because random forest variable importance measures may in some cases favour imputed data (Liaw & Wiener 2002), we excluded mean depth from the seasonal data as it was found to have a disproportionate influence.

Evaluation of model performance

We assessed model predictions using three different measures related to the correlation coefficient (R^2), which describes the variation in the response explained by the predictors. First, model R^2 was assessed based on the full sets of predictor and response variables from each individual model with the lowest AIC_c from the LMs (Approach 1) and GAMs (Approach 2). We also combined the model-averaged predictions from the LM and spatial GAMs, as well as the bagged predictions from random forest (Approach 3), and fit simple regression lines through the observed and predictive values to assess the overall agreement (Piñeiro *et al.* 2008).

While the method described above is adequate to assess the explanatory power of a model based on the full predictor and response data, it may not fully assess how well a model can predict where the response variable is completely unknown (i.e., lake TP is not available to be included in the training set) (Catherine *et al.* 2010). We therefore applied cross-validation using a random subset of 80% of the observations for training the models and 20% of the observations for model validation, with 25 iterations each (implemented with the 'caret' package by Kuhn

(2008)). Cross-validation was applied individually to each of the LMs with the lowest AIC_c (Approach 1), the non-spatial GAM with the lowest AIC_c (Approach 2), and the random forest models (Approach 2). The root-mean square error (RMSE) based on the average residual distance of each predicted value from the observed data was also calculated (Kuhn 2008). This procedure helps to assess the robustness of each approach for prediction of TP in other lakes, as well as the sensitivity of our models to the specific lakes used.

Spatial autocorrelation

Given the spatial nature of our data, we assessed the degree of spatial autocorrelation in the response variables based on a Moran's *I* statistic using a search radius of eight nearest-neighbours (the eight closest neighbours for any given lake) with the R package 'spdep' (Bivand 2012). Additionally, we assessed the degree of spatial autocorrelation in the residuals of each model-averaged set after accounting for the different covariates in each model, as described below for linear models (Approach 1) and GAMs (Approach 2). Random forest models are theoretically less susceptible to spatial autocorrelation because they use a randomized bootstrap sample of data (Cutler *et al.* 2007; Dormann *et al.* 2012).

5.3. RESULTS

Our global analysis included lakes from across a broad spatial gradient with highly heterogeneous biophysical conditions, land use settings, and trophic statuses (Figure 1). Lake TP varied from 3.4 µg L^{-1} to 941 µg L^{-1}, with a median of 31.6 µg P L^{-1} for those lakes sampled on an annual basis. For lakes sampled during a single season (i.e., summer or dry season), TP varied from 0.5 µg L^{-1} to 1689 µg L^{-1}, with a median of 24.3 µg L^{-1}. The mean annual temperature (MAT) of lake locations varied from -6 to 27°C (median of ~7°C) over a range of altitudes (mean watershed elevation -1 to 3000 m). Watersheds ranging from 0 to 100% urban or agriculture were represented (mean ~14% urban and ~34% agricultural, differing little between the two TP datasets). Lakes in the seasonal data set were typically smaller and shallower than the annually sampled lakes (median lake area of 1.2 km^2 and overall mean depth of 9 m for the seasonal versus median lake area of 2.9 km^2 and 15 m mean depth for the annual lakes). Watersheds with predominantly loamy soil textures were most common for all of our study lakes.

Model predictive performance across the three approaches was relatively high when using both watershed biophysical and anthropogenic driver predictors, with the R^2 between the observed and predicted values ranging from a low of 0.50 to a high of 0.79 (Figure 3). In contrast, simple bivariate models considering only the effects of the two major anthropogenic drivers (proportion of watershed in agriculture and watershed population density) had considerably lower explanatory power (average R^2 across the three approaches of 0.19±0.06 for the annual lake TP observations and average R^2 of 0.39±0.04 for the seasonal observations, with standard deviations). These differences in model fits indicate that there are interdependencies between watershed biophysical characteristics and anthropogenic drivers in determining lake TP.

For each of the three approaches, we found broadly consistent results in terms of the most influential predictors of TP for models that incorporated both anthropogenic drivers and biophysical characteristics. Climate and soil type were typically the most influential biophysical determinants of lake TP, while watershed cropland extent, pasture extent, and population density were the most influential anthropogenic drivers across the three modelling approaches. However, differences in both the regression slopes and intercepts for lakes with particular watershed characteristics (Approach 1) indicates that some groups of lakes have different relationships with anthropogenic drivers than others.

5.3.1. Approach 1: Lake TP trends across CART subsets and LM predictions

Soil type and climate were the main biophysical variables that divided lakes across both the annual and seasonal datasets in the regression trees (Figures 4A and 4B). Lower level divisions of lakes occurred based on watershed to lake area ratio (WA:LA) (annual samples), watershed slope (seasonal samples), and soil texture (both sets), which indicates potential interactions between these and either climate or soil type. The regression trees for annually sampled lakes show the particular influence of mean annual precipitation (MAP), which divided the lakes at 72 mm per month, as well as lower level divisions on mean annual temperature (MAT) that separated some temperate, tropical, and relatively arid lakes. The seasonal lake TP observations, which are clustered in central Europe and North America, showed more prominent divisions according to soil orders that possibly account for variation at the sub-regional level. Boxplots at the base of each tree show the distribution of residualized TP (controlling for the effects of agriculture and population density) in each terminal node; these indicate the degree to which lakes in each subset were influenced by agriculture and population density alone (e.g., lakes were highly influenced where partial residuals had a median value close to zero). Most of the lakes in each terminal node encompassed a broad latitudinal and climate gradient (Table 4).

Standardized linear regression slopes from across the terminal node models and model-averaged 95% confidence sets show the change in log-transformed TP for every per unit change in a predictor (Figures 5A and 5B). Our results reveal some variability in the direction and importance of the key anthropogenic drivers considered here; however, population density, cropland extent, and pasture extent showed typically positive and relatively steep slopes within both the annual and seasonal lake TP measurement data. Population density was the most important driver overall for the annual lake TP samples (∑Akaike weights >0.50 for 9 out of 12 terminal node models). In contrast, watershed pasture extent was the most consistently important driver for the seasonal lake samples (∑Akaike weights >0.50 for 6 out of 8 models), followed by population density and cropland extent. The three remaining anthropogenic drivers included in these models (cropland fertilizer use, total manure P production, and mean historical agriculture extent) showed less consistent directionality in the relationship with TP, but were nonetheless considered relatively important variables in several models. Log-transformed mean watershed slope showed a typically weak negative relationship with TP across several of the individual models.

Linear model interactions for agriculture and population density

In total, 8 of the 12 annual models showed a significant (P<0.05) bivariate relationship between TP and total agricultural extent (cropland plus pasture) (Figure 6A), while 7 of the 8 models had a significant relationship for the seasonal data (Figure 6B). However, there was considerable variation in the explanatory power and significance of these bivariate models (R^2 ranged from 0.01 to 0.44).

The interaction effect of the terminal node groups with total agriculture was barely significant (ANCOVA P=0.05 for both data sets), indicating that the slope of the relationship was not overly different between groups. In contrast, there was a significant interaction of terminal node model with population density (not shown; ANCOVA P<0.0001 for both datasets). However, both agriculture and population density had significantly different intercepts among groups (P <0.0001), which may be indicative of different baseline TP between groups of lakes based on biophysical setting (Taranu and Gregory-Eaves, 2008).

5.3.2. Approach 2: Global predictions from GAMs

At least five TP predictors were highly significant ($P \leq 0.001$) using the annual samples (WA:LA ratio, MAT, population density historic agriculture extent, and the spatial interaction term; Figure 7A). At least four predictors were highly significant for the seasonal TP samples (watershed slope, population density, the spatial interaction term, as well as the categorical soil order term; Figure 7B). These fitted curves control for the global additive effects of each predictor as well as the interaction between latitude and longitude in the annual and seasonal TP models with the lowest overall AIC_c. A less conservative cut-off (P<0.01) suggests that four additional predictors had some evidence of a significant effect (WA:LA ratio, MAP, cropland P fertilizer use, and livestock manure production) in either the annual or seasonal TP sets (Figures 7A and 7B).

Most of the predictors considered indicated at least some degree of nonlinearity (estimated degrees of freedom >1.0). Relatively wide confidence bands were present for some curves, indicating comparably poor explanatory power in spans with fewer data points (e.g., high IDW livestock manure P production values). Other terms not shown in the figures were either penalized out of the models, excluded based on high degrees of multivariate correlation, or had poor explanatory power based on AIC_c.

Adding spatial coordinates (latitude and longitude) to the GAMs increased the predictive power considerably, with an ~16% increase in the variation explained for both the annual and seasonal measurements (compare R^2 for the spatial and non-spatial GAMs in Tables 2 and 3). Additionally, the AIC_c was considerably lower ($\Delta AIC_c = 30$) for both spatial GAMs compared to the non-spatial GAMs, which can be considered a highly significant improvement in fit (Burnham and Anderson, 2003). However, even though none of the predictors considered were strongly correlated with latitude and longitude coordinates (Spearman's rho <0.7), adding the spatial interaction term did reduce the overall significance of some of the other predictors. For example, MAP was highly significant (P<0.001) in the non-spatial GAMs for both the annual and seasonal TP observations. Adding spatial coordinates may therefore account for some of the effects of other terms, resulting in relatively lower significance for these predictors (Figures 7A and 7B).

5.3.3. Approach 3: Global predictions from random forest

Aggregating hundreds of regression trees allowed for a large number of predictors to be considered simultaneously in our global models. Only MAT, MAP, and cropland extent had random forest variable importance measures >50% for both the annual and seasonal data sets (Figures 8A and 8B). This indicates that these three variables considerably improved the accuracy of the TP predictions across the regression trees for which they were selected. Watershed slope and population density were relatively important variables for the annual TP data only, while pasture and historic agricultural extent were relatively more important for the seasonal TP data. In contrast, population density was considerably less important for the seasonal lakes and had a relatively lower importance than more detailed or synthetic indicators of population pressure, such as distance-weighted P yield in wastewater from human populations (Figure 8B). Overall, various predictors describing different soil characteristics were slightly more influential for the seasonal than annual TP samples (e.g., soil organic carbon content, soil texture class, as well as soil sand and clay contents). Both random forest models showed lower importance values for the land management (e.g., fertilizer use) and lake morphometric variables (e.g., catchment to lake area) than more broad factors such as land cover and climate.

We also examined the marginal effects of four key predictors in the random forest models (MAT, MAP, watershed slope, cropland, and population density) controlling for the average effect of all other predictors (Figure 9). This shows that small increases in cropland extent and temperature had particularly strong positive effects on lake TP when controlling for other variables. For example, there was a sharp rise in TP for watersheds that had distance-weighted cropland proportions >0.10. Although there was also a sharp increase in lake TP with small initial increases in distance-weighted population density for the annual lakes, this strong threshold effect was less prominent for the seasonal lakes. Similarly, TP declined sharply for both data sets as watershed slope increased, which also supports the results from the GAM models (Figure 7). Random forest models also showed possible interactions between MAT and MAP, wherein annual and seasonal TP increased as temperature rose and precipitation fell.

5.3.4. Evaluation of the three statistical approaches

Comparison of global predicted versus observed TP indicates that the spatial GAMs provided considerably better overall fits (R^2 = 0.79) than the CART/LM or random forest approaches (R^2 =

0.72 and 0.58, respectively) for the seasonally sampled lakes (Figure 3). There was little difference in the strengths of the fits between the CART/LM and spatial GAMs for the lakes sampled annually, and the overall R^2 values were more comparable to random forest (R^2 ranged from 0.50 to 0.59). The global LM and GAM fits were improved using model-averaged parameter values for the predictors that fell into the 95% confidence sets (non-model averaged predictions are not shown, but often had slightly lower R^2).

Overall, all of the models had relatively low absolute root-mean square errors in terms of untransformed predicted TP (1.73 to 2.89 μg P L^{-1}) given the very wide range in TP among the 1010 lakes studied here (see Tables 2 and 3 for LM results specific to each terminal node group, the non-spatial GAMs, and random forest models). However, random forest models had slightly more scatter in the predictions for some lakes around the 1:1 (which indicates a perfect fit with the observed values; Figure 3).

Cross-validation helped to understand how predictive capacity was influenced by the specific lake TP observations used to train the models (Tables 2 and 3). Random forest was the most robust approach based on cross-validation, with no change in the R^2 between the full and training data subsets. In contrast, some of the linear models had considerable deviation in R^2 depending on which subset of training data was used. The variation in lake TP explained by the non-spatial GAMs was similarly 8 to 12% lower than when predicting from the full dataset based on the cross-validation subsets, suggesting that there could be slight inflation in the predictive power of the spatial GAMs using the full dataset over a predictive model where lake TP is completely unknown. The specific subsets of points chosen to fit these models may therefore influence the accuracy of the predicted results. However, cross-validation of the LM and GAM models was based on predictors from a single model with the lowest respective AIC_c, given computational constraints, whereas we used model-averaged parameters from the 95% confidence sets for prediction that may be subject to lower uncertainty (Figure 3).

5.3.5. Spatial autocorrelation

We found moderate positive spatial autocorrelation in our response variables, with Moran's *I* of 0.42 and 0.53 (both $P<0.0001$) for the annual and seasonal observations, respectively. This indicates that lakes with high or low TP were clustered in certain areas. The methods used here did not explicitly address the variance structure of these models in order to reduce spatial autocorrelation in the response variable (Dormann *et al.* 2007). However, the predictors included in the spatial GAMs, sub-setting of lakes into more homogeneous groups (CART/LM), and use of model-averaged parameter estimates eliminated or greatly reduced autocorrelation in the model residuals that could confound model inference. Only three of the twenty linear models tested contained significant spatial autocorrelation in the model-averaged residuals (Moran's $I \leq 0.15$, $P<0.05$). Similarly, while the non-spatial GAM models had some residual spatial autocorrelation (Moran's I of 0.14 and 0.05, both $P<0.01$), significant autocorrelation was absent following the addition of the spatial interaction term ($P>0.05$).

5.4. DISCUSSION

Each of our three independent modelling approaches suggest that there is an important interplay between watershed biophysical context (climate, soil type, and watershed slope) and anthropogenic drivers (agricultural extent and population density) for determining TP across these globally distributed lakes. We consistently found important roles of watershed agricultural extent, population density, precipitation, temperature, watershed slope, and either soil order or soil texture for each approach and for both lake TP datasets. The proportion of agriculture and population density in a watershed typically had a positive relationship with TP in our study. In particular, we found strong positive relationships between TP and watershed cropland extent, pasture extent, or both (Figures 5 and 9) as well as a strong curvilinear relationship between TP and mean historic agricultural extent (Figure 7A). However, models that contained several anthropogenic and biophysical predictors indicated far better overall fits than simple bivariate models containing watershed agricultural extent and population density alone.

Global predictive models of lake TP based on watershed data appear to be feasible, even without controlling for reference TP conditions (Approaches 2 and 3). Our results indicate that it is possible to obtain reasonably accurate predictions of TP for lakes across these distinct regions by using only anthropogenic drivers and simple indicators of watershed characteristics (Figure 3), despite the myriad determinants of P cycling within lakes that are not considered in our study (Kalff 2002). At the same time, inconsistency or heterogeneity observed in the TP relationships with some key anthropogenic and biophysical variables are clearly displayed in our results. In particular, we found evidence that some lakes responded differently to increased watershed population density and agricultural extent. Accounting for inherent differences in lake TP due to watershed biophysical characteristics (e.g., Approach 1), considering nonlinear additive effects (Approach 2), and allowing complex interactions among as many predictors as possible (Approach 3) offered considerably greater explanatory power for assessing trends in lake TP relative to other large-scale studies that focussed on either anthropogenic (Taranu & Gregory-Eaves 2008) or biophysical factors (Liu *et al.* 2011). One explanation of this is that human influence and various biophysical landscape characteristics act together to mediate lake TP, which has been described as "landscape filtering" (Heathwaite 2010).

Our TP prediction error rates are typically within the ranges necessary to estimate lake trophic status using generalized limits, such as ~30 to 100 µg TP L^{-1} for eutrophic lakes (Nürnberg 1996; Rast & Thornton 2004). Median observed lake TP concentrations for both the annual and seasonal samples were slightly above the common water quality target of 24 µg TP L^{-1}, considered by Carpenter and Bennett (2011) as a global boundary between mesotrophy and eutrophy due to anthropogenic P loading. Given that our predicted median TP values were within 0.3 to 4.4 µg L^{-1} of the observed global median TP, each of our models should enable relatively accurate classification of lakes at risk of eutrophication.

5.4.1. Agriculture and population density as drivers of lake TP

Our findings collectively highlight the positive relationship between population density and TP across the >1000 lakes in this study. Significant differences in the population density regression slopes among lake groups derived from CART models (ANCOVA with $P<0.0001$) suggests that lakes in some settings may respond differently to increases in population (Figure 5). Major efforts have been undertaken in recent decades to reduce point-source wastewater P inputs to surface waters, especially in more developed regions (Carpenter *et al.* 1998). Disparities in P removal via wastewater treatment among urban watersheds in different countries (Van Drecht *et al.* 2009), or rural watersheds subject to P leakage from residential septic systems (Muscutt & Withers 1996), may therefore account for some differences in population density effects. This is supported by our finding that P yield in wastewater was more important than population density alone for lakes with seasonal TP observations (Figure 8B). However, more urban watersheds may simply have elevated P runoff due to greater impervious surface areas (Soranno *et al.* 1996; Carpenter *et al.* 1998; Fraterrigo & Downing 2008).

A meta-analysis by Taranu and Gregory-Eaves (2008) found that the proportion of agriculture in a watershed explained 28% of the variation in TP across 358 temperate lakes, but with some variation in the magnitude of the effect for individual studies. We found roughly the same explanatory power of total agriculture for seasonally sampled lakes (average R^2 of 0.27 ±0.13 across 8 models), but a lower explanatory power for the annually sampled lakes (average R^2 of 0.13 ±0.12 across 12 models). This may relate to whether pasture or cropland is more prevalent in the watershed, as we found some difference in the influence of these among different lake groups (Figure 5A).

We also observed an interesting potential interaction in some lakes' responses to population and agriculture. For example, at least five groups of lakes had weak relationships between watershed cropland extent and TP (where cropland was considered a relatively unimportant predictor), but stronger relationships with population density (where population density was considered a relatively important predictor) (Figures 5A and 5B). As urbanization occurs, the watershed area in agriculture may simply be reduced, which would make agriculture appear to be a less important driver for TP overall.

Some of the patterns we observed may relate to inherent geographic differences in the lakes represented within our annual and seasonal lake TP analyses, as well as clustering of lakes within certain regions due to data availability. For example, lake locations in our study with annual TP observations were more likely to have both relatively high watershed population densities and high proportions of agriculture, but population had a stronger overall effect on TP for these lakes. Lakes with seasonal TP samples were typically smaller and shallower (roughly half the median size of the annually sampled lakes), but the geographic distribution of lakes in each set also correspond to important inter-regional differences in land use that may affect the lake trophic statuses represented in our study (for example, we have little representation of lakes in Central Asia, Africa, and Oceania, as well as no lakes north of 60° latitude; Figure 1). Average watershed population density for the lakes falling in the annual subsets was almost three times higher than for the seasonally sampled lakes; however, the two lake datasets had minimal differences in current agricultural extent (~3% difference), mean historic agricultural extent (~8%), and urban land cover extent (~1.5%). Although agriculture theoretically influences TP in both lake sets in a similar way after controlling for biophysical setting (Figure 8A and 8B), population density may have a greater influence on the lakes in the annual subset simply because these lakes are in more densely populated settings. The importance of pasture relative to cropland in the seasonal TP set also indicates the need to account for different types of agricultural land use, given that pasture is much more widespread globally than cropland (Ramankutty *et al.* 2008).

5.4.2. Incorporating watershed context and interactions among predictors

Inherent differences in reference TP related to regional and local context (Soranno *et al.* 2011) could influence how we interpret the relative influence of key predictors, such as agriculture and population density. In our study, we addressed this by grouping lakes according to relatively homogenous watershed climate and soil types (Approach 1). Our finding that there were significantly different intercepts of the regression lines for both the agriculture and population density ANCOVA tests (Figure 6) indicates that lakes among specific groups may differ in their reference TP in the absence of these human pressures (Dodds & Oakes 2004; Taranu & Gregory-Eaves 2008). While our global model results appear to be robust to this heterogeneity, some of the GAM curves showed relatively weaker than expected relationships with TP (e.g., those representing agricultural extent; Figures 7A and 7B), which is likely attributable to these differences in reference TP.

