The role of feeding ecology in persistent organic pollutant accumulation of killer whales across the North Atlantic Ocean

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July 2023

A thesis submitted to McGill University in partial fulfillment of the requirements of the degree of Doctor of Philosophy (Ph.D.) Renewable Resources

Photo taken in Vestmannaeyjar, Iceland: Anaïs Remili / The Icelandic Orca Project

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LIST OF ABBREVIATIONS

Accronym	Definition
AA	Amino Acid
AIC	Akaike Information Criterion
ANOVA	Analysis Of Variance
ARTS	Aerial Remote Tag System
ASE	Accelerated Solvent Extraction
BB	Brominated Biphenyl
BDE	Brominated Diphenyl Ether
BEH-TEBP	Bis(2-Ethylhexyl)-Tetrabromophthalate
BFR	Brominated Flame Retardants
BTBPE	1,2-Bis(2,4,6-Tribromophenoxy)Ethane
CB	Chlorinated Biphenyl
CC	Calibration Coefficient
CHL	Chlordane
CI	Confidence Interval
CYP450	Cytochrome P450
DBDPE	Decabromodiphenyl Ethane
DBE-DBCH	Decabromodiphenyl
DBHCTD	Hexachlorocyclopentadienyl-Dibromocyclooctane
DCM	Dichloromethane
DDC or DDC-CO	Dechlorane Plus
DDD	Dichlorodiphenyldichloroethane
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
DK	Denmark
DNA	Deoxyribonucleic Acid
DPTE	2,3-Dibromopropyl-2,4,6-Tribromophenyl Ether
ECCC	Environment And Climate Change Canada
ECNI	Electron Capture Negative Ionization
EH-TBB	2-Ethylhexyl-2,3,4,5-Tetrabromobenzoate
EI	Electron Ionization
EPA	Environmental Protection Agency
FA	Fatty Acid
FAME	Fatty Acid Methyl Esters
FID	Flame Ionization Detection
FR	Flame Retardants
FRQNT	Fond De Recherches Du Québec Nature Et Technologie
GC	Gas Chromatograph
GLM	Generalized Linear Model
GPC	Gas Permeation Chromatography
HBB	Hexabromobenzene

HBCD or HBCDD	Hexabromocyclododecane
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
HRMS	High Resolution Mass Spectrometry
ID	Identification
IS	Internal Standard
KL	Kullback Leibler
KW	Killer Whale
LOD	Limit Of Detection
LOPO	Leave One Prey Out
MLOD	Method Limit Of Detection
MLOQ	Method Limit Of Quantitation
MS	Mass Spectrometer
MUFA	Mono-Unsaturated Fatty Acid
NA	North Atlantic
NIST	National Institute Of Standards And Technology
NOAA	National Oceanic And Atmospheric Administration
NSERC	Natural Sciences And Engineering Research Council
NWRC	National Wildlife Research Center
OBTMPI	2,3,4,5-Tetrabromophenyl)-1H-Indene
OC	Organochlorines
ON	Ontario
PBB	Polybrominated Biphenyl
PBDE	Polybrominated Diphenyl Ethers
PBEB	Pentabromoethylbenzene
PBP	Pentabromophenol
PBT	Polybutylene Terephthalate
PCA	Principal Component Analysis
PCB	Polychlorinated Biphenyl
PCDD	Polychlorinated Dibenzodioxins
PCDF	Polychlorinated Dibenzofurans
PFAS	Polyfluoroalkyl Substances
PFH	Perfluorohexane
PFOA	Perfluorooctanoic Acid
PFOS	Perfluorooctane Sulfonic Acid
PFOSF	Perfluorooctane Sulfonyl Fluoride
PON	Paraoxonase
POP	Persistent Organic Pollutants
PUFA	Poly-Unsaturated Fatty Acid
QA	Quality Assessment
QC	Quebec
QFASA	Quantitative Fatty Acid Signature Analysis
QFASAR	Quantitative Fatty Acid Signature Analysis For R