Our study confirms the important role of geography in global lake TP models (Figures 1 and 7), both in terms of the inter-regional differences, but also with respect to how local biophysical context varies within regions. While our CART method typically grouped nearby lakes together, resulting in some spatial clustering, these groupings were not based solely on geography. Most subsets encompassed a wide range of latitudes and climates (see Table 4), and approximately half of the subsets included lakes from multiple continents. This supports previous lake water quality studies that have shown the importance of using models that allow for flexibility in model parameterization for different lake systems or landscape settings, not just based on geographic regions (Cheng *et al.* 2010; Wagner *et al.* 2011).

Soil characteristics (i.e., soil order, texture, drainage class, and organic matter content) were also found to be relatively influential in each of our analyses. Soil order describes features of both soil parent material and geology, which are known to influence inherent lake TP concentrations (Dillon & Kirchner 1975; Kosten *et al.* 2009). Both CART models (Figures 4A and 4B) as well as the random forest model for the seasonal TP observations (Figure 8B) suggested that soil texture (e.g., relative sand or clay content) and soil drainage were influential on TP. Soil texture and drainage are likely to influence erosion rates, the magnitude of P-sorption to soil particles, as well as hydrological factors that affect the magnitude of runoff (Kalff 2002; Fraterrigo & Downing 2008; Diebel *et al.* 2009).

Explicit consideration of interactions or additive effects among numerous predictors may help to address complexities in lake TP determinants at the global scale. We found evidence of potentially complex nonlinear interactions or even thresholds in the relationships between some watershed biophysical characteristics and anthropogenic drivers for predicting lake TP. For example, our CART model results using residualized TP typically displayed interactions between either climate and soils or climate and watershed morphometry, which was corroborated by using a very large number of regression trees and all available predictors with random forest. Both the GAM and random forest approaches indicated a strong negative and nonlinear effect of mean watershed slope on TP (Figures 7 and 9); this finding is somewhat counterintuitive, as we might expect watershed slope to increase potential for P runoff, but this could be explained by the fact that lakes in settings with strong topographic relief are also less likely to be influenced by either urban or agricultural land use. Additionally, we found a strong divergence in the effects of MAT and MAP based on random forest models. A possible explanation for this may be that lakes in more arid settings are likely to have higher inherent TP and thus lower degrees of biomass P-limitation (Williams 2004).

5.4.3. Global eutrophication models and spatial data

Our findings strongly support the applications of global spatial data, including hydrological, climate, soils, agriculture, and population, in describing lake TP trends. This is despite mismatches in the resolution of some global datasets relative to the size of the watersheds considered here. For example, the median watershed areas for our annual and seasonal lakes are 32.6 km^2 and 106.9 km^2, which indicates roughly 1:3 and 1:1 ratios with the resolution of the cropland extent grids from Ramankutty et al. (2007) for our seasonal and annual TP data, respectively. That we found a relationship between the global cropland data and lake TP is surprising given that the global cropland data contains tens of thousands of individual grid cells that are each roughly 100 km^2 in size (grid areas decrease with distance from the equator). This may, in part, confirm the importance of regional context as an important determinant of local lake water quality (Soranno *et al.* 2010; Fergus *et al.* 2011).

Although global spatial data describing agricultural extent and population density were typically related to lake TP in the way that we would hypothesize, we found somewhat less consistent effects using other agriculture-related predictors with coarser resolutions. For example, we observed negative relationships between lake TP and cropland P fertilizer use for some groups of lakes in Approach 1 (Figure 5). Possible explanations for this include mismatches in the resolution of data (Table 1), redundancy in the explanatory power of agricultural land cover and land management at more local levels in our data set, as well as the fact that some global data representing land management factors is derived using broader national or relatively coarse sub-national data to derive spatially distributed estimates (Potter *et al.* 2010; MacDonald *et al.* 2011). Land management data representing conditions for a single year may also offer less insight on cumulative water quality implications than longer-term management data (MacDonald & Bennett 2009).

Our hydrologically-based spatial framework will enable application of these models to hundreds of thousands of lakes globally, allowing for detailed mapping of lake eutrophication risk across regions. In particular, high-resolution hydrological data from HydroSHEDS (Lehner *et al.* 2008) can be used to calculate detailed watershed boundaries globally, which is essential for hydrologically meaningful assessment of the potential landscape drivers of P loading for individual lakes. We tested various relatively simple surrogates of watershed morphometry (watershed slope, lake area), the degree of lake 'flushing' (WA:LA ratio), and water residence time (lake discharge) using these hydrological data. Lake area and WA:LA ratio were selected in both CART models (Figures 5A and 5B), and WA:LA ratio had a positive effect on TP based on the GAMs (Figure 7A and 7B), which agrees with established models regarding elevation of TP concentrations when lake surface area is relatively small (Ahlgren *et al.* 1988; Håkanson 1995). Similarly, the consistently negative effect of increasing watershed slope on TP across all of our models could be a result of increased runoff resulting in shorter water residence times. The development of more detailed estimates of lake volume and water residence time globally using high-resolution digital elevation data (Hollister *et al.* 2011) is therefore likely to greatly improve our ability to model P runoff.

5.4.4. Insights from different statistical approaches

Overall, we found that all three approaches to predicting lake TP offered distinct advantages, although the specific lakes used to train a model can influence the R^2 considerably. Data sub-setting based on landscape setting clearly helped to reduce heterogeneity in conditions (Approach 1), and largely supports the view that in larger scale analyses, not all systems will respond to landscape drivers of nutrient loading in the same way (Wagner *et al.* 2011). While we apply a fixed modelling framework in Approach 1 because we were interested in how the role of key predictors differed among separate groups of lakes, possible extensions of this approach include applications of hierarchical and mixed models where global models are applied but with variability in parameter estimates among lake groups (Taranu & Gregory-Eaves 2008). For example, several recent studies have applied Bayesian hierarchical models that account for intersystem parameterization based on covariates that are known to vary across lakes (Lamon *et al.* 2008; Cheng *et al.* 2010; Wagner *et al.* 2011).

Less parametric global modelling approaches, such as GAMs and random forest (Approaches 2 and 3) may be better suited to dealing with heterogeneity in the relationship between landscape drivers and lake TP globally when explicit incorporation of background nutrient levels is not available. Random forest in particular appears to be a robust method that could be easily extended to incorporate additional lakes with unknown TP resulting in reasonable predictions except for at the highest and lowest TP values, especially because it can handle such a large number of potentially correlated predictors (Cutler *et al.* 2007; Dormann *et al.* 2012). We also found that model averaging using multi-model inference (Burnham & Anderson 2002) reduced some of the uncertainty in our models by incorporating all predictors that had some level of support rather than using a single 'best' model.

5.5. Conclusions

While watershed agricultural extent and population density typically had positive influences on lake TP, there is some variation in the particular role these key drivers play. Differences in the strength or direction of the relationship between lake TP and anthropogenic drivers for some groups of relatively more homogenous lakes illustrates that watershed biophysical context can have a confounding influence on large-scale models of lake TP concentrations. Accounting for potential interdependencies among many predictors of lake TP, such as the interactions between biophysical context (especially climate and soil type) and anthropogenic drivers (e.g., agriculture and population density), allows for more broadly applicable models with relatively robust predictions of TP even across diverse lakes from many regions. Simple surrogates of watershed morphometry, particularly watershed area to lake area ratio (WA:LA) and mean watershed slope, also offer critical insight on lake drainage and P runoff that can easily be applied to a very large number of lakes.

Global spatial data describing key watershed anthropogenic drivers and various biophysical characteristics offers important opportunities for detailed, spatially explicit modelling of lake water quality. Extension of these predictive models to understand spatial patterns of lake eutrophication risk globally is a promising avenue for future research that will provide critical new insight on the ecological implications of the modern human-dominated P cycle (Carpenter and Bennett, 2011).

5.6. Acknowledgements

We thank Matthew Mitchell for detailed comments on a previous draft of this chapter. Data contributions from Zofia Taranu, Jesse Vermaire, and Vera Huszar were greatly appreciated. Alex MacDonald provided helpful feedback on the statistical approach. GKM and EMB received funding from the Natural Sciences and Engineering Research Council of Canada (NSERC).

5.7. References

Ahlgren, I., Frisk, T. & Kamp-Nielsen, L. (1988) Empirical and theoretical models of phosphorus loading, retention and concentration vs. Lake trophic state. *Hydrobiologia,* **170,** 285-303.

Alcamo, J., Döll, P., Henrichs, T., Kaspar, F., Lehner, B., Rösch, T. & Siebert, S. (2003) Development and testing of the watergap 2 global model of water use and availability. *Hydrological Sciences Journal,* **48,** 317-337.

Arbuckle, K. E. & Downing, J. A. (2001) The influence of watershed land use on lake n:P in a predominantly agricultural landscape. *Limnology and Oceanography,* **46,** 970-975.

Barton, K. (2011) *Mumin: Multi-model inference 1.6 for R version 2.14.2. Http://r-forge.R-project.Org/projects/mumin/*

Bennett, E., Carpenter, S. & Cardille, J. (2008) Estimating the risk of exceeding thresholds in environmental systems. *Water, Air, & Soil Pollution,* **191,** 131-138.

Bivand, R. (2012) Spdep: Spatial dependence: Weighting schemes, statistics and models, v. 0.5-46. R package.

Breiman, L. (2001) Random forests. *Machine Learning,* **45,** 5-32.

Burnham, K. P. & Anderson, D. R. (2002) *Model selection and multimodel inference: A practical information-theoretic approach*. Springer Verlag, New York, NY.

Carlisle, D. M., Falcone, J., Wolock, D. M., Meador, M. R. & Norris, R. H. (2010) Predicting the natural flow regime: Models for assessing hydrological alteration in streams. *River Research and Applications,* **26,** 118-136.

Carpenter, S. R. (2005) Eutrophication of aquatic ecosystems: Bistability and soil phosphorus. *Proceedings of the National Academy of Sciences of the United States of America,* **102,** 10002-10005.

Carpenter, S. R. & Bennett, E. M. (2011) Reconsideration of the planetary boundary for phosphorus. *Environmental Research Letters,* **6,** 014009.

Carpenter, S. R., Caraco, N. F., Correll, D. L., Howarth, R. W., Sharpley, A. N. & Smith, V. H. (1998) Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecological Applications,* **8,** 559-568.

Catherine, A., Mouillot, D., Escoffier, N., Bernard, C. & Troussellier, M. (2010) Cost effective prediction of the eutrophication status of lakes and reservoirs. *Freshwater Biology,* **55,** 2425-2435.

Cheng, V., Arhonditsis, G. & Brett, M. (2010) A revaluation of lake-phosphorus loading models using a bayesian hierarchical framework. *Ecological Research,* **25,** 59-76.

Cutler, D. R., Edwards, T. C., Jr., Beard, K. H., Cutler, A., Hess, K. T., Gibson, J. & Lawler, J. J. (2007) Random forests for classification in ecology. *Ecology,* **88,** 2783-2792.

D'Arcy, P. & Carignan, R. (1997) Influence of catchment topography on water chemistry in southeastern québec shield lakes. *Canadian Journal of Fisheries and Aquatic Sciences,* **54,** 2215-2227.

De'ath, G. (2002) Multivariate regression trees: A new technique for modeling species-environment relationships. *Ecology,* **83,** 1105-1117.

De'ath, G. & Fabricius, K. E. (2000) Classification and regression trees: A powerful yet simple technique for ecological data analysis. *Ecology,* **81,** 3178-3192.

Diebel, M., Maxted, J., Robertson, D., Han, S. & Vander Zanden, M. J. (2009) Landscape planning for agricultural nonpoint source pollution reduction iii: Assessing phosphorus and sediment reduction potential. *Environmental Management,* **43,** 69-83.

Dillon, P. J. & Kirchner, W. B. (1975) The effects of geology and land use on the export of phosphorus from watersheds. *Water Research,* **9,** 135-148.
Dodds, W. K. & Oakes, R. M. (2004) A technique for establishing reference nutrient concentrations across watersheds impacted by humans. *Limnology and Oceanography Methods,* **2,** 333-341.
Döll, P., Kaspar, F. & Lehner, B. (2003) A global hydrological model for deriving water availability indicators: Model tuning and validation. *Journal of Hydrology,* **270,** 105-134.
Dormann, C. F., Elith, J., Bacher, S., Buchmann, C., Carl, G., Carré, G., Marquéz, J. R. G., Gruber, B., Lafourcade, B., Leitão, P. J., Münkemüller, T., McClean, C., Osborne, P. E., Reineking, B., Schröder, B., Skidmore, A. K., Zurell, D. & Lautenbach, S. (2012) Collinearity: A review of methods to deal with it and a simulation study evaluating their performance. *Ecography***,** *In press*.
Dormann, C. F., M. McPherson, J., B. Araújo, M., Bivand, R., Bolliger, J., Carl, G., G. Davies, R., Hirzel, A., Jetz, W., Daniel Kissling, W., Kühn, I., Ohlemüller, R., R. Peres-Neto, P., Reineking, B., Schröder, B., M. Schurr, F. & Wilson, R. (2007) Methods to account for spatial autocorrelation in the analysis of species distributional data: A review. *Ecography,* **30,** 609-628.
Elser, J. J., Bracken, M. E. S., Cleland, E. E., Gruner, D. S., Harpole, W. S., Hillebrand, H., Ngai, J. T., Seabloom, E. W., Shurin, J. B. & Smith, J. E. (2007) Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecology Letters,* **10,** 1135-1142.
EU-EEA (2011) Waterbase - lakes dataset version: May 2011. European Environment Agency, Copenhagen, Denmark.
FAO/IIASA (2012) Harmonized world soil database (version 1.2; FAO/IIASA/ISRIC/ISSCAS/JRC). United Nations-FAO, Italy and IIASA, Laxenburg, Austria.
Faraway, J. (2009) *Linear models with R*. Chapman & Hall/CRC, Boca Raton.
Fergus, C. E., Soranno, P. A., Cheruvelil, K. S. & Bremigan, M. T. (2011) Multiscale landscape and wetland drivers of lake total phosphorus and water color. *Limnology & Oceanography,* **56,** 2127-2146.
Fraterrigo, J. & Downing, J. (2008) The influence of land use on lake nutrients varies with watershed transport capacity. *Ecosystems,* **11,** 1021-1034.
Gergel, S. E. (2005) Spatial and non-spatial factors: When do they affect landscape indicators of watershed loading? *Landscape Ecology,* **20,** 177-189.
Håkanson, L. (1995) Models to predict lake annual mean total phosphorus. *Journal of Aquatic Ecosystem Stress and Recovery (Formerly Journal of Aquatic Ecosystem Health),* **4,** 25-58.
Harrison, J. A., Bouwman, A. F., Mayorga, E. & Seitzinger, S. (2010) Magnitudes and sources of dissolved inorganic phosphorus inputs to surface fresh waters and the coastal zone: A new global model. *Global Biogeochem. Cycles,* **24,** GB1003.
Heathwaite, A. L. (2010) Multiple stressors on water availability at global to catchment scales: Understanding human impact on nutrient cycles to protect water quality and water availability in the long term. *Freshwater Biology,* **55,** 241-257.
Hijmans, R. J., Cameron, S. E., Parra, J. L., Jones, P. G. & Jarvis, A. (2005) Very high resolution interpolated climate surfaces for global land areas. International Journal of Climatology, 25, 1965-1978.

Hollister, J. W., Milstead, W. B. & Urrutia, M. A. (2011) Predicting maximum lake depth from surrounding topography. *PLoS ONE,* **6,** e25764.

Huszar, V. L. M., Caraco, N. F., Roland, F. & Cole, J. (2006) Nutrient-chlorophyll relationships in tropical-subtropical lakes: Do temperate models fit? Biogeochemistry, 79:239-250.

Nitrogen cycling in the americas: Natural and anthropogenic influences and controls. 239-250.

Kalff, J. (2002) *Limnology: Inland water ecosystems*. Prentice-Hall., Upper Saddle River, NJ.

Keatley, B. E., Bennett, E. M., MacDonald, G. K., Taranu, Z. E. & Gregory-Eaves, I. (2011) Land-use legacies are important determinants of lake eutrophication in the anthropocene. *PLoS ONE,* **6,** e15913.

Keele, L. (2008) *Semiparametric regression for the social sciences*. John Wiley & Sons, Ltd.

King, R. S., Baker, M. E., Whigham, D. F., Weller, D. E., Jordan, T. E., Kazyak, P. F. & Hurd, M. K. (2005) Spatial considerations for linking watershed land cover to ecological indicators in streams. *Ecological Applications,* **15,** 137-153.

Klein Goldewijk, K., Beusen, A. & Janssen, P. (2010) Long-term dynamic modeling of global population and built-up area in a spatially explicit way: Hyde 3.1. *The Holocene,* **20,** 565-573.

Kleinman, P. J. A., Sharpley, A. N., McDowell, R. W., Flaten, D. N., Buda, A. R., Tao, L., Bergstrom, L. & Zhu, Q. (2011) Managing agricultural phosphorus for water quality protection: Principles for progress. *Plant and Soil,* **349,** 169-182.

Kleinman, P. J. A., Srinivasan, M. S., Dell, C. J., Schmidt, J. P., Sharpley, A. N. & Bryant, R. B. (2006) Role of rainfall intensity and hydrology in nutrient transport via surface runoff. *J. Environ. Qual.,* **35,** 1248-1259.

Kosten, S., Huszar, V. L. M., Mazzeo, N., Scheffer, M., Sternberg, L. d. S. L. & Jeppesen, E. (2009) Lake and watershed characteristics rather than climate influence nutrient limitation in shallow lakes. *Ecological Applications,* **19,** 1791-1804.

Kuhn, M. (2008) Building predictive models in r using the caret package. *Journal of Statistical Software,* **28,** 1-26.

Lamon, I. E. C., Malve, O. & Pietiläinen, O.-P. (2008) Lake classification to enhance prediction of eutrophication endpoints in finnish lakes. *Environmental Modelling & Software,* **23,** 938-947.

Lehner, B. & Döll, P. (2004) Development and validation of a global database of lakes, reservoirs and wetlands. *Journal of Hydrology,* **296,** 1-22.

Lehner, B. & Grill, G. (*In preparation*) Global-scale mean monthly river discharge at a spatial resolution of 15 sec (450 m).

Lehner, B., Verdin, K. & Jarvis, A. (2008) New global hydrography derived from spaceborne elevation data. *Eos Trans. AGU,* **89**.

Liaw, A. & Wiener, M. (2002) Classification and regression by randomforest. *R News,* **2,** 18-22.

Liu, W., Zhang, Q. & Liu, G. (2011) Effects of watershed land use and lake morphometry on the trophic state of chinese lakes: Implications for eutrophication control. *CLEAN – Soil, Air, Water,* **39,** 35-42.

MacDonald, G. & Bennett, E. (2009) Phosphorus accumulation in saint lawrence river watershed soils: A century-long perspective. *Ecosystems,* **12,** 621-635.

MacDonald, G. K., Bennett, E. M., Potter, P. A. & Ramankutty, N. (2011) Agronomic phosphorus imbalances across the world's croplands. *Proceedings of the National Academy of Sciences,* **108,** 3086-3091.

Mayorga, E., Seitzinger, S. P., Harrison, J. A., Dumont, E., Beusen, A. H. W., Bouwman, A. F., Fekete, B. M., Kroeze, C. & Van Drecht, G. (2010) Global nutrient export from watersheds 2 (news 2): Model development and implementation. *Environmental Modelling & Software,* **25,** 837-853.

Muscutt, A. D. & Withers, P. J. A. (1996) The phosphorus content of rivers in england and wales. *Water Research,* **30,** 1258-1268.

ORNL (2006) Landscan: Global population database (2006 release). Oak Ridge National Laboratory, Oak Ridge, TN.

NBSC (2008) Large freshwater lakes, reservoirs water quality. National Bureau of Statistics of China, Chinese Ministry of Water Resources.

NGA/NASA (2003) Srtm water body data product (specific guidance). *Version 2.0.* pp. 4.

Nõges, T. (2009) Relationships between morphometry, geographic location and water quality parameters of european lakes. *Hydrobiologia,* **633,** 33-43.

Nürnberg, G. K. (1984) The prediction of internal phosphorus load in lakes with anoxic hypolimnia. *Limnology and Oceanography,* **29,** 111-124.

Nürnberg, G. K. (1996) Trophic state of clear and colored, soft- and hardwater lakes with special consideration of nutrients, anoxia, phytoplankton and fish. *Lake and Reservoir Management,* **12,** 432-447.

Nürnberg, G. K. (1998) Prediction of annual and seasonal phosphorus concentrations in stratified and polymictic lakes. *Limnology and Oceanography,* **43,** 1544-1552.

Piñeiro, G., Perelman, S., Guerschman, J. P. & Paruelo, J. M. (2008) How to evaluate models: Observed vs. Predicted or predicted vs. Observed? *Ecological Modelling,* **216,** 316-322.

Pionke, H. B., Gburek, W. J. & Sharpley, A. N. (2000) Critical source area controls on water quality in an agricultural watershed located in the chesapeake basin. *Ecological Engineering,* **14,** 325-335.

Potter, P., Ramankutty, N., Bennett, E. M. & Donner, S. D. (2010) Characterizing the spatial patterns of global fertilizer application and manure production. *Earth Interactions,* **14,** 1-22.

Ramankutty, N., Evan, A. T., Monfreda, C. & Foley, J. A. (2008) Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000. *Global Biogeochemical Cycles,* **22**.

Ramankutty, N. & Foley, J. A. (1999) Estimating historical changes in global land cover: Croplands from 1700 to 1992. *Global Biogeochem. Cycles,* **13,** 997-1027.

Rast, W. & Thornton, J. A. (2004) The phosphorus loading concept and the oecd eutrophication programme: Origin, application and capabilities. *The Lakes Handbook: Lake Restoration and Rehabilitation, Volume 2.* P. O'Sullivan & C. S. Reynolds, pp. 712. Wiley-Blackwell, Malden, MA.

Robertson, D., Saad, D. & Heisey, D. (2006) A regional classification scheme for estimating reference water quality in streams using land-use-adjusted spatial regression-tree analysis. *Environmental Management,* **37,** 209-229.

SEDAC-CIESIN (2004) Global rural-urban mapping project, version 1 (GRUMP v1). Socioeconomic Data and Applications Center, Center for International Earth Science Information Network, Columbia University, New York, NY.

Smith, V. (2003) Eutrophication of freshwater and coastal marine ecosystems a global problem. *Environmental Science and Pollution Research,* **10,** 126-139.

Soranno, P., Wagner, T., Martin, S., McLean, C., Novitski, L., Provence, C. & Rober, A. (2011) Quantifying regional reference conditions for freshwater ecosystem management: A comparison of approaches and future research needs. *Lake and Reservoir Management,* **27,** 138-148.

Soranno, P. A., Hubler, S. L., Carpenter, S. R. & Lathrop, R. C. (1996) Phosphorus loads to surface waters: A simple model to account for spatial pattern of land use. *Ecological Applications,* **6,** 865-878.

Soranno, P. A., Spence Cheruvelil, K., Webster, K. E., Bremigan, M. T., Wagner, T. & Stow, C. A. (2010) Using landscape limnology to classify freshwater ecosystems for multi-ecosystem management and conservation. *Bioscience,* **60,** 440-454.

Taranu, Z. & Gregory-Eaves, I. (2008) Quantifying relationships among phosphorus, agriculture, and lake depth at an inter-regional scale. *Ecosystems,* **11,** 715-725.

Taranu, Z. E., Zurawell, R., Pick, F. & Gregory-Eaves, I. (*In Press*) Climate and nutrients as drivers of contemporary cyanobacteria dynamics across lakes: Relative importance of drivers varies with lake mixis. *Global Change Biology*.

Therneau, T. M. & Atkinson, B. (2011) An introduction to recursive partitioning using the rpart routines. Department of Health Sciences Research: Mayo Clinic, Rochester, MN.

Therneau, T. M. & Atkinson, B. (2012) Rpart: Recursive partitioning, v.3.1-53. R package.

US-EPA (2009) National lakes assessment data. Lakes dataset version: 2007. U.S. Environmental Protection Agency, Washington D.C.

Van Drecht, G., Bouwman, A. F., Harrison, J. & Knoop, J. M. (2009) Global nitrogen and phosphate in urban wastewater for the period 1970 to 2050. *Global Biogeochem. Cycles,* **23,** GB0A03.

Vermaire, J. C., Prairie, Yves T. & Gregory-Eaves, I. (2012) Diatom-inferred decline of macrophyte abundance in lakes of southern quebec, canada. *Canadian Journal of Fisheries and Aquatic Sciences,* **69,** 511-524.

Wagner, T., Soranno, P. A., Webster, K. E. & Cheruvelil, K. S. (2011) Landscape drivers of regional variation in the relationship between total phosphorus and chlorophyll in lakes. *Freshwater Biology,* **56,** 1811-1824.

Williams, W. D. (2004) Lakes in arid environments. *The Lakes Handbook: Lake Restoration and Rehabilitation, Volume 2.* P. O'Sullivan & C. S. Reynolds, pp. 712. Wiley-Blackwell, Malden, MA.

Wood, S. (2006) *Generalized additive models: An introduction with r*. Chapman and Hall/CRC, Boca Raton, Florida.

Zuur, A. F., Ieno, E. N., Walker, N., Saveliev, A. A. & Smith, G. M. (2009) *Mixed effects models and extensions in ecology with r*. Springer, New York, NY.

5.8. Tables and Figures

Table 1. Data used in the CART/LM, GAM, and random forest models. Some predictors were excluded from the GAM and CART/LM approach due to presumed overlap with other predictors, or because of missing data. Additionally, not all variables listed were ultimately included the linear models and GAMs following exclusion due to multi-collinearity or multivariate correlation. Data resolutions are approximate values near the equator. *Data sources are listed on following page.*

Theme	**Dataset** (units)	**Model types**			**Resolution** (km x km)	**Data source**
		CART/LM	*GAM*	*RF*		
Biophysical characteristics						
Hydrological Morphometric	Lake area (km^2)	■ (RT)	■	■	n/a	*
	Mean lake depth (m)	■ (RT)		■	n/a	*
	Lake volume (million m^3)	■ (RT)		■	n/a	*
	Watershed to lake area ratio (WA:LA)	■ (RT)	■	■	~0.5 x 0.5	*
	Discharge at lake outflow (m^3/s)	■ (RT)	■	■	~0.5 x 0.5	***1***
	Mean slope of watershed (°)	■ (RT)	■	■	~0.5 x 0.5	***2***
Climate	Mean annual temperature (MAT; °C)	■ (RT)	■	■	~1 x 1	***3***
	Mean annual precipitation (MAP; mm)	■ (RT)	■	■	~1 x 1	***3***
Soils	Clay content (%)	■ (RT)	■	■	~5 x 5	***4***
	Sand content (%)	■ (RT)	■	■	~5 x 5	***4***
	Organic carbon content (%)			■	~5 x 5	***4***
	Dominant soil order (FAO)	■ (RT)	■	■	~5 x 5	***4***
	Dominant soil drainage class (FAO)	■ (RT)		■	~5 x 5	***4***
	Dominant soil texture class (USDA)			■	~5 x 5	***4***
Anthropogenic drivers						
Land use	Cropland extent in watershed (%)	■ (LM)	■	■	~10 x 10	***5***
	Pasture extent in watershed (%)	■ (LM)	■	■	~10 x 10	***5***
	Historic agricultural extent (%)	■ (LM)	■	■	~50 x 50	***6***
Land management	Cropland P fertilizer use (kg ha^{-1})	■ (LM)	■	■	~50 x 50	***7***
	Manure P produced (kg yr^{-1} x 100)	■ (LM)	■	■	~50 x 50	***8***
	Cropland soil P balance (kg ha-1)			■	~50 x 50	***7***
Human population	Population density (persons km^{-2})	■ (LM)	■	■	~1 x 1	***9***
	Historic population density (p km^{-2})	■ (LM)	■	■	~1 x 1	***10***
	P yield in domestic wastewater (kg yr^{-1})			■	>1 x 1	***11***
	Urban land cover extent (%)			■	~1 x 1	***12***

Data source key corresponding to Table 1:

*** :** Reported by sources or calculated as described in Chapter 5 text for lake areas.

1 : HydroSHEDs and WaterGap (Alcamo *et al.* (2003); Lehner *et al.* (2006); Lehner & Grill (*In preparation*))

2 : NGA/NASA (2003)

3 : WorldClim (Hijmans *et al.* (2005))

4 : Harmonized World Soil Database (FAO/IIASA (2012))

5 : Ramankutty *et al.* (2008)

6 : Ramankutty & Foley (1999; updated in 2011)

7 : MacDonald *et al.* (2011) – Chapter 2 (this thesis)

8 : Potter *et al.* (2010)

9 : Landscan (ORNL (2006)

10 : Hyde v.3.1 (Klein-Goldewijk *et al.* (2010))

11 : Calculated using data from Van Drecht *et al.* (2009) and Landscan (ORNL (2006))

12 : GRUMP (SEDAC-CIESIN (2004))

Table 2. Model performance results for the annual lake TP observation data with cross-validation (25 iterations of each model, with 80% training and 20% test sets to assess predictions). Because cross-validation was not possible for the spatial GAM model, the R^2 of the model with the lowest AIC_c is shown. RMSE is root mean square error.

Approach	Model	RMSE (μg P L^{-1})	Cross-validated R^2	R^2 - Std. deviation
CART / LM	*LM #1*	6.71	0.26	0.18
	LM #2	4.03	0.54	0.17
	LM #3	5.73	0.36	0.20
	LM #4	3.87	0.27	0.19
	LM #5	5.25	0.19	0.15
	LM #6	4.13	0.24	0.23
	LM #7	8.47	0.45	0.32
	LM #8	11.48	0.27	0.18
	LM #9	3.53	0.46	0.21
	LM #10	6.79	0.32	0.24
	LM #11	4.66	0.59	0.32
	LM #12	5.78	0.57	0.31
GAM	*Non-spatial*	7.52	0.38	0.06
	Spatial (lowest AIC_c)	--	0.54	--
Random Forest	*500 trees x 12 predictors*	6.76	0.45	0.06

Table 3. Model performance results for the seasonal lake TP observation data with cross-validation. The R^2 of the spatial GAM model with the lowest AIC_c is shown.

Approach	Model	RMSE (μg P L^{-1})	Cross-validated R^2	R^2 - Std. deviation
CART/LM	*LM #1*	10.00	0.33	0.25
	LM #2	3.71	0.62	0.24
	LM #3	4.27	0.51	0.14
	LM #4	10.23	0.43	0.24
	LM #5	4.27	0.41	0.25
	LM #6	6.89	0.54	0.32
	LM #7	6.08	0.54	0.32
	LM #8	9.59	0.37	0.20
GAM	*Non-spatial*	8.75	0.56	0.08
	Spatial (lowest AIC_c)	--	0.73	--
Random Forest	*500 trees x 12 predictors*	8.11	0.58	0.08

Table 4. Geographic, climate, and soil descriptors for the lakes with annual TP observations grouped according to the CART watershed biophysical characteristic divisions (Approach 1). Linear model (LM) numbers correspond to those in other figures and tables.

Model	**Latitude range** (degrees N or S)	**Dominant soil orders**	**MAT** (median C°)	**MAP** (median monthly)	**Median annual TP** ($\mu g\ L^{-1}$)
LM #1	-20S to 58N	Leptosols	8.5	101	12.6
LM #2	41N to 59N	Podzols	6.1	55	16.2
LM #3	-42S to 57N	Cambisols	9.6	97	22.7
LM #4	-24S to 58N	Leptosols/Luvisols	8.4	103	14.0
LM #5	-9S to 52N	Cambisols/Luvisols	13.8	50	43.1
LM #6	42N to 59N	Luvisols/Cambisols	6.2	55	35.8
LM #7	-43S to 55N	Cambisols	8.9	57	32.2
LM #8	-36S to 44N	Cambisols	16.0	105	105.3
LM #9	20N to 56N	Podzols/Cambisols	7.3	55	40.8
LM #10	42N to 57N	Cambisols	12.4	50	69.3
LM #11	-6S to 25N	Cambisols/Luvisols	9.3	53	57.6
LM #12	-8S to 58N	Podzols/Leptisols	10.6	55	131.3

FIGURES

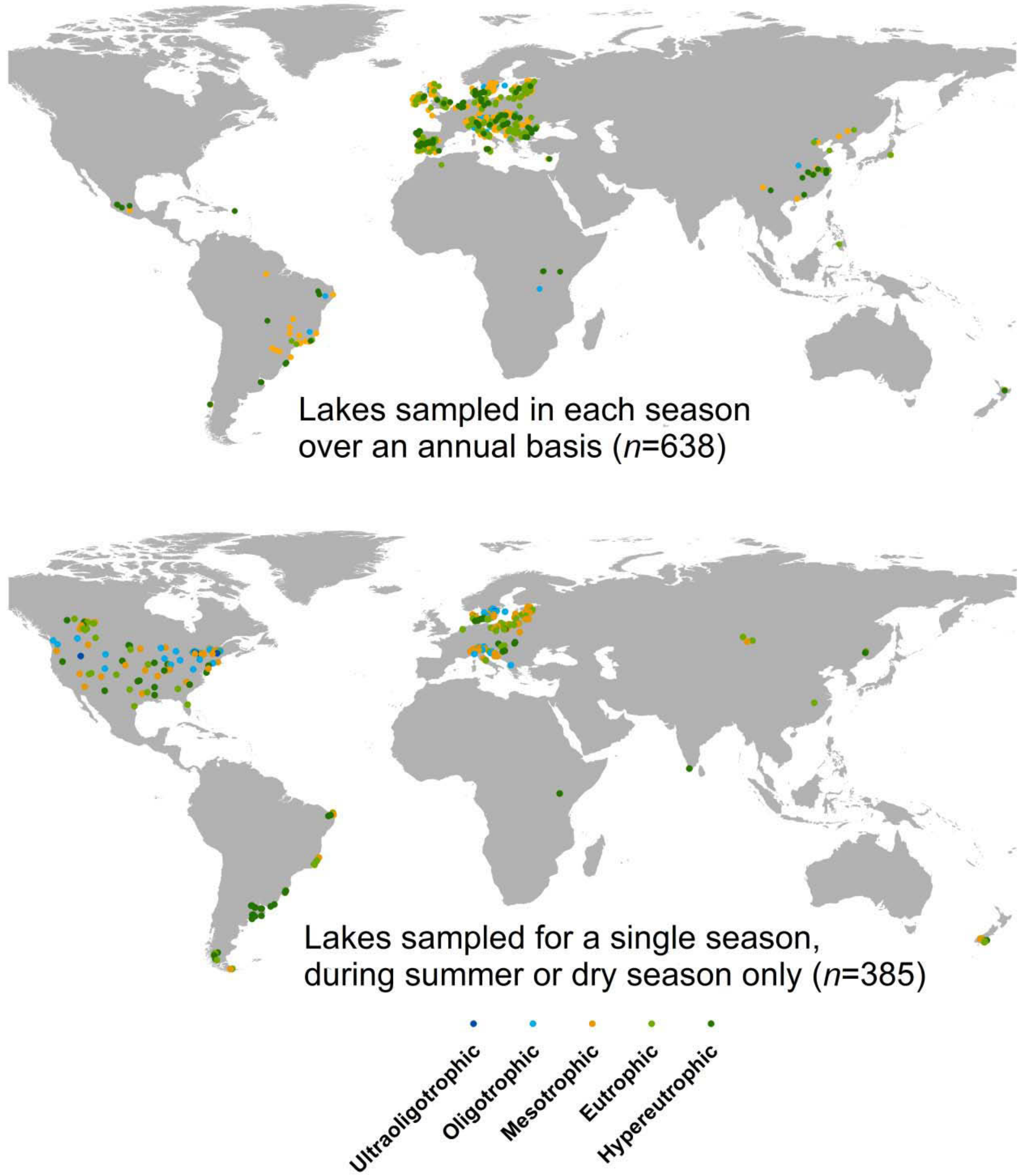


Fig. 1. Maps of the locations of all lakes included in each of the two datasets analyzed in this study (i.e., annual or seasonal TP samples). The point colours pertain to trophic status groupings inferred from TP following the OECD worldwide lakes classification (Rast & Thornton 2004).

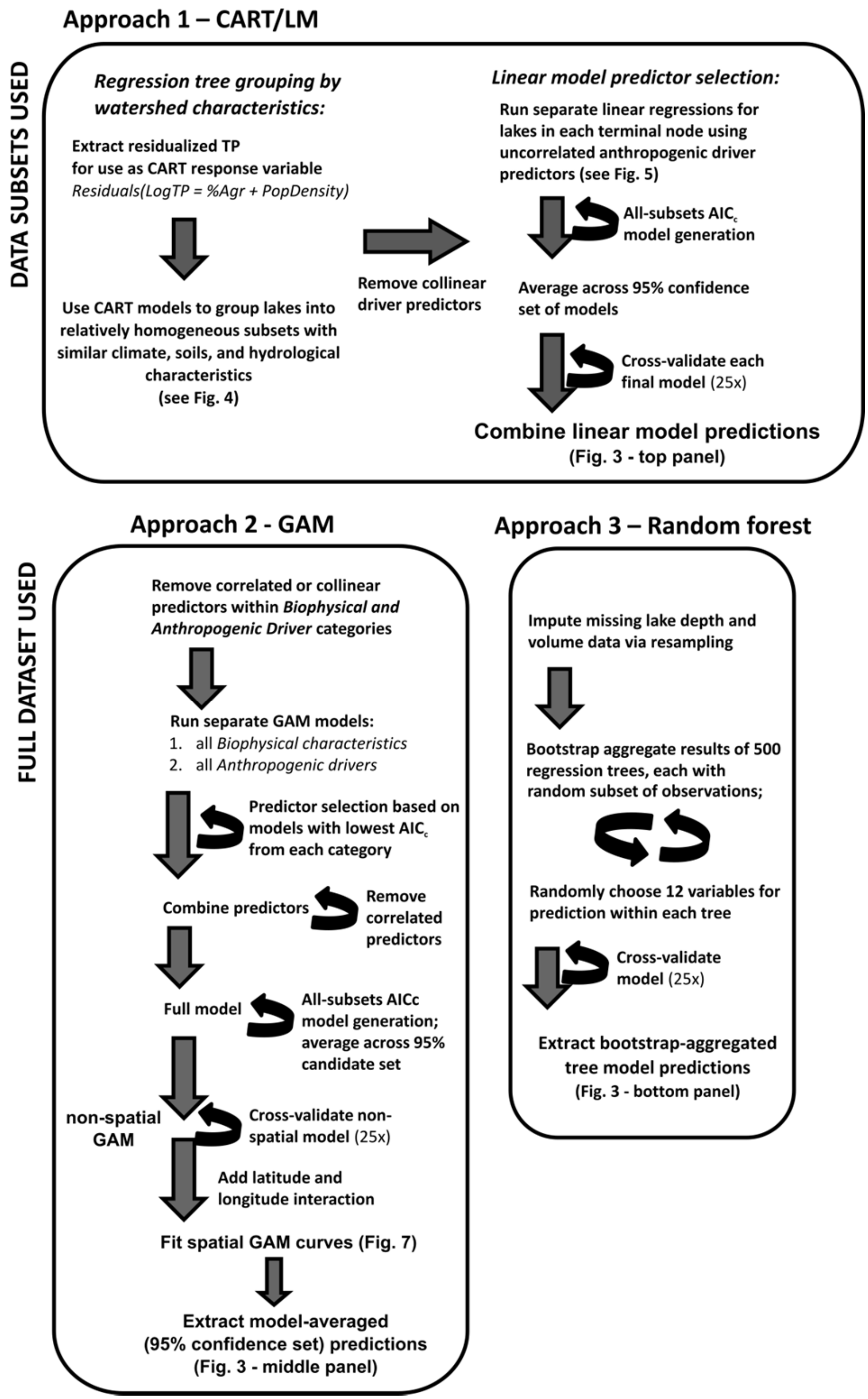


Fig. 2. Conceptual model outlining the three statistical approaches used in this study.