RGPIN	Discovery Grants Program – Individual
SCCP	Short-Chain Chlorinated Paraffins
SE	Standard Error
SEA	Standard Ellipse Area
SFA	Saturated Fatty Acid
SIBER	Stable Isotope Bayesian Ellipses In R
SIM	Selected Ion Monitoring
SPE	Solid-Phase Extraction
SPM	Saint Pierre & Miquelon
SRM	Standard Reference Material
SW	SeaWorld
TBCT	Tetrabromo- O-Chlorotoluene
TBP-AE	Tribromophenyl Allyl Ether
TBX	2,3,5,6-Tetrabromo-P-Xylene
TOC	Table Of Contents
UV	Ultraviolet

GENERAL ABSTRACT

As the oceans' top marine mammal predators, killer whales are threatened by synthetic pollutants like industrial chemicals, flame retardants, and pesticides. These pollutants usually partition into fatty tissues and concentrate through food webs to reach high levels in killer whales. A high accumulation of these chemicals can cause immune and reproductive problems. A recent study even developed models forecasting risk of decline in multiple killer whale populations by 2100 because of these pollutants. Previous studies suggested that feeding ecology plays an important role in the accumulation of contaminants like persistent organic pollutants (POPs).

While most studies have been performed in North Pacific killer whales, off the West coast of North America, research has been lacking in the North Atlantic, especially in the Western North Atlantic. This knowledge gap regarding the feeding habits of killer whales and the potential risks associated with the accumulation of harmful contaminants served as the driving force behind this doctoral project. Over a span of just over four years and four data chapters, our international, collaborative, trans-Atlantic research effort successfully shed light on the feeding ecology of North Atlantic killer whales, their contaminant levels, and the associated risks.

Chapter Two of this doctoral thesis provides a comprehensive overview of the existing literature up until the contributions of this thesis to the field. Chapter Three investigates variation in contaminant exposures based on dietary patterns among Icelandic killer whales. This chapter focuses on individual feeding specialization within the population (obtained through observation and stable isotopes) and uncovers significant variation in contaminant levels among Icelandic killer whales, primarily influenced by dietary preferences (fish-only diets vs. diets consisting of both fish and marine mammals). Chapter Four develops a novel method to precisely determine

the diet composition of wild killer whales. By analyzing archived blubber samples from killer whales housed at Sea World and their prey, I measured the fatty acid compositions of both predators and prey. I then calculated calibration coefficients for use in quantitative fatty signature analysis and validated the method using harvested Greenlandic killer whales with known stomach contents. Chapter Five then applies this new method to ~200 North Atlantic killer whales spanning from the Canadian Arctic to Northern Norway, revealing large differences between and within populations of North Atlantic killer whales. Finally, Chapter Six demonstrates that these dietary differences are the primary factor driving contaminant accumulation among killer whales across this Ocean. It highlights that killer whales feeding on marine mammals, particularly in the Western North Atlantic, face high risks of health effects due to polychlorinated biphenyl burdens. Through examination of the role of feeding habits in the contaminant-associated risks of North Atlantic killer whales, the knowledge provided by this thesis can further motivate policy makers and stakeholders to improve actions to dispose of contaminated waste and underscores the need to prevent the release of new contaminants into the environment to protect the top ocean predator.

RESUME GENERAL

En tant que principaux prédateurs des mammifères marins, les orques sont menacées par les polluants synthétiques tels que les produits chimiques industriels, les retardateurs de flamme et les pesticides. Ces polluants se répartissent généralement dans les tissus adipeux et se concentrent à travers les réseaux alimentaires pour atteindre des niveaux élevés chez les orques. Une forte accumulation de ces produits chimiques peut entraîner des problèmes immunitaires et reproductifs. Une étude récente a même développé des modèles prévoyant un risque de déclin de plusieurs populations d'orques d'ici 2100 à cause de ces polluants. Des études antérieures ont suggéré que l'écologie alimentaire joue un rôle important dans l'accumulation de contaminants tels que les polluants organiques persistants.

Alors que la plupart des études ont été réalisées sur les orques du Pacifique Nord, au large de la côte ouest de l'Amérique du Nord, les recherches ont été insuffisantes dans l'Atlantique Nord, en particulier dans l'Atlantique Nord occidental. Ce manque de connaissances sur les habitudes alimentaires des orques et les risques liés à l'accumulation de contaminants nocifs a été la force motrice de ce projet de doctorat. En l'espace d'un peu plus de quatre ans