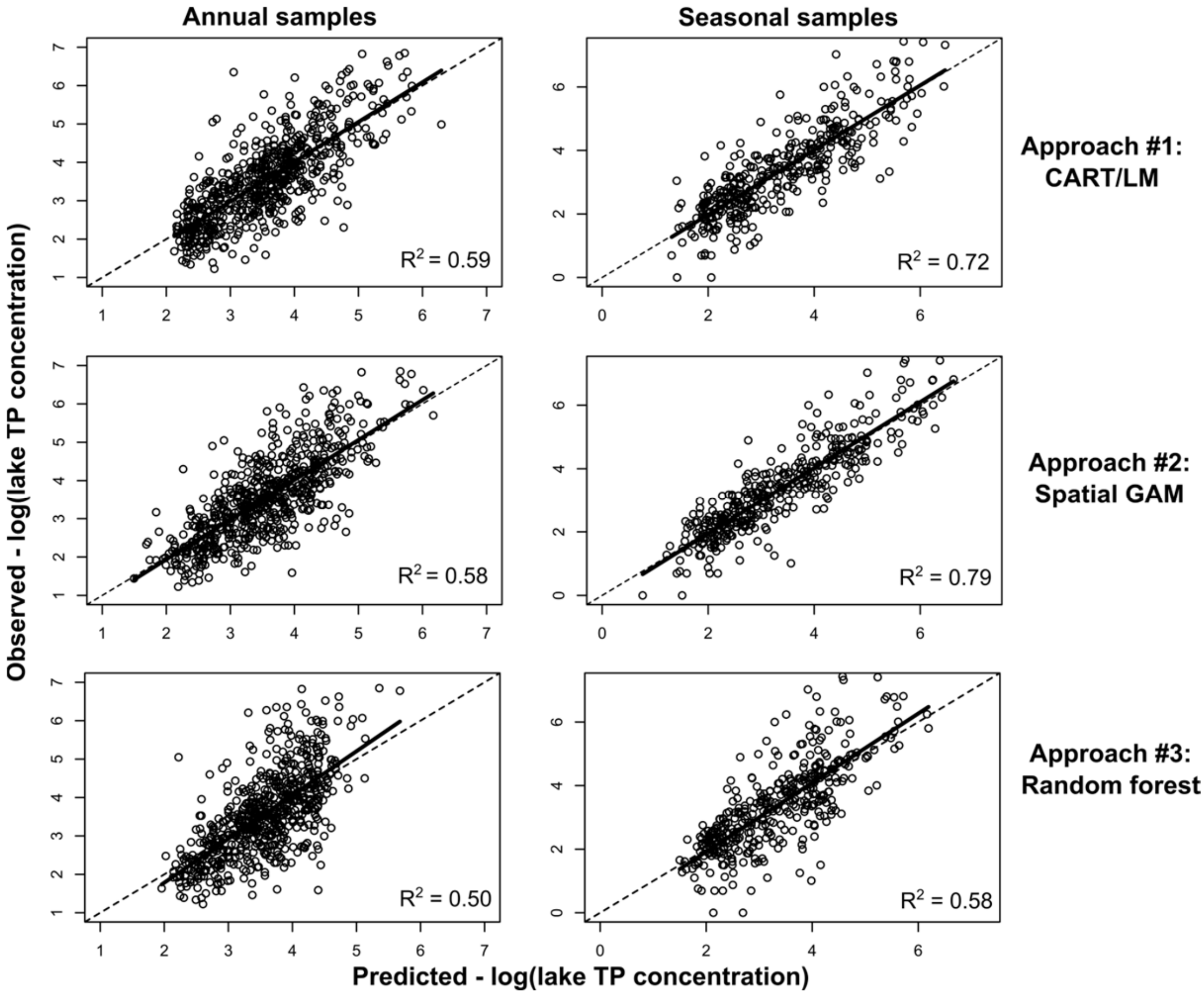


Fig. 3. Plots showing the predicted results versus observed lake TP from each of the modelling approaches used here. Linear regression lines and associated R^2 values fitted through the data are shown with thick black lines and 1:1 lines indicating perfect fit between observed and predicted values are shown as dotted black lines.

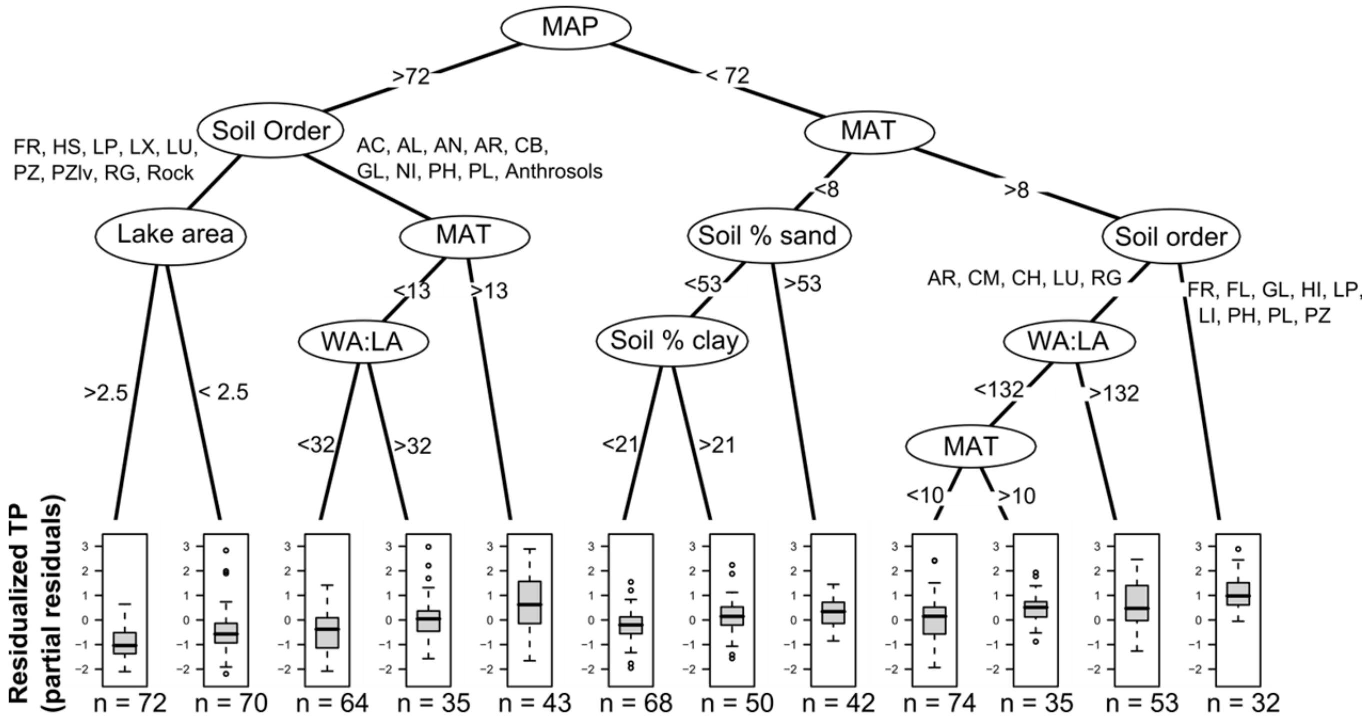


Fig. 4A. Regression tree showing the grouping of lakes with relatively similar watershed characteristics for the annual lake TP dataset. The response variable is residualized lake TP, extracted from the initial regression in Approach 1. The distribution of the partial residuals (LogTP ~ %Agriculture + PopulationDensity) for each terminal node are shown with box plots centred around zero.

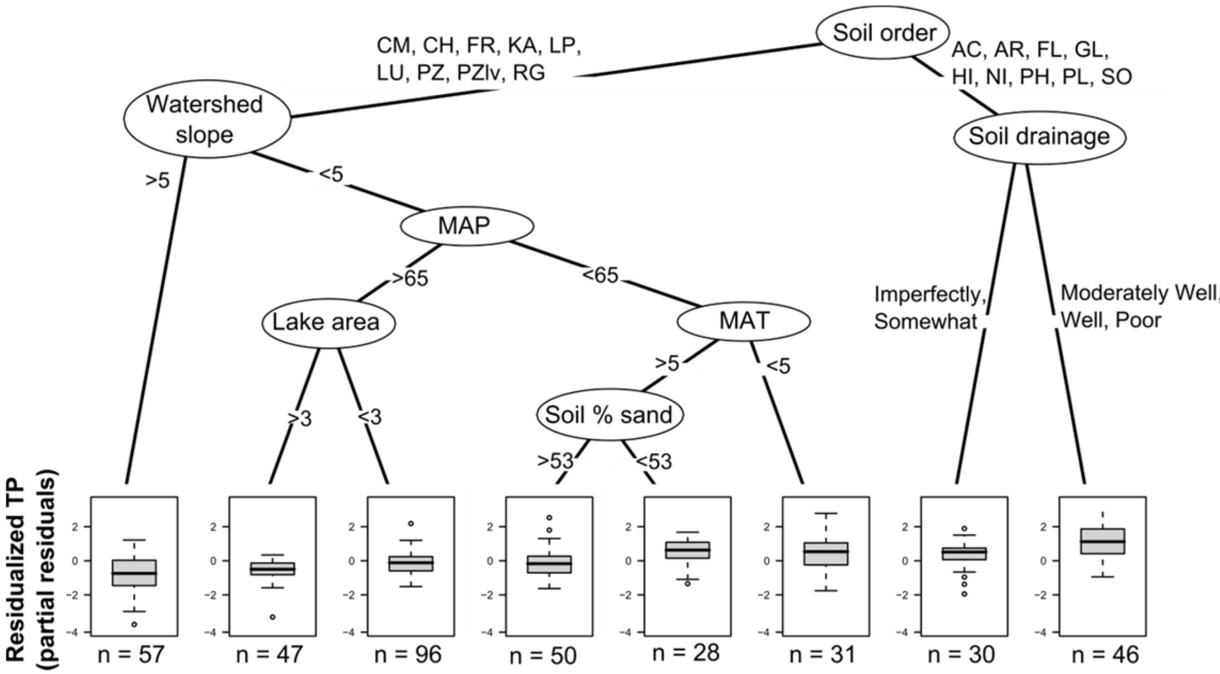


Fig. 4B. Regression tree showing the grouping of lakes with relatively similar watershed characteristics for the seasonal lake TP dataset. The response variable is residualized lake TP, extracted from the initial regression in Approach 1. The distribution of the partial residuals (LogTP ~ %Agriculture + PopulationDensity) for each terminal node are shown with box plots centred around zero.

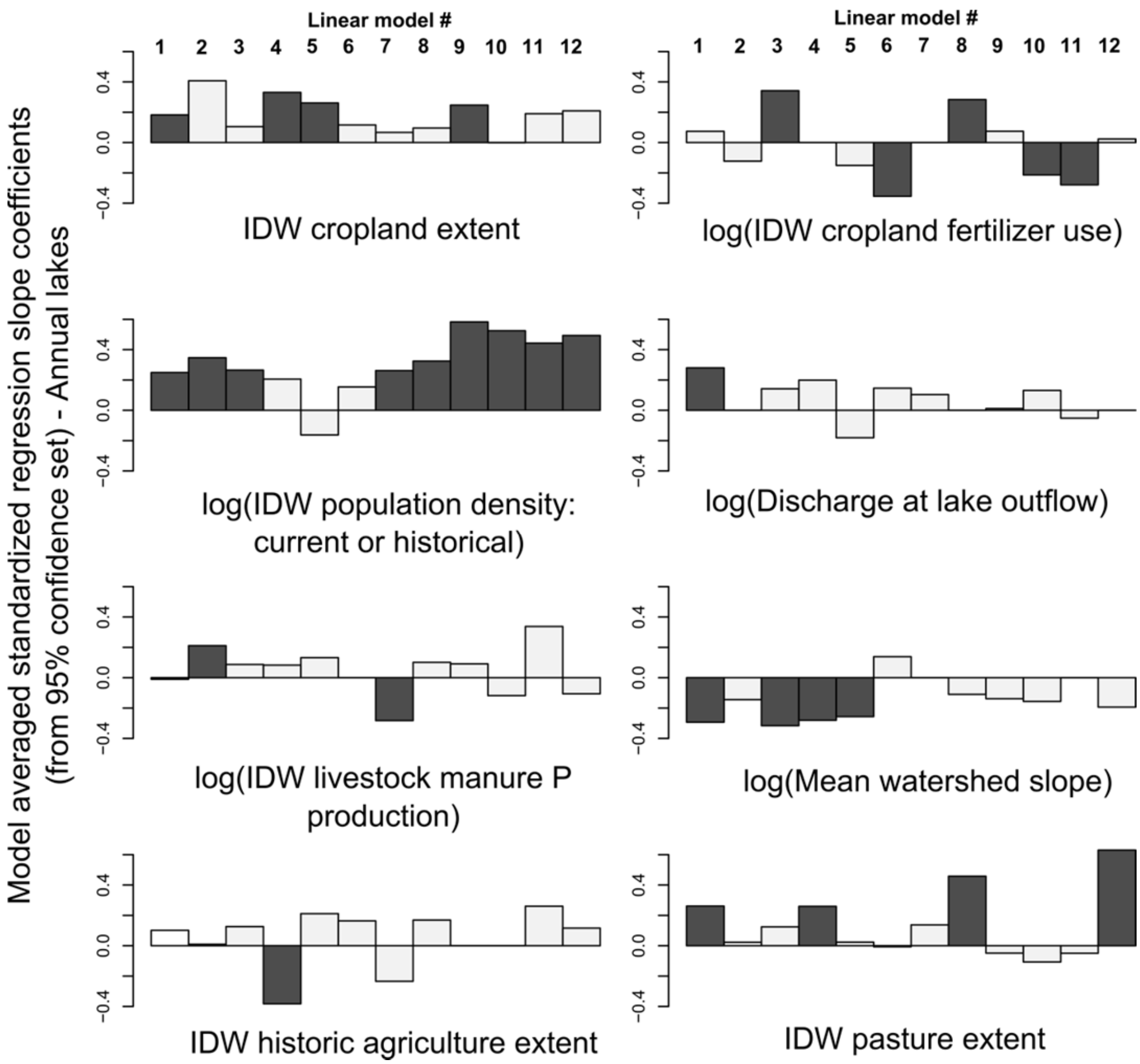


Fig. 5A. Plots showing the model-averaged standardized regression coefficients (slopes) for lakes with annual TP samples (Approach 1). Each bar corresponds to a linear model for lakes in a terminal node group from Fig. 4A. Bars shaded in grey indicate relatively important predictors across the 95% confidence set (Akaike weights >0.50). Any bar with a value of zero means that the predictor was not used due to multicollinearity.

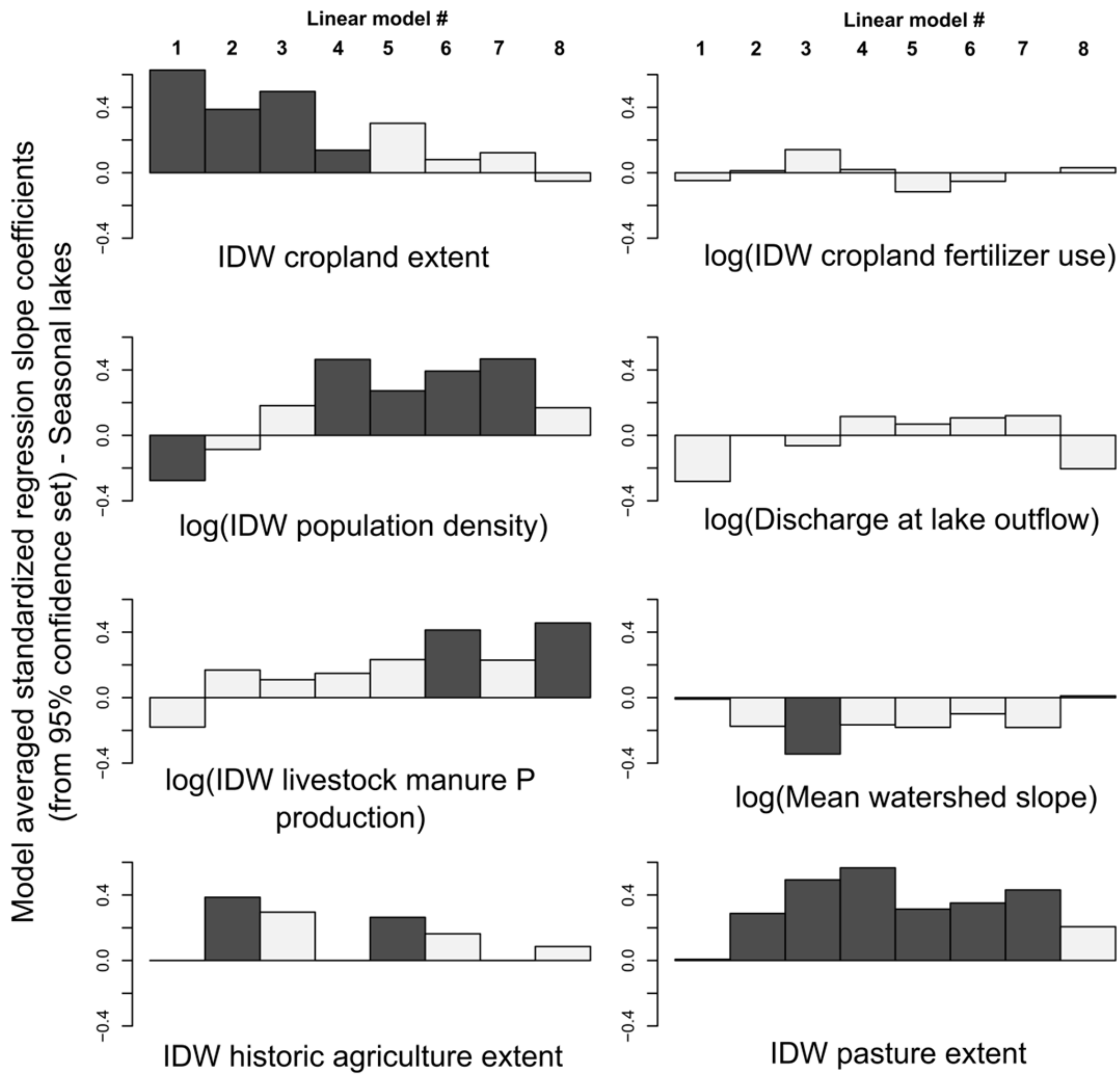


Fig. 5B. Plots showing the model-averaged standardized regression coefficients for lakes with seasonal TP samples (Approach 1). Each bar corresponds to a linear model for lakes in a terminal node group from Fig. 4B. Bars shaded in grey indicate relatively important predictors across the 95% confidence set (Akaike weights >0.50). Any bar with a value of zero means that the predictor was not used due to multicollinearity.

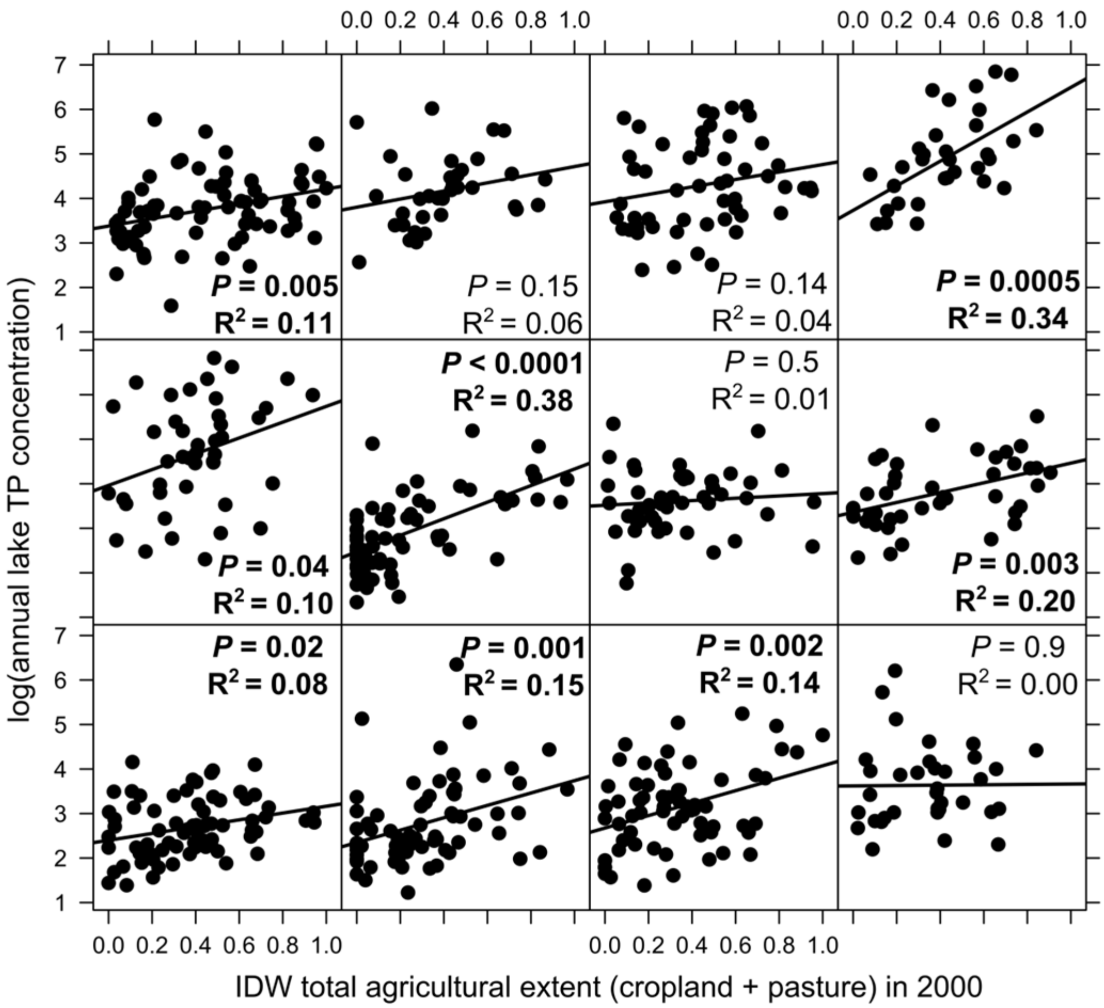


Fig. 6A. Plots showing the relationship between total agricultural extent (proportion of watershed area under agriculture) and lake TP for the Annual lakes dataset. Each plot corresponds to a terminal node group of lakes in Fig. 4A. Linear regression lines and associated *P* values are indicated, with significant relationships (*P*<0.05) indicated in bold.

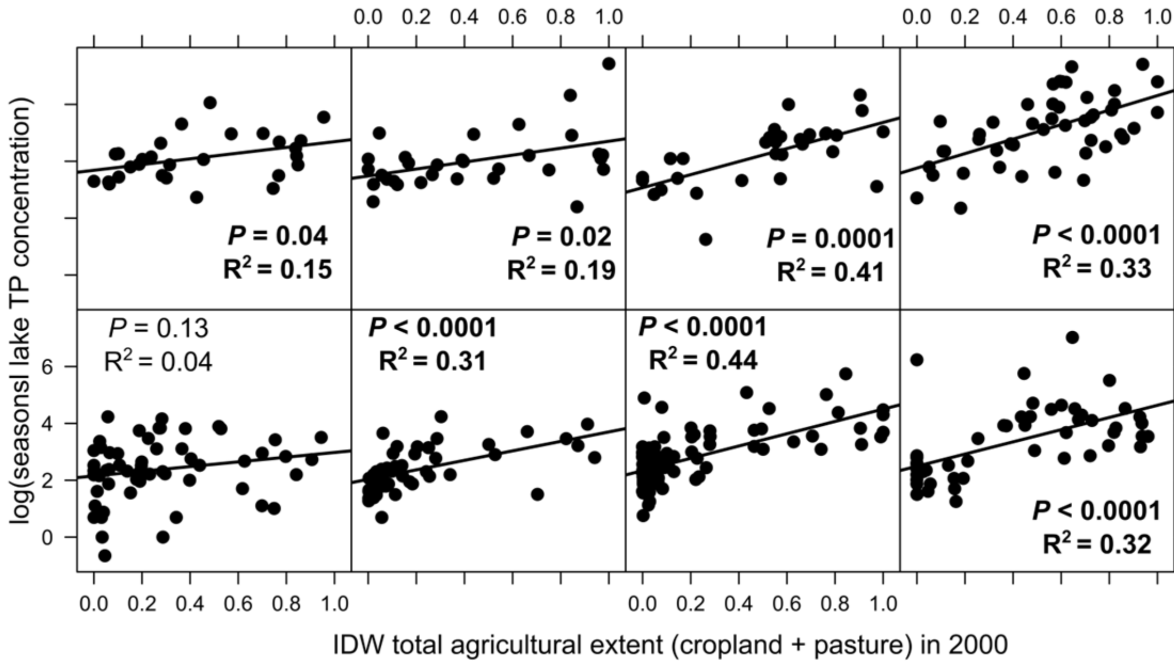


Fig. 6B. Plots showing the relationship between total agricultural extent (proportion of watershed area under agriculture) and lake TP for the Seasonal lakes dataset. Each plot corresponds to a terminal node group of lakes in Fig. 4B. Linear regression lines and associated *P* values are indicated, with significant relationships (*P*<0.05) indicated in bold

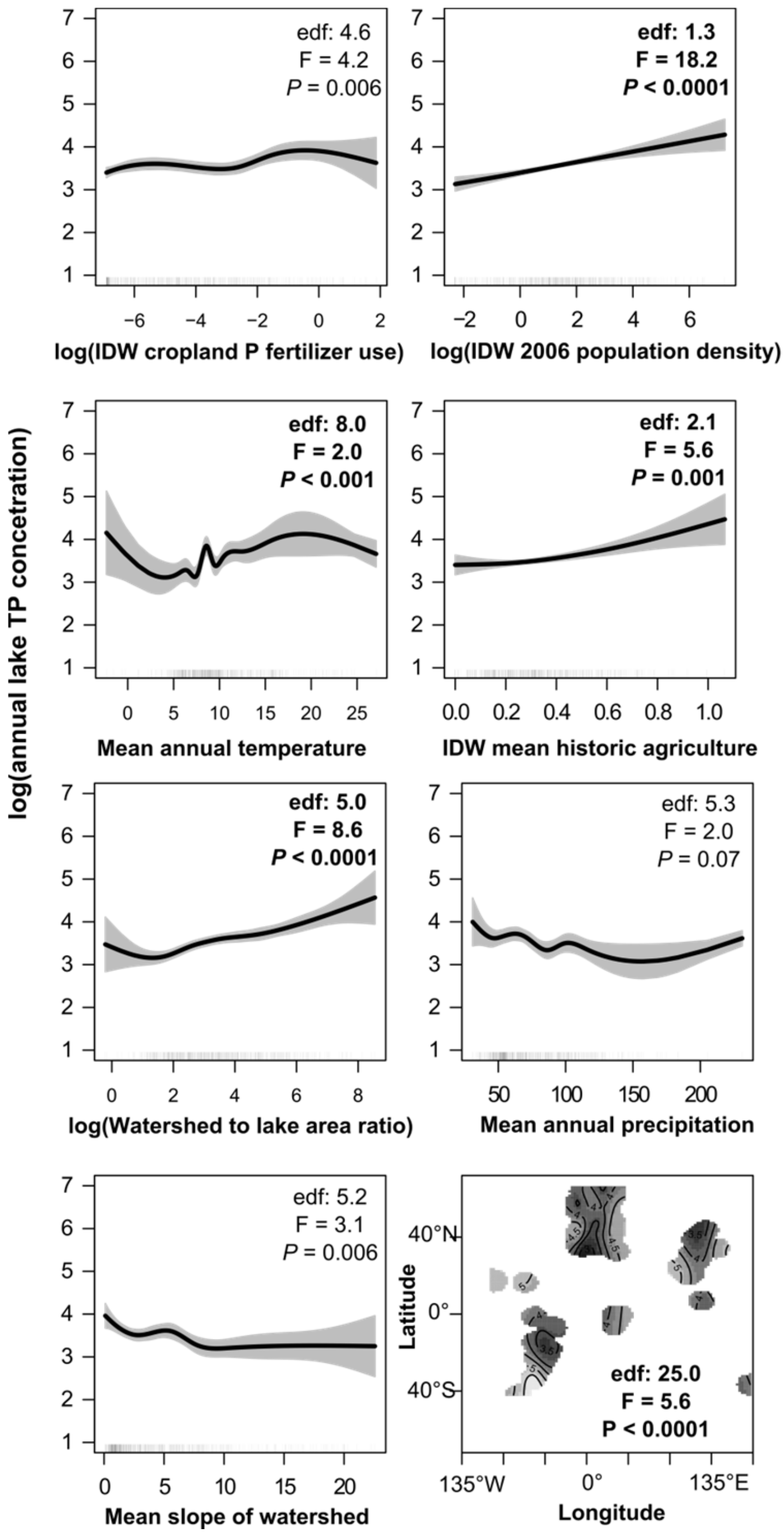


Fig. 7A. Fitted curves for predictors retained in the spatial GAM model with the lowest AIC_c (annual lakes). Each curve controls for the additive effect of other predictors and the spatial interaction (latitude by longitude; darker colours indicate lower predicted TP values and lighter colours indicate higher predicted TP). Highly significant curves are in bold (P < 0.001). Estimated degrees of freedom (edf) >1.0 indicates nonlinearity.

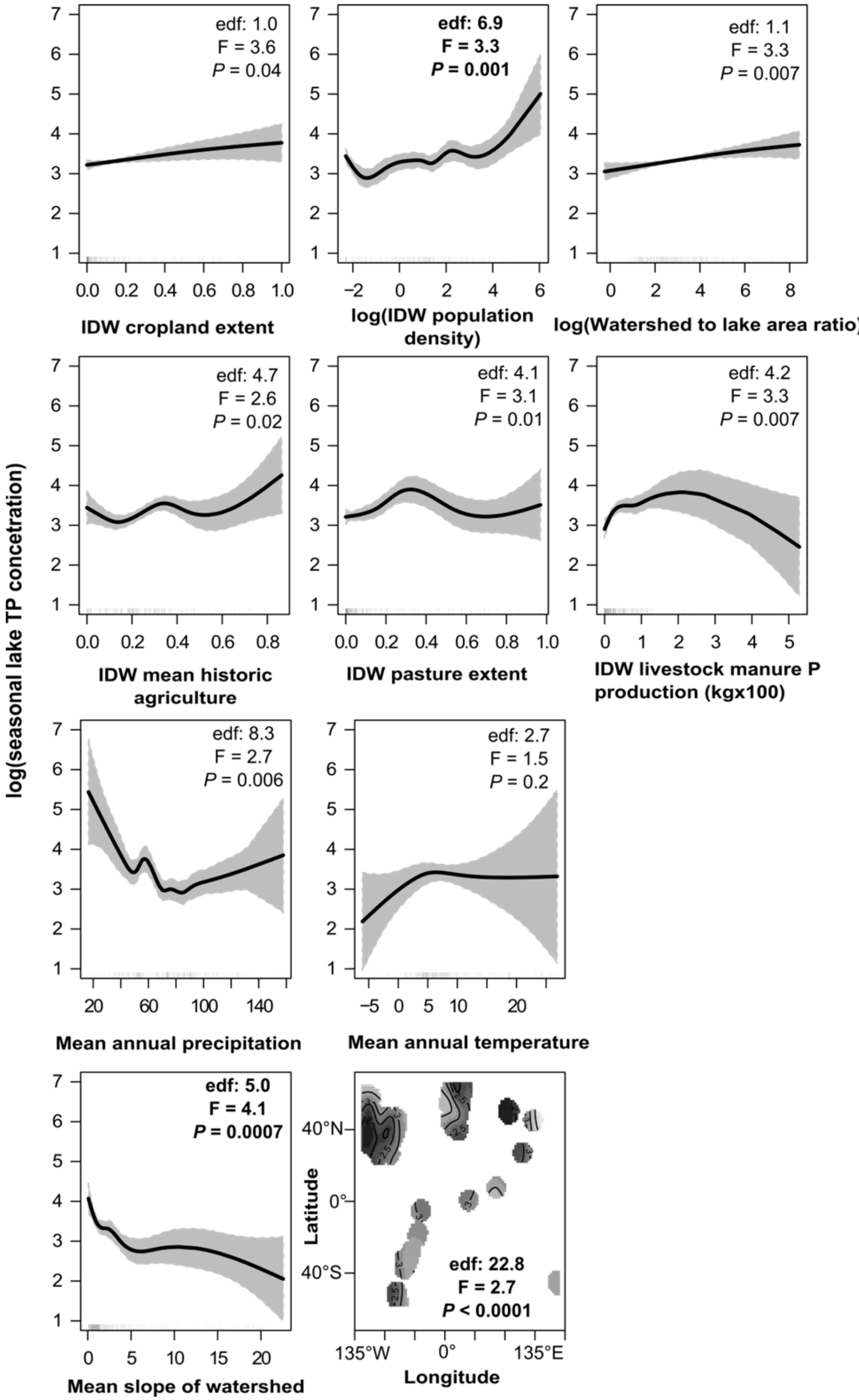


Fig. 7B. Fitted curves for all predictors retained in the spatial GAM model with the lowest AIC_c for the seasonal lakes dataset. Soil order (categorical predictor) was also retained in this model (*P* < 0.001), but is not shown. See Fig. 7A for annotations.

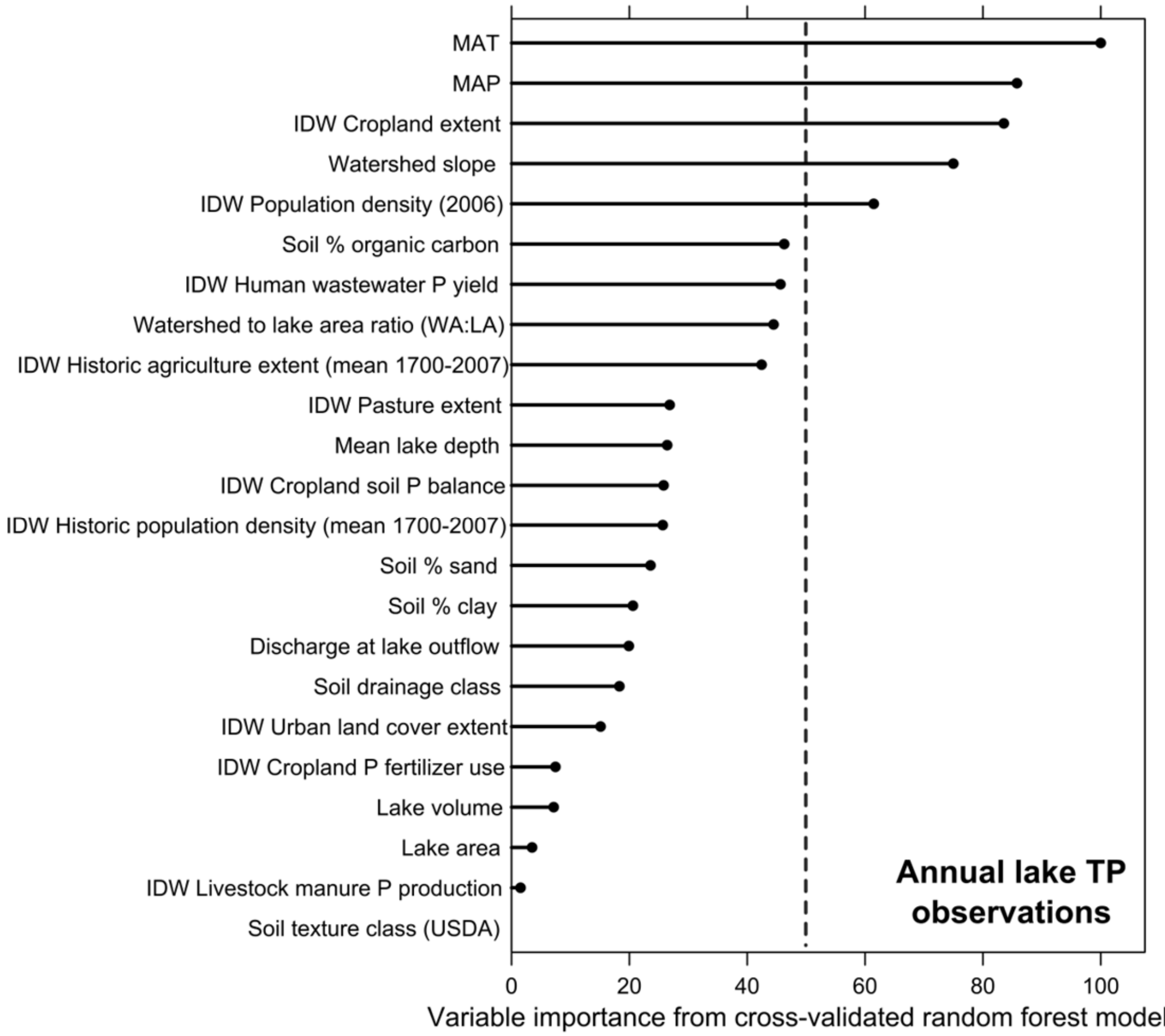


Fig. 8A. Variable importance results for the random forest models (annually sampled lakes).

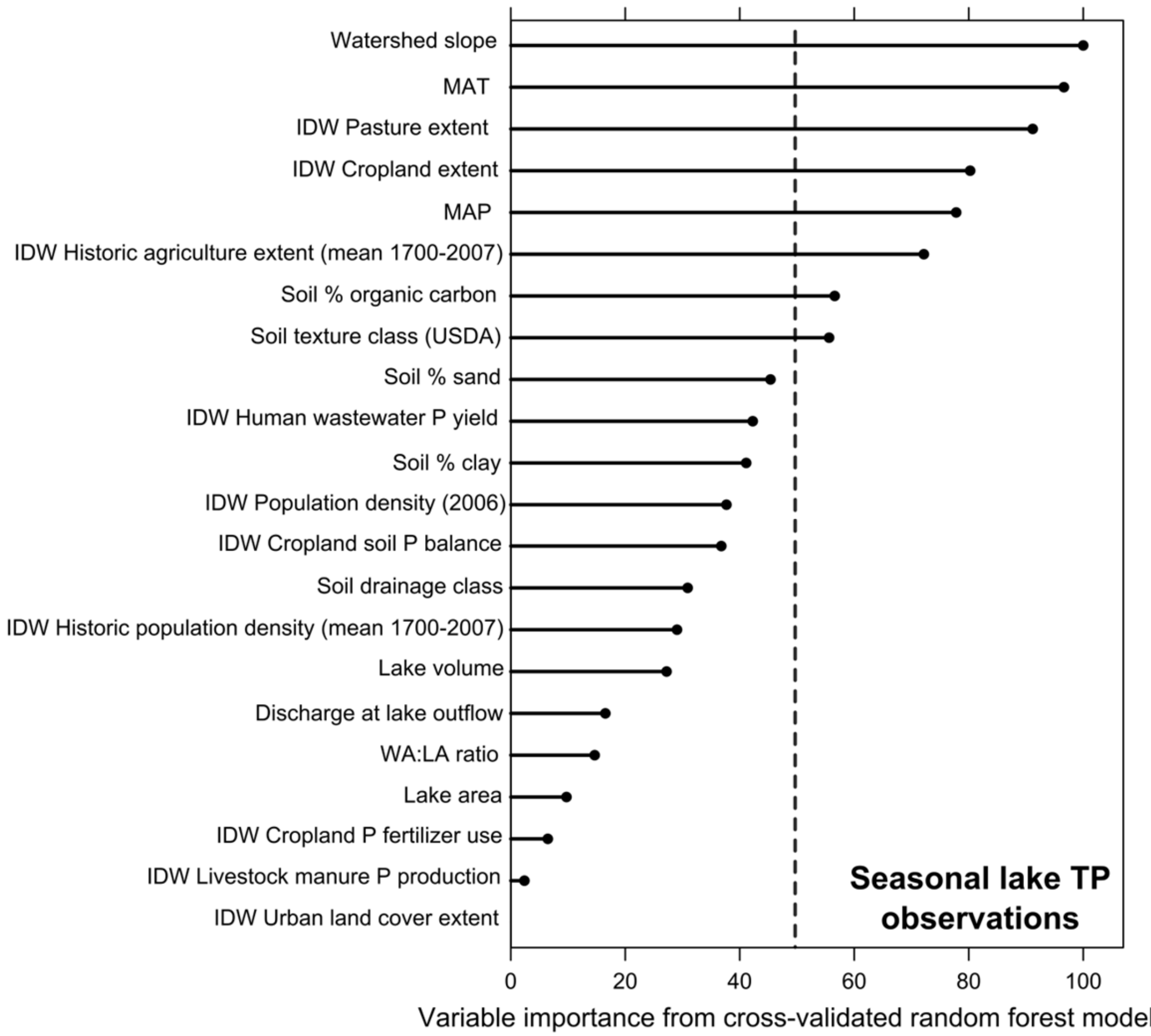


Fig. 8B. Variable importance results for the random forest models (seasonally sampled lakes).

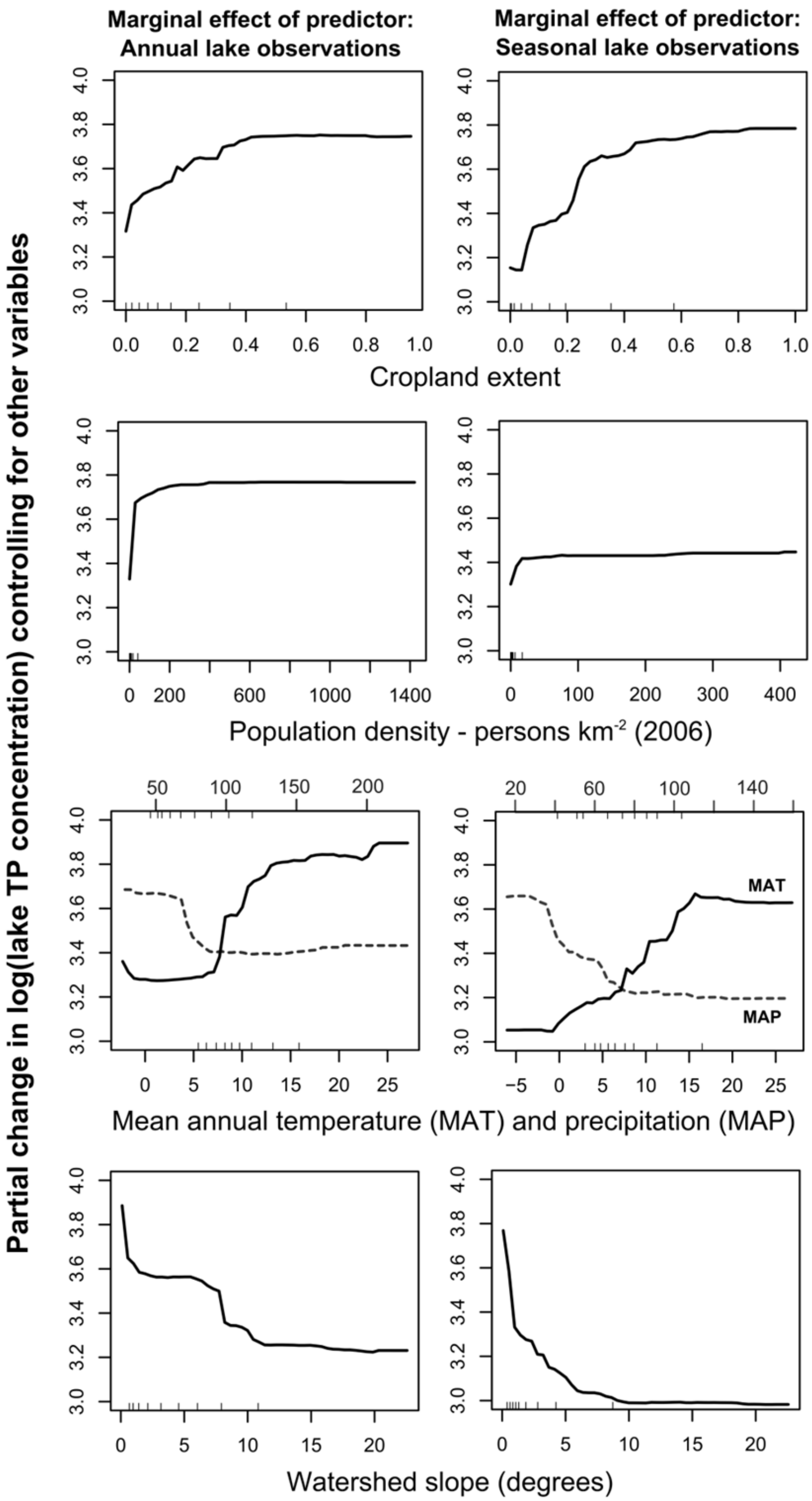


Fig. 9. Partial dependence plots showing the marginal effect of key anthropogenic and envionmental predictors on log-transfomed lake TP concentrations when controlling for all other predictors in the random forest models. The tick marks at the bottom of each plot indicate the deciles (10^{th} percentile intervals) for the predictor data.

CHAPTER 6: SYNTHESIS & CONCLUDING REMARKS

6.1. CONTRIBUTIONS TO KNOWLEDGE

I presented four novel studies in this thesis that explored the influence of human activity, particularly agriculture, on the alteration of phosphorus (P) flows across space, time, and ecological systems. Each of these studies considered unique components of the modern P cycle with different spatial emphases. These studies also addressed key knowledge gaps, providing quantitative results that are policy relevant (Chapters 2 and 3) and insightful for ecosystem management (Chapters 3 and 4).

- *P imbalances in cropland soils globally* (Chapter 2): This spatially-detailed study uniquely addressed both the drivers and distribution of P surpluses and deficits for croplands globally, building considerably on previous global research that considered more aggregate results (e.g., Smil 2000; Bennett *et al.* 2001; Bouwman *et al.* 2009). Nutrient imbalances are often generalized for entire regions in global studies. However, the results of this study indicated that there is considerable sub-regional variation in P imbalances due to the distinct roles of P fertilizer versus manure P use in different locations, and how these relate to the both the types of crops grown and their yields. These results also provided new insight on the efficiency of P use globally, highlighting important inefficiencies in P fertilizer use across many regions. This study was the first to propose redistribution of P from areas of surplus to areas of deficit at large scales using quantitative estimates.

- *Global connections in P use for a regional agricultural system* (Chapter 3): This comprehensive analysis of the regional P situation in the US (a key agricultural producing and consuming nation) addressed the cumulative implications of agricultural management, globalization, livestock and biofuel production on P use. Based on a detailed model of the US agricultural system, I compared the amount of P used in the production of specific commodities (e.g., animal products, processed crops, and corn ethanol), as well as the potential losses of this P throughout the food system (from agricultural soils to the food consumed in human diets). This work offered novel examination of the linkages in fertilizer use between the US and its trading partners globally for specific agricultural commodities (by comparing P flows related to US production for domestic consumption, foreign production for US consumption, and US production for foreign consumption). Finally, I placed quantitative estimates concerning how potential changes to food system management would influence regional P use, which have typically been discussed only conceptually in large-scale studies. The scenarios results indicated that improved P management at the farm, distributional, and consumer level could reduce annual P fertilizer applications in the US by as much as half

- *Soil P legacies among globally distributed sites* (Chapter 4): While several studies have reported persistent changes in soil P pools due to past agriculture, broader understanding of these legacies has been clouded by immense variation across studies. This study was the first empirical analysis of agricultural land-use legacies on soil P, using results compiled from a large sample of studies (n=94) worldwide. The quantitative synthesis of soil P data from abandoned agricultural areas around the world comprehensively addressed how farming history, soil characteristics, and biophysical factors influence soil P pools over time following cessation of agriculture. These insights into the trajectories of soil P following land-use change from agriculture are essential to understanding the ecological implications of land-use history and the potential to remediate these effects at the ecosystem scale. In particular, we found that some agriculturally-induced changes in soil P may be reduced over time, which challenges the notion that these changes are largely irreversible.

- *Watershed drivers of total P concentrations for a diverse set of lakes globally* (Chapter 5): This study is the first to use defined watershed boundaries for a large sample of lakes worldwide to test the effect of multiple anthropogenic and biophysical variables on lake total P (representing both contemporary and historical land uses, agricultural land management, watershed drainage, edaphic characteristics, as well as climate). The findings of this study demonstrated that it is possible to obtain fairly accurate predictions of lake total P using estimates of watershed characteristics derived from global land use, hydrological, soils, and climate datasets. These results provide new insight on the relative influence of various drivers of lake total P at inter-regional scales; in particular, the influence of land use attributes, such as the proportion of a watershed in agriculture and watershed population density, appears to be interdependent with watershed biophysical characteristics for determining lake TP. These models will serve as the basis for mapping spatial patterns of eutrophication risk for individual lakes worldwide.

6.2. Cross-cutting themes

Each of the disparate components of the P cycle considered in this thesis are interrelated, with collective implications for P as a source of pollution and P as a critical nutrient for humanity (Elser 2012). Finding operational solutions to resolve P imbalances and inefficiencies at different scales will be essential for achieving sustainable agricultural P use that has more positive agronomic and ecological outcomes (Vitousek *et al.* 2009; Carpenter & Bennett 2011; Elser & Bennett 2011; Kleinman et al. 2011; Townsend & Porder 2011; Wang *et al.* 2011; Cordell *et al.* 2012; Townsend & Porder 2012).

6.2.1. Opportunities to mitigate P imbalances and reduce systemic P losses

Assessing geographic patterns of agricultural P use is essential to understanding the factors that contribute most to nutrient imbalances at the regional scale and opportunities to improve P-use efficiency. The analysis presented in Chapter 2 helped to pinpoint the relative influence of agronomic P inputs (either fertilizer, manure, or both) in relation to crop harvest on nutrient imbalance trends globally. I showed that the most intense P surpluses at the global scale were primarily associated with fertilizer use, and that fertilizer P was surprisingly less efficient at improving crop productivity than manure in some regions. Some of the global patterns of P imbalances and associated P-use efficiencies were clearly related to broad regional variation in agricultural systems (e.g., farming intensities and crop productivity). This results in a concentration of P imbalances in specific areas. However, generalizing about P use across whole regions fails to account for sub-regional variations in P surpluses and deficits that are linked to more local drivers, such as livestock densities, the types of crops produced and their yields. These results also show that global P imbalances could be mitigated by redistributing P from areas of surplus to areas of deficit at smaller scales (e.g., Bateman *et al.* 2011).

Exactly what agricultural commodities are produced with the mineral P applied to croplands, and where this P goes once these crops leave the farm, is central to leveraging P use in globally-integrated agricultural systems. Chapter 3 demonstrated how P fertilizer use in the US is dominated by a few commodities that strongly impacted the fate of mineral P inputs throughout the agricultural supply chain (e.g., red meat and corn ethanol). For example, this study quantitatively demonstrated the degree of P ‘leakage’ from the food system associated with meat production, primarily because manure P is inadequately recycled back to croplands at the national scale (Steinfeld *et al.* 2006; Menzi *et al.* 2010). However, scenario analysis suggests that tackling particular P inefficiencies in the US agricultural system (e.g., national P surpluses for two major crops, losses of P during processing of crops and via consumer waste, etc.), could dramatically reduce annual P fertilizer requirements with no net change in production or caloric intake.

6.2.2. Global integration of agricultural systems and the P cycle

Researchers are beginning to acknowledge the importance of agricultural trade on the P cycle and what this means for managing finite reserves of phosphate rock (Matsubae *et al.* 2011; Schipanski & Bennett 2012). My findings underpin the need to explicitly consider agricultural trade in national P budgets (Cordell *et al.* 2012), particularly in the case of major agricultural net-exporters such as the US (Chapter 3).

Agricultural trade has an important effect on overall P fertilizer applications in the US and its trading partners, by diverting P fertilizer use to other countries (via imports of certain products, such as beef from Canada) or concentrating P fertilizer use in countries producing exports (e.g., for US corn exports to East Asia). Agricultural trade therefore has a concomitant role in contributing to the P surpluses and deficits identified in Chapter 2. In the US context, decreasing the magnitude of P fertilizer required to produce exports by improving P management could lessen future geo-political concerns surrounding depletion of US domestic phosphate rock reserves (Childers *et al.* 2011). Accounting for the P use embodied in the production of traded commodities will be essential to sustainable and equitable management of the modern P cycle.

6.2.3. Enduring soil P legacies and eutrophication risk

My subsequent analyses served to integrate some of the above concepts (namely, soil P inputs and associated imbalances across the landscape) with our understanding of the ecological effects that these anthropogenic P flows can have over time (Chapter 4) and space (Chapter 5). In the context of this thesis, these studies represent a starting point to help translate our understanding of agriculturally-driven changes to the P cycle into useful conceptual or empirical models for ecosystem management with global applicability (e.g., Carpenter & Bennett 2011; Dumas *et al.* 2011; Sattari *et al.* 2012).

In Chapter 4, I addressed land-use legacies on soil P, which are integral to ecosystem management in the context of an important global land-use change trend (agricultural abandonment) (McLauchlan 2006). This meta-analysis confirmed that enduring legacies of past agriculture do occur for both plant available and total P pools in many areas following agricultural abandonment. However, there was a reduction in the magnitude of this soil P alteration over time since abandonment when studies were viewed together. More specifically, I observed that the type of vegetation established following cessation of agriculture and overall soil types appeared to mediate broader land-use legacies on soil P across studies and regions. Soil characteristics, such as clay content and pH, were particularly influential for plant available P pools in post-agricultural ecosystems. Importantly, the magnitude of soil P legacies appears to differ considerably between soils that received soil amendments (e.g., fertilizer or manure applications) during the agricultural period and those that did not.

This insight on the dynamics of soil P related to past agriculture helps to understand the role that land-use history can have for current ecosystem management (Foster *et al.* 2003; McLauchlan 2006; Keatley *et al.* 2011; Townsend & Porder 2012), Yet, it also suggests the importance of considering factors specific to a given location, particularly soil and vegetation types, in understanding the persistence of past P imbalances on current soil P. However, the finding of a decrease in the magnitude of soil P alteration over time in Chapter 4 challenges the notion that agriculturally-induced changes in soil P are largely irreversible (Dupouey *et al.* 2002).

Finally, in Chapter 5, I examined how key anthropogenic drivers (agricultural land use and population density) were related to lake total P concentrations around the world. While these drivers were typically associated with elevation of total P (e.g., Taranu & Gregory-Eaves 2008; Keatley *et al.* 2011), the strength of these relationships varied considerably among lakes, possibly due to factors such as differences in background TP concentrations. As observed in Chapter 4 for soils P legacies, I also found that watershed biophysical characteristics (e.g., precipitation, temperature, soil type, watershed slope and other factors that influence drainage into lakes) had a role in mediating the influence of anthropogenic drivers. This confirms the importance of "landscape filtering" potential on controlling P runoff to lakes, particularly in the context of large-scale water quality models (Heathwaite 2010). Using models that accounted for the combined effects of watershed land use and biophysical factors, I demonstrated how it is possible to obtain reasonably accurate predictions of lake seasonal and annual TP for a large sample of individual lakes, even in the absence of detailed local sampling information (e.g., water chemistry).

6.2.4. Overarching conclusions

These studies each point to the need to consider interrelated local, regional, and global factors that affect P flows through agricultural systems and its ecological outcomes over space and time. For example, agricultural P management varies tremendously across the globe, with local factors such as the types of crops grown and livestock densities being important contributing factors (Chapter 2). The potential legacies of agricultural changes to soil P pools and the influence that these could have for P loading to lakes also depends, in large part, on more local factors such as soil type or watershed hydrological context (Chapters 4 and 5).

At the same time, processes occurring at more regional scales, such as trade, consumer behaviour, and government policy, are likely to influence P management decisions by individual farmers locally (Chapters 2 and 3). As a result, we ultimately need to be concerned about the cumulative implications that these heterogeneous factors can have on ecological outcomes across scales, such as freshwater eutrophication, as well as depletion of finite phosphate rock reserves. Greater attention to the spatial variation in both problems and solutions related to P, as well as their complex temporal dimensions, will be essential for advancing the science and policies needed to achieve greater P sustainability in agriculture while simultaneously ensuring healthy aquatic ecosystems.

6.3. References

Bateman, A., van der Horst, D., Boardman, D., Kansal, A. & Carliell-Marquet, C. (2011) Closing the phosphorus loop in england: The spatio-temporal balance of phosphorus capture from manure versus crop demand for fertiliser. *Resources Conservation and Recycling,* **55,** 1146-1153.

Bennett, E. M., Carpenter, S. R. & Caraco, N. F. (2001) Human impact on erodable phosphorus and eutrophication: A global perspective. *Bioscience,* **51,** 227-234.

Bouwman, A. F., Beusen, A. H. W. & Billen, G. (2009) Human alteration of the global nitrogen and phosphorus soil balances for the period 1970-2050. *Global Biogeochemical Cycles,* **23**.

Carpenter, S. R. & Bennett, E. M. (2011) Reconsideration of the planetary boundary for phosphorus. *Environmental Research Letters,* **6,** 014009.

Childers, D. L., Corman, J., Edwards, M. & Elser, J. J. (2011) Sustainability challenges of phosphorus and food: Solutions from closing the human phosphorus cycle. *Bioscience,* **61,** 117-124.

Cordell, D., Neset, T. S. S. & Prior, T. (2012) The phosphorus mass balance: Identifying 'hotspots' in the food system as a roadmap to phosphorus security. *Current Opinion in Biotechnology*.

Dumas, M., Frossard, E. & Scholz, R. W. (2011) Modeling biogeochemical processes of phosphorus for global food supply. *Chemosphere,* **84,** 798-805.

Dupouey, J., Dambrine, E., Laffite, J. & Moares, C. (2002) Irreversible impact of past land use on forest soils and biodiversity. *Ecology,* **83,** 2978-2984.

Elser, J. & Bennett, E. (2011) Phosphorus cycle: A broken biogeochemical cycle. *Nature,* **478,** 29-31.

Elser, J. J. (2012) Phosphorus: A limiting nutrient for humanity? *Current Opinion in Biotechnology,* ***In press***.

Foster, D., Swanson, F., Aber, J., Burke, I., Brokaw, N., Tilman, D. & Knapp, A. (2003) The importance of land-use legacies to ecology and conservation. *Bioscience,* **53,** 77-88.

Heathwaite, A. L. (2010) Multiple stressors on water availability at global to catchment scales: Understanding human impact on nutrient cycles to protect water quality and water availability in the long term. *Freshwater Biology,* **55,** 241-257.

Keatley, B. E., Bennett, E. M., MacDonald, G. K., Taranu, Z. E. & Gregory-Eaves, I. (2011) Land-use legacies are important determinants of lake eutrophication in the anthropocene. *PLoS ONE,* **6,** e15913.

Kleinman, P. J. A., Sharpley, A. N., McDowell, R. W., Flaten, D. N., Buda, A. R., Tao, L., Bergstrom, L. & Zhu, Q. (2011) Managing agricultural phosphorus for water quality protection: Principles for progress. *Plant and Soil,* **349,** 169-182.

Matsubae, K., Kajiyama, J., Hiraki, T. & Nagasaka, T. (2011) Virtual phosphorus ore requirement of japanese economy. *Chemosphere,* **84,** 767-772.

McLauchlan, K. (2006) The nature and longevity of agricultural impacts on soil carbon and nutrients: A review. *Ecosystems,* **9,** 1364-1382.

Menzi, H., Oenema, O., Burton, C., Shipin, O., Gerber, P., Robinson, T. & Franceschini, G. (2010) Impacts of intensive livestock production and manure management on the environment. (eds. H. Steinfeld, H. Mooney, F. Schneider & L. Neville), pp. 416. Island Press, Washington, DC.

Sattari, S. Z., Bouwman, A. F., Giller, K. E. & van Ittersum, M. K. (2012) Residual soil phosphorus as the missing piece in the global phosphorus crisis puzzle. *Proceedings of the National Academy of Sciences,* **109,** 6348-6353.
Schipanski, M. & Bennett, E. (2012) The influence of agricultural trade and livestock production on the global phosphorus cycle. *Ecosystems,* **15,** 256-268.
Smil, V. (2000) Phosphorus in the environment: Natural flows and human interferences. *Annual Review of Energy and the Environment,* **25,** 53-88.
Steinfeld, H., Gerber, P., Wassenaar, T., Castel, V., Rosales, M. & De Haan, C. (2006) Livestock's long shadow - environmental issues and options pp. 390. Food and Agriculture Organization of the United Nations, Rome, Italy.
Taranu, Z. & Gregory-Eaves, I. (2008) Quantifying relationships among phosphorus, agriculture, and lake depth at an inter-regional scale. *Ecosystems,* **11,** 715-725.
Townsend, A. R. & Porder, S. (2011) Boundary issues. *Environmental Research Letters,* **6,** 011001.
Townsend, A. R. & Porder, S. (2012) Agricultural legacies, food production and its environmental consequences. *Proceedings of the National Academy of Sciences,* **109,** 5917-5918.
Vitousek, P. M., Naylor, R., Crews, T., David, M. B., Drinkwater, L. E., Holland, E., Johnes, P. J., Katzenberger, J., Martinelli, L. A., Matson, P. A., Nziguheba, G., Ojima, D., Palm, C. A., Robertson, G. P., Sanchez, P. A., Townsend, A. R. & Zhang, F. S. (2009) Nutrient imbalances in agricultural development. *Science,* **324,** 1519-1520.
Wang, F., Sims, J. T., Ma, L., Ma, W., Dou, Z. & Zhang, F. (2011) The phosphorus footprint of china's food chain: Implications for food security, natural resource management, and environmental quality. *J. Environ. Qual.,* **40,** 1081-1089.

APPENDIX 1. LIST OF STUDIES INCLUDED IN THE SOIL PHOSPHORUS LAND-USE LEGACIES META-ANALYSIS FROM CHAPTER 4.

Abubakar, S. M. (1996) Rehabilitation of degraded lands by means of fallowing in a semi-arid area of northern Nigeria. *Land Degradation & Development*, *7*(2), 133-144.

Aweto, A. O. (1981) Secondary Succession and Soil Fertility Restoration in South-Western Nigeria: II. Soil Fertility Restoration. *The Journal of Ecology*, *69*(2), 609.

Baeten, L., Hermy, M., & Verheyen, K. (2009) Environmental limitation contributes to the differential colonization capacity of two forest herbs. *Journal of Vegetation Science*, *20*(2), 209-223.

Baeten, L., Verstraeten, G., De Frenne, P., Vanhellemont, M., Wuyts, K., Hermy, M. & Verheyen, K. (2011) Former land use affects the nitrogen and phosphorus concentrations and biomass of forest herbs. *Plant Ecology*, 212, 901-909.

Brand, J., & Pfund, J. L. (1998) Site-and watershed-level assessment of nutrient dynamics under shifting cultivation in eastern Madagascar. *Agriculture, Ecosystems & Environment*, *71*(1-3), 169-183.

Buschbacher, A. R., Uhl, C, & Serrao, E. A. S. (1988) Abandoned Pastures in Eastern Amazonia . II . Nutrient Stocks in the Soil and Vegetation. *Journal of Ecology*, *76*(3), 682-699.

Compton, J. E., & Boone, R. D. (2000) Long-Term Impacts of Agriculture on Soil Carbon and Nitrogen in New England Forests. *Ecology*, *81*(8), 2314-2330.

Daroub, S. H., Ellis, B. G. & Robertson, G. P. (2001) Effect of Cropping and Low-Chemical Input Systems on Soil Phosphorus Fractions. *Soil Science*, 166, 281-291.

Davidson, E. A., de Carvalho, C. J. R., Figueira, A. M., Ishida, F. Y., Ometto, J. P. H. B., Nardoto, G. B., Saba, R. T., Hayashi, S. N., Leal, E. C., Vieira, I. C. G. & Martinelli, L. A. (2007) Recuperation of nitrogen cycling in Amazonian forests following agricultural abandonment. *Nature*, 447, 995-998.

Dechert, G., Veldkamp, E., & Anas, I. (2004) Is soil degradation unrelated to deforestation? Examining soil parameters of land use systems in upland Central Sulawesi, Indonesia. *Plant and Soil*, *265*(1-2), 197-209.

Degn, H. (2001) Succession from farmland to heathland: a case for conservation of nature and historic farming methods. *Biological Conservation*, *97*(3), 319-330.

de Keersmaeker, L., Martens, L., Veryheyen, K., Hermy, M., de Schrijver, A., Lust, N. (2004) Impact of soil fertility and insolation on diversity of herbaceous woodland species colonizing afforestations in Muizen forest (Belgium). *Forest Ecology and Management*, *188*(1-3), 291-304.

Diekmann, L. O., Lawrence, D., & Okin, G. S. (2007) Changes in the spatial variation of soil properties following shifting cultivation in a Mexican tropical dry forest. *Biogeochemistry*, *84*(1), 99-113.

Dormaar, J. F., Adams, B. W., & Willms, W. D. (1994) Effect of Grazing and Abandoned Cultivation on a Stipa-Bouteloua Community. *Journal of Range Management*, *47*(1), 28.

Duguy, B., Rovira, P., & Vallejo, R. (2007) Land-use history and fire effects on soil fertility in eastern Spain. *European Journal of Soil Science*, *58*(1), 83-91.

Dyer, J. M. (2010) Land-use legacies in a central Appalachian forest: differential response of trees and herbs to historic agricultural practices. *Applied Vegetation Science*, *13*(2), 195-206.

Eberhardt, R. W., Foster, D. R., Motzkin, G., & Hall, B. (2003) Conservation of Changing Landscapes: Vegetation and Land-Use History of Cape Cod National Seashore. *Ecological Applications*, *13*(1), 68-84.

Falkengren-Grerup, U., Brink, D., & Brunet, J. (2006) Land use effects on soil N, P, C and pH persist over 40–80 years of forest growth on agricultural soils. *Forest Ecology and Management*, *225*(1-3), 74-81.

Feldpausch, T. R., Rondon, M. A., Fernandes, E. C. M., Riha, S. J., & Wandelli, E. (2004) Carbon and Nutrient Accumulation in Secondary Forests Regenerating on Pastures in Central Amazonia. *Ecological Applications*, *14*(sp4), 164-176.

Fisher, R. F. (1995) Amelioration of Degraded Rain Forest Soils by Plantations of Native Trees. *Soil Science Society of America Journal*, *70*(1), 305-a.

Flinn, K. M., & Marks, P L. (2007) Agricultural legacies in forest environments: tree communities, soil properties, and light availability. *Ecological Applications*, *17*(2), 452-63.

Flinn, K. M., Vellend, M., & Marks, P.L. (2005) Environmental causes and consequences of forest clearance and agricultural abandonment in central New York, USA. *Journal of Biogeography*, *32*(3), 439-452.

Foote, R. L., & Grogan, P. (2010) Soil Carbon Accumulation During Temperate Forest Succession on Abandoned Low Productivity Agricultural Lands. *Ecosystems*, *13*(6), 795-812.

Fraterrigo, J. M., Turner, M. G., Pearson, S. M., & Dixon, P. (2005) Effects of Past Land Use on Spatial Heterogeneity of Soil Nutrients in Southern Appalachian Forests. *Ecological Monographs*, *75*(2), 215-230.

Frizano, J., Vann, D. R., Johnson, A. H., Johnson, C. M., Vieira, I. C. G., & Zarin, D. J. (2003). Labile Phosphorus in Soils of Forest Fallows and Primary Forest in the Bragantina Region, Brazil. *Biotropica*, *35*(1), 2.

Genxu, W., Haiyan, M., Ju, Q., & Juan, C. (2004) Impact of land use changes on soil carbon, nitrogen and phosphorus and water pollution in an arid region of northwest China. *Soil Use and Management*, *20*(1), 32-39.

Giddens, K M; Parfitt, R. L., & Percival, H. J. (1997) Comparison of some soil properties under Pinus radiata and improved pasture. *New Zealand Journal of Agricultural Research*, *40*, 409-416.

Gong, J., Chen, L., Fu, B., Huang, Y., Huang, Z., & Peng, H. (2006) Effect of land use on soil nutrients in the loess hilly area of the Loess Plateau, China. *Land Degradation & Development*, *17*(5), 453-465.

Graae, B. (2003) Vegetation and soil differences in ancient opposed to new forests. *Forest Ecology and Management*, *177*(1-3), 179-190.

Grossmann, E. B., & Mladenoff, D. J. (2008) Farms, fires, and forestry: Disturbance legacies in the soils of the Northwest Wisconsin (USA) Sand Plain. *Forest Ecology and Management*, *256*(4), 827-836.

Guariguata, M. R., Chazdon, R. L., Denslow, J. S., Dupuy, J. M., & Anderson, L. (1997) Structure and floristics of secondary and old-growth forest stands in lowland Costa Rica. *Plant Ecology*, 132, 107-120.

Holl, K. D. (1999) Factors Limiting Tropical Rain Forest Regeneration in Abandoned Pasture: Seed Rain, Seed Germination, Microclimate, and Soil. *Biotropica*, *31*(2), 229-242.

Hooper, E., Legendre, P., & Condit, R. (2005) Barriers to forest regeneration of deforested and abandoned land in Panama. *Journal of Applied Ecology*, *42*(6), 1165-1174.

Hughes, R. F., Kauffman, J. B., & Jaramillo, V. J. (1999). Biomass , Carbon , and Nutrient Dynamics of Secondary Forests in a Humid Tropical Region of México. *Ecology*, *80*(6), 1892-1907.

Hughes, R. F., Kauffman, J. B., & Cummings, D. L. (2000) Fire in the Brazilian Amazon. *Oecologia*, *124*(4), 574-588.

Johnson, N. C., Zak, D. R., Tilman, D., & Pfleger, F. L. (1991) Dynamics of vesicular-arbuscular mycorrhizae during old field succession. *Oecologia*, *86*, 349-358.

Kalisz, P. J. (1986) Soil Properties of Steep Appalachian Old Fields. *Ecology*, 67, 1011-1023.

Kalinina, O., Goryachkin, S. V., Karavaeva, N. A., Lyuri, D. I., Najdenko, L., & Giani, L. (2009) Self-restoration of post-agrogenic sandy soils in the southern Taiga of Russia: Soil development, nutrient status, and carbon dynamics. *Geoderma*, *152*(1-2), 35-42.

Kang, B. T., Salako, F. K., Akobundu, I. O., Pleysier, J. L. & Chianu, J. N. (1997) Amelioration of a degraded Oxic Paleustalf by leguminous and natural fallows. *Soil Use and Management*, 13, 130-136.

Koerner, W., Dupouey, J., Dambrine, E. & Benoit, M. (1997) Influence of past land use on the vegetation and soils of present day forest in the Vosges mountains, France. *Journal of Ecology*, 85(3), 351-358.

Kuznetsova, I. V., Tikhonravova, P. I., & Bondarev, A. G. (2009) Changes in the properties of cultivated gray forest soils after their abandoning. *Eurasian Soil Science*, *42*(9), 1062-1070.

Lauber, C. L., Strickland, M. S., Bradford, M. a, & Fierer, N. (2008) The influence of soil properties on the structure of bacterial and fungal communities across land-use types. *Soil Biology and Biochemistry*, *40*(9), 2407-2415.

Lawrence, D., & Schlesinger, W. H. (2001) Changes in Soil Phosphorus during 200 Years of Shifting Cultivation in Indonesia. *Ecology*, *82*(10), 2769-2780.

Lawrence, D., D'Odorico, P., Diekmann, L., Delonge, M., Das, R., & Eaton, J. (2007) Ecological feedbacks following deforestation create the potential for a catastrophic ecosystem shift in tropical dry forest. *Proceedings of the National Academy of Sciences of the United States of America*, *104*(52), 20696-701.

Lee, C., You, Y., & Robinson, G. R. (2002) Secondary Succession and Natural Habitat Restoration in Abandoned Rice Fields of Central Korea. *Restoration Ecology*, *10*(2), 306-314.

Lemenih, M. (2004) Comparison of soil attributes under Cupressus lusitanica and Eucalyptus saligna established on abandoned farmlands with continuously cropped farmlands and natural forest in Ethiopia. *Forest Ecology and Management*, *195*(1-2), 57-67.

Lemma, B., & Olsson, M. (2006) Soil δ15N and nutrients under exotic tree plantations in the southwestern Ethiopian highlands. *Forest Ecology and Management*, *237*(1-3), 127-134.

Leprun, J.-C., Grouzis, M., & Randriambanona, H. (2009) Post-cropping change and dynamics in soil and vegetation properties after forest clearing: Example of the semi-arid Mikea Region (southwestern Madagascar). *Comptes Rendus Geosciences*, *341*(7), 526-537.

Lindell, L., Aström, M., & Oberg, T. (2010) Land-use versus natural controls on soil fertility in the Subandean Amazon, Peru. *The Science of the total environment*, *408*(4), 965-75.

Liu, X., Li, F.-M., Liu, D.-Q., & Sun, G.-J. (2010) Soil Organic Carbon, Carbon Fractions and Nutrients as Affected by Land Use in Semi-Arid Region of Loess Plateau of China. *Pedosphere*, *20*(2), 146-152.

Magid, J. (1993). Vegetation effects on phosphorus fractions in set-aside soils. *Plant and Soil*, *149*(1), 111-119.

Manlay, R. J., Masse, D., Chotte, J.-luc, Feller, C., Kairé, M., Fardoux, J., & Pontanier, R. (2002) Carbon, nitrogen and phosphorus allocation in agro-ecosystems of a West African savanna II. The soil component under semi-permanent cultivation. *Agriculture, Ecosystems & Environment*, *88*, 233-248.

Markewitz, D, & Sartori, F. (2002) Soil change and carbon storage in longleaf pine stands planted on marginal agricultural lands. *Ecological applications*, *12*(5), 1276-1285.

Markewitz, Daniel, Davidson, E., Moutinho, P., & Nepstad, D. (2004) Nutrient Loss and Redistribution After Forest Clearing on a Highly Weathered Soil in Amazonia. *Ecological Applications*, *14*(sp4), 177-199.

Matlack, G. R. (2009) Long-term changes in soils of second-growth forest following abandonment from agriculture. *Journal of Biogeography*, *36*(11), 2066-2075.

Montagnini, F., Fanzeres, A., & Vinha, S. G. D. (1995) The Potentials of 20 Indigenous Tree Species for Soil Rehabilitation in the Atlantic Forest Region of Bahia, Brazil. *The Journal of Applied Ecology*, *32*(4), 841.

Moreira, F. M. D. S., Nóbrega, R. S. A., Jesus, E. D. C., Ferreira, D. F., & Pérez, D. V. (2009) Differentiation in the fertility of Inceptisols as related to land use in the upper Solimões river region, western Amazon. *The Science of the total environment*, *408*(2), 349-55.

Morris, Sherri J., Bohm, S., Haile-Mariam, S., & Paul, E. A. (2007). Evaluation of carbon accrual in afforested agricultural soils. *Global Change Biology*, *13*(6), 1145-1156.

Motavalli, P. P. & McConnell, J. (1998) Land Use and Soil Nitrogen Status in a Tropical Pacific Island Environment. *Journal of Environmental Quality*, 27, 119-123.

Odum, E. P., Pinder J. E., and Christiansen T. A. (1984) Nutrient Losses from Sandy Soils during Old-field Succession. *American Midland Naturalist, 111*(1), 148-154.

Ogunkunle, A. O. & Eghaghara, O. O. (1992) Influence of land use on soil properties in a forest region of Southern Nigeria. *Soil Use and Management*, 8, 121-124.

Orczewska, A. (2009) The impact of former agriculture on habitat conditions and distribution patterns of ancient woodland plant species in recent black alder (Alnus glutinosa (L.) Gaertn.) woods in south-western Poland. *Forest Ecology and Management*, *258*(5), 794-803.

Paul, E. A., Morris, S J, Six, J., Paustian, K., & Gregorich, E. G. (2002) Interpretation of Soil Carbon and Nitrogen Dynamics in Agricultural and Afforested Soils, 1620-1629.

Podrázský, V., Remeš, J., Hart, V., & Moser, W. K. (2009) Production and humus form development in forest stands established on agricultural lands – Kostelec nad Černými lesy region, *Journal of Forest Science, 55*(7), 299-305.

Pywell, R. F., Webb, N. R. & Putwain, P. D. (1994) Soil fertility and its implications for the restoration of heathland on farmland in Southern Britain. *Biological Conservation*, 70, 169-181.

Ralte, V., Pandey, H. N., Barik, S. K., Tripathi, R. S., & Prabhu, S. D. (2005) Changes in microbial biomass and activity in relation to shifting cultivation and horticultural practices in subtropical evergreen forest ecosystem of north-east India. *Acta Oecologica*, *28*, 163-172.

Ramakrishnan, P. & Toky, O. (1981) Soil nutrient status of hill agro-ecosystems and recovery pattern after slash and burn agriculture (Jhum) in north-eastern India. *Plant and Soil*, 60, 41-64.

Ritter, E. (2009) Development of bioavailable pools of base cations and P after afforestation of volcanic soils in Iceland. *Forest Ecology and Management*, *257*, 1129-1135.

Ritter, E. (2007) Carbon, nitrogen and phosphorus in volcanic soils following afforestation with native birch (Betula pubescens) and introduced larch (Larix sibirica) in Iceland. *Plant and Soil*, 295(1-2), 239-251.

Roder, W., Phengchanh, S. & Keoboulapha, B. (1995) Relationships between soil, fallow period, weeds and rice yield in slash-and-burn systems of Laos. *Plant and Soil*, 176(1), 27-36.

Römermann, C., Dutoit, T., Poschlod, P. & Buisson, E. (2005) Influence of former cultivation on the unique Mediterranean steppe of France and consequences for conservation management. *Biological Conservation*, 121, 21-33.

Ross, D. (1999) Land-use change: effects on soil carbon, nitrogen and phosphorus pools and fluxes in three adjacent ecosystems. *Soil Biology and Biochemistry*, *31*(6), 803-813.

Ruecker, G., Schad, P., Alcubilla, M. M., & Ferrer, C. (1998) Natural regeneration of degraded soils and site changes on abandoned agricultural terraces in Mediterranean Spain. *Land Degradation & Development*, *188*, 179-188.

Sánchez-de León, Y. & Zou, X. (2004) Plant influences on native and exotic earthworms during secondary succession in old tropical pastures. *Pedobiologia*, 48, 215-226.

Sandoval-Pérez, A. L., Gavito, M. E., García-Oliva, F. & Jaramillo, V. J. (2009) Carbon, nitrogen, phosphorus and enzymatic activity under different land uses in a tropical, dry ecosystem. *Soil Use and Management*, 25, 419-426.

Sciama, D., Augusto, L., Dupouey, J.-L., Gonzalez, M., & Moares Domínguez, C. (2009) Floristic and ecological differences between recent and ancient forests growing on non-acidic soils. *Forest Ecology and Management*, *258*(5), 600-608.

Standish, R. J., Cramer, V. a, Hobbs, R. J., & Kobryn, H. T. (2006) Legacy of Land-Use Evident in Soils of Western Australia's Wheatbelt. *Plant and Soil*, *280*(1-2), 189-207.

Switzer, G. L., Shelton, M. G., & Nelson, L. (1979) Successional Development of the Forest Floor and Soil Surface on Upland Sites of the East Gulf Coastal Plain. *Ecology*, *60*(6), 1162-1171.

Tarrega, R., Calvo, L., Taboada, A., Garciatejero, S., & Marcos, E. (2009) Abandonment and management in Spanish dehesa systems: Effects on soil features and plant species richness and composition. *Forest Ecology and Management*, *257*(2), 731-738.

Tiessen, H., Salcedo, I., & Sampaio, E. (1992) Nutrient and soil organic matter dynamics under shifting cultivation in semi-arid northeastern Brazil. *Agriculture, Ecosystems & Environment*, *38*(3), 139-151.

Uhl, C. (1987) Factors Controlling Succession Following Slash-and-Burn Agriculture in Amazonia. *Journal of Ecology*, *75*(2), 377-407.

Valdespino, P; Romulado, R; Cadenazzi, L; Campo, J. (2009) Phosphorus cycling in primary and secondary seasonally dry tropical forests in Mexico. *Annals of Forest Science*, 66(1), 107.

Valtinat, K., Bruun, H. H., & Brunet, Jorg. (2008) Restoration of oak forest: Effects of former arable land use on soil chemistry and herb layer vegetation. *Scandinavian Journal of Forest Research*, *23*(6), 513-521.

van der Wal, A, Vanveen, J., Smant, W., Boschker, H., Bloem, J., Kardol, P., Vanderputten, W., et al. (2006) Fungal biomass development in a chronosequence of land abandonment. *Soil Biology and Biochemistry*, *38*(1), 51-60.

von Oheimb, G., Härdtle, W., Naumann, P. S., Westphal, C., Assmann, T., & Meyer, H. (2008) Long-term effects of historical heathland farming on soil properties of forest ecosystems. *Forest Ecology and Management*, *255*(5-6), 1984-1993.

Wall, A., Hytonen, J. (2005) Soil fertility of afforested arable land compared to continuously. *Plant and Soil*, *275*, 247-260.

Wang, B., Liu, G. B., Xue, S., & Zhu, B. (2010) Changes in soil physico-chemical and microbiological properties during natural succession on abandoned farmland in the Loess Plateau. *Environmental Earth Sciences*, *62*(5), 915-925.

Wilson, B R; Moffatt, A J; Nortcliff, S. (1997) The nature of three ancient woodland soils in southern England. *Journal of Biogeography*, *24*(5), 633-646.

Yuan, Z.-hua, Lu, X.-guo, & Zhou, J. (2006) Cumulative effects of different cultivating patterns on properties of albic soil in Sanjiang Plain. *Chinese Geographical Science*, *16*(2), 133-140.

Zhang, K., Dang, H., Tan, S., Wang, Z., & Zhang, Q. (2010) Vegetation community and soil characteristics of abandoned agricultural land and pine plantation in the Qinling Mountains, China. *Forest Ecology and Management*, *259*(10), 2036-2047.

Zhao, W., Xiao, H., Liu, Z., & Li, J. (2005) Soil degradation and restoration as affected by land use change in the semiarid Bashang area, northern China. *Catena*, *59*(2), 173-186.

Appendix 2. Chapter 4 model selection results (95% candidate set of models – labile P). Each linear continuous (shown as a numeric parameter value), categorical (shown as '+'), and spline smoother continuous ('+') variable that was retained in a given model is indicated.

Model rank	Intercept	Aridity Index	Clay	FLU	LOA	MaxDepth	Plantation	Region	pHratio	TST	k	AICc	Δ AICc	AIw (weight)
1	-0.64		-0.02			0.01	+	+	+	+	16	468.10	0.00	0.16
2	-0.26		-0.02			0.01		+	+	+	15	469.00	0.88	0.10
3	0.59		-0.02			0.01			+	+	11	470.10	1.95	0.06
4	-0.81	0.00	-0.02			0.01	+	+	+	+	17	470.30	2.14	0.05
5	-0.67	0.01	-0.02			0.01		+	+	+	16	470.40	2.34	0.05
6	0.47		-0.02			0.01	+		+	+	12	471.00	2.84	0.04
7	-0.65		-0.02		+	0.01	+	+	+	+	19	471.10	3.04	0.04
8	-1.05					0.01	+	+	+	+	15	472.00	3.86	0.02
9	-0.51		-0.01				+	+	+	+	15	472.10	4.03	0.02
10	-1.23	0.01	-0.02		+	0.01	+	+	+	+	20	472.10	4.03	0.02
11	-0.62		-0.02	+		0.01	+	+	+	+	18	472.30	4.16	0.02
12	0.54	0.00	-0.02			0.01			+	+	12	472.30	4.18	0.02
13	0.82		-0.02		+	0.01			+	+	14	472.70	4.59	0.02
14	-0.14		-0.02		+	0.01		+	+	+	18	472.80	4.70	0.02
15	-0.68		-0.01			0.01	+	+		+	14	472.80	4.72	0.02
16	-0.25		-0.02	+		0.01		+	+	+	17	472.90	4.84	0.01
17	-0.97	0.02	-0.02		+	0.01		+	+	+	19	473.00	4.91	0.01
18	0.53		-0.02	+		0.01			+	+	13	473.20	5.05	0.01
19	0.52	0.00	-0.02			0.01	+		+	+	13	473.20	5.09	0.01
20	-0.11		-0.01					+	+	+	14	473.30	5.18	0.01
21	0.68		-0.02		+	0.01	+		+	+	15	473.70	5.57	0.01
22	0.69		-0.02						+	+	10	473.70	5.59	0.01
23	-0.62		-0.01		+	0.01	+	+		+	17	473.70	5.59	0.01
24	-0.87						+	+	+	+	14	473.90	5.77	0.01
25	0.38		-0.02	+		0.01	+			+	12	473.90	5.79	0.01
26	0.42		-0.02	+		0.01	+		+	+	14	474.10	6.00	0.01
27	-0.71	0.01	-0.01				+	+	+	+	16	474.20	6.07	0.01
28	-1.15	0.00				0.01	+	+	+	+	16	474.20	6.12	0.01
29	0.56		-0.02				+		+	+	11	474.30	6.17	0.01
30	0.54		-0.02	+		0.01				+	11	474.30	6.20	0.01
31	-0.77	0.00	-0.02	+		0.01	+	+	+	+	19	474.50	6.40	0.01
32	-0.66					0.01		+	+	+	14	474.60	6.45	0.01
33	-0.57	0.01	-0.02					+	+	+	15	474.60	6.46	0.01
34	-0.63	0.01	-0.02	+		0.01		+	+	+	18	474.60	6.48	0.01
35	0.53	0.01	-0.02		+	0.01			+	+	15	474.70	6.58	0.01

Table continued on following page.

Model rank	Intercept	Aridity Index	Clay	FLU	LOA	MaxDepth	Plantation	Region	pHratio	TST	k	AICc	Δ AICc	AIw (weight)
36	-0.54		-0.02	+		0.01	+	+		+	16	474.80	6.67	0.01
37	0.44		-0.02			0.01	+			+	10	475.00	6.86	0.01
38	-1.05					0.01	+	+		+	13	475.00	6.94	0.01
39	-0.61		-0.02	+	+	0.01	+	+	+	+	21	475.10	6.99	0.01
40	-0.64	0.00	-0.01			0.01	+	+		+	15	475.10	7.02	0.01
41	0.61		-0.02	+	+	0.01	+			+	15	475.40	7.32	0.00
42	0.53	0.00	-0.02	+		0.01			+	+	14	475.50	7.34	0.00
43	0.76		-0.02	+	+	0.01			+	+	16	475.50	7.38	0.00
44	-0.94	0.01	-0.01		+	0.01	+	+		+	18	475.60	7.46	0.00
45	-0.47		-0.02	+	+	0.01	+	+		+	19	475.60	7.53	0.00
46	-0.53		-0.01		+		+	+	+	+	18	475.70	7.57	0.00
47	0.48	0.00	-0.02		+	0.01	+		+	+	16	475.80	7.72	0.00
48	-1.05				+	0.01	+	+	+	+	18	475.80	7.72	0.00
49	0.59	0.00	-0.02						+	+	11	475.90	7.76	0.00
50	0.54	0.00	-0.02	+		0.01	+			+	13	476.00	7.92	0.00
51	-0.54		-0.01				+	+		+	13	476.00	7.93	0.00
52	-1.02	0.00					+	+	+	+	15	476.00	7.94	0.00
53	0.81		-0.02	+	+	0.01				+	14	476.10	8.02	0.00
54	-1.04	0.01				0.01		+	+	+	15	476.20	8.05	0.00
55	-0.47							+	+	+	13	476.20	8.14	0.00
56	-1.18	0.01	-0.02	+	+	0.01	+	+	+	+	22	476.30	8.17	0.00
57	0.51	0.00	-0.02	+		0.01	+		+	+	15	476.40	8.27	0.00
58	-1.17	0.01	-0.01		+		+	+	+	+	19	476.40	8.32	0.00
59	-0.49		-0.01	+			+	+	+	+	17	476.50	8.35	0.00
60	0.63		-0.02	+	+	0.01	+		+	+	17	476.50	8.36	0.00
61	-0.12		-0.02	+	+	0.01		+	+	+	20	476.50	8.40	0.00
62	0.57	0.00	-0.02				+		+	+	12	476.50	8.42	0.00
63	0.59	0.00	-0.02	+		0.01				+	12	476.50	8.44	0.00
64	-1.04			+		0.01	+	+	+	+	17	476.60	8.45	0.00
65	0.68		-0.02		+	0.01	+			+	13	476.70	8.58	0.00
66	-0.86						+	+		+	12	476.70	8.61	0.00
67	0.65		-0.02			0.01				+	9	476.90	8.78	0.00
68	0.91		-0.02		+				+	+	13	477.00	8.84	0.00
69	-0.98				+	0.01	+	+		+	16	477.00	8.85	0.00
70	0.64		-0.02	+					+	+	12	477.00	8.88	0.00
71	-0.91	0.02	-0.02	+	+	0.01		+	+	+	21	477.00	8.91	0.00
72	0.60	0.00	-0.02			0.01	+			+	11	477.00	8.94	0.00
73	-0.01		-0.02	+		0.01		+		+	15	477.10	9.02	0.00
74	-0.54	0.00	-0.02	+		0.01	+	+		+	17	477.10	9.03	0.00
75	-0.97	0.00				0.01	+	+		+	14	477.30	9.17	0.00
76	0.48		-0.02	+			+			+	11	477.30	9.18	0.00
77	-1.53	0.01			+	0.01	+	+	+	+	19	477.40	9.24	0.00

Appendix 3. Chapter 4 model selection results (95% candidate set – total P).
Each linear continuous (numeric parameter value), categorical (+), and spline smoother continuous (+) variable that was retained in a given model is indicated.

Model rank	Intercept	Clay	FLU	LOA	MaxDepth	pHratio	Region	Plantation	TST	k	AICc	Δ AICc	AIw (weight)
1	0.92				-0.005			+	+	13	80.66	0.00	0.25
2	0.62				-0.004	0.28		+	+	14	82.17	1.50	0.12
3	0.87				-0.005		+	+	+	14	82.89	2.23	0.08
4	0.91	0.00			-0.005			+	+	14	83.06	2.40	0.07
5	0.88							+	+	12	84.17	3.51	0.04
6	0.51				-0.004	0.32	+	+	+	15	84.19	3.53	0.04
7	0.91		+		-0.005			+	+	15	84.45	3.79	0.04
8	0.61	0.00			-0.004	0.28		+	+	15	84.59	3.92	0.03
9	0.24					0.54	+	+	+	14	84.84	4.18	0.03
10	0.92			+	-0.005			+	+	16	84.91	4.25	0.03
11	0.86	0.00			-0.005		+	+	+	15	85.31	4.65	0.02
12	0.36	0.00				0.50		+	+	14	85.39	4.73	0.02
13	0.61		+		-0.004	0.28		+	+	16	86.02	5.36	0.02
14	0.85						+	+	+	13	86.44	5.77	0.01
15	0.88	0.00						+	+	13	86.55	5.89	0.01
16	0.49	0.00			-0.004	0.32	+	+	+	16	86.61	5.95	0.01
17	0.65			+	-0.004	0.25		+	+	17	86.68	6.01	0.01
18	0.85		+		-0.005		+	+	+	16	86.7	6.04	0.01
19	0.34		+			0.50		+	+	15	86.87	6.21	0.01
20	0.90	0.00	+		-0.005			+	+	16	86.89	6.22	0.01
21	0.86			+	-0.005		+	+	+	17	87.14	6.47	0.01
22	0.22	0.00				0.55	+	+	+	15	87.25	6.58	0.01
23	0.92	0.00		+	-0.005			+	+	17	87.43	6.76	0.01
24	0.38			+		0.48		+	+	16	87.59	6.93	0.01
25	0.87		+					+	+	14	87.99	7.33	0.01
26	0.48		+		-0.004	0.32	+	+	+	17	88.04	7.38	0.01
27	0.88			+				+	+	15	88.46	7.80	0.01
28	0.59	0.00	+		-0.004	0.28		+	+	17	88.47	7.80	0.01
29	0.52			+	-0.004	0.30	+	+	+	18	88.68	8.01	0.00
30	0.20		+			0.54	+	+	+	16	88.69	8.02	0.00

APPENDIX 4. DATA SOURCES FOR LAKE TOTAL PHOSPHORUS ANALYSIS

Studies from which lake TP data was compiled during the literature search in Chapter 5:

Ballot, A., K. Kotut, E. Novelo, and L. Krienitz. (2009) Changes of phytoplankton communities in Lakes Naivasha and Oloidien, examples of degradation and salinization of lakes in the Kenyan Rift Valley. Hydrobiologia 632 (1):359-363.

Davies, J.-M., Nowlin, W. H. & Mazumder, A. (2004) Temporal changes in nitrogen and phosphorus codeficiency of plankton in lakes of coastal and interior british columbia. Canadian Journal of Fisheries and Aquatic Sciences, 61, 1538-1551.

Diaz, M., F. Pedrozo, C. Reynolds, and P. Temporetti (2007) Chemical composition and the nitrogen-regulated trophic state of Patagonian lakes. Limnologica 37 (1):17-27.

EEA (2011) Waterbase - Lakes. Lakes dataset version: May 2011. European Environment Agency, Copenhagen. Available at: http://www.eea.europa.eu/data-andmaps/data/waterbase-lakes-7 (visited August 2011).

EPA (2007) National Lakes Assessment Data. Lakes dataset version: 2007. U.S. Environmental Protection Agency, Washington D.C. Available at: http://water.epa.gov (visited August 2011).

Finkel, Z. V., Vaillancourt, C. J., Irwin, A. J., Reavie, E. D. & Smol, J. P. (2009) Environmental control of diatom community size structure varies across aquatic ecosystems. Proceedings of the Royal Society B: Biological Sciences, 276, 1627-1634.

Havens, K. E., T. Kukushima, P. Xie, T. Iwakuma, R. T. James, N. Takamura, T. Hanazato, and T. Yamamoto (2001) Nutrient dynamics and the eutrophication of Shallow lakes Kasumigaura (Japan), Donghu (PR China), and Okeechobee (USA). Environmental Pollution 111 (2):263-272.

Huszar, V. L. M., L. H. S. Silva, M. Marinho, P. Domingos, and C. L. Sant'Anna. (2000) Cyanoprokaryote assemblages in eight productive tropical Brazilian waters. Hydrobiologia 424:67-77.

Ishii, Y., S. Harigae, S. Tanimoto, T. Yabe, T. Yoshida, K. Taki, N. Komatsu, K. Watanabe, M. Negishi, and H. Tatsumoto (2010) Spatial variation of phosphorus fractions in bottom sediments and the potential contributions to eutrophication in shallow lakes. Limnology 11 (1):5-16.

Kang, S. A., X. R. Peng, L. Zhang, M. Liu, Y. Zhang, and Ieee (2009) The assessment of the present eutrophication status and characteristic analysis of Xingkai Lake, 2009 3rd International Conference on Bioinformatics and Biomedical Engineering, Vols 1-11. New York.

Kitaka, N., Harper, D. M. & Mavuti, K. M. (2002) Phosphorus inputs to Lake Naivasha, Kenya, from its catchment and the trophic state of the lake. Hydrobiologia, 488, 73-80.

Kosten, S., V. L. M. Huszar, N. Mazzeo, M. Sche_er, L. D. L. Sternberg, and E. Jeppesen (2009) Lake and watershed characteristics rather than climate influence nutrient limitation in shallow lakes. Ecological Applications 19 (7):1791-1804.

Lagomarsino, L., G. L. Perez, R. Escaray, J. Bustingorry, and H. E. Zagarese. (2011) Weather variables as drivers of seasonal phosphorus dynamics in a shallow hypertrophic lake (Laguna Chascomus, Argentina). Fundamental and Applied Limnology 178 (3):191-201.

Masson, S., B. Pinel-Alloul, and V. H. Smith (2000) Total phosphorus-chlorophyll

a size fraction relationships in southern Quebec lakes. Limnology and Oceanography 45(3):732-740.
Nowlin, W. H., J. M. Davies, and A. Mazumder (2007) Planktonic phosphorus pool sizes and cycling efficiency in coastal and interior British Columbia lakes. Freshwater Biology 52 (5):860-877.
Nyakoojo, C., and S. M. Byarujali (2010) An ecological study of two shallow, equatorial lakes: Lake Mburo and Lake Kachera, Uganda. African Journal of Ecology 48(4):860-864.
Oduor, S. O., Schagerl, M. & Mathooko, J. M. (2003) On the limnology of Lake Baringo (Kenya): I.Temporal physico-chemical dynamics. Hydrobiologia, 506-509, 121-127.
Pacheco, J., C. Iglesias, M. Meerhof, C. Fosalba, G. Goyenola, F. Teixeira-de Mello, S. Garcia, M. Gelos, and F. Garcia-Rodriguez (2010) Phytoplankton community structure in five subtropical shallow lakes with different trophic status (Uruguay): a morphology based approach. Hydrobiologia 646 (1):187-197.
Rennella, A. M., and R. Quiros (2006) The effects of hydrology on plankton biomass in shallow lakes of the Pampa Plain. Hydrobiologia 556:181-191.
Sheela, A. M., J. Letha, and S. Joseph (2011) Environmental status of a tropical lake system. Environmental Monitoring and Assessment 180 (1-4):427-449.
Shinneman, A. L. C., J. E. Almendinger, C. E. Umbanhowar, M. B. Edlund, and S. Nergui (2009) Paleolimnologic Evidence for Recent Eutrophication in the Valley of the Great Lakes (Mongolia). Ecosystems 12 (6):944-960.
Silvoso, J., I. Izaguirre, and L. Allende (2011) Picoplankton structure in clear and turbid eutrophic shallow lakes: A seasonal study. Limnologica 41 (3):181-190.
Taranu, Z. & Gregory-Eaves, I. (2008) Quantifying relationships among phosphorus, agriculture, and lake depth at an inter-regional scale. Ecosystems, 11, 715-725.
Taranu, Z. E., Zurawell, R., Pick, F. & Gregory-Eaves, I. (In press) Climate and nutrients as drivers of contemporary cyanobacteria dynamics across lakes: Relative importance of drivers varies with lake mixis. Global Change Biology.
Trolle, D., D. P. Hamilton, C. Hendy, and C. Pilditch (2008) Sediment and nutrient accumulation rates in sediments of twelve New Zealand lakes: inuence of lake morphology, catchment characteristics and trophic state. Marine and Freshwater Research 59(12):1067-1078.
Trolle, D., G. W. Zhu, D. Hamilton, L. C. Luo, C. McBride, and L. Zhang (2009) The influence of water quality and sediment geochemistry on the horizontal and vertical distribution of phosphorus and nitrogen in sediments of a large, shallow lake. Hydrobiologia 627 (1):31-44.
Valdovinos, C., and P. Pedreros (2007) Geographic variations in shell growth rates of the mussel Diplodon chilensis from temperate lakes of Chile: Implications for biodiversity conservation. Limnologica 37 (1):63-75.
Vermaire, J. C., Prairie, Yves T. & Gregory-Eaves, I. (2012) Diatom-inferred decline of macrophyte abundance in lakes of southern quebec, canada. Canadian Journal of Fisheries and Aquatic Sciences, 69, 511-524.
Villalobos, L., O. Parra, M. Grandjean, E. Jaque, S. Woel, and H. Campos (2003) A study of the river basins and limnology of five humic lakes on Chiloe Island. Revista Chilena De Historia Natural 76 (4):563-590.
Wang, Maolan, Wenbin Zhou, and Yongping Ai (2011) The Hydrochemistry and

Nutrient Distribution in Poyang Lake, China. Bioinformatics and Biomedical Engineering, (iCBBE) 2011 5th International Conference, 10-12 May 2011.
Xie, Liqiang, Ping Xie, Sixin Li, Huijuan Tang, and Hong Liu (2003) The low TN:TP ratio, a cause or a result of Microcystis blooms? Water Research 37 (9):2073-2080.
Yang, X. D., J. Shen, X. H. Dong, E. F. Liu, and S. M. Wang (2006) Historical trophic evolutions and their ecological responses from shallow lakes in the middle and lower reaches of the Yangtze River: Case studies on Longgan Lake and Taibai Lake. Science in China Series D-Earth Sciences 49:51-61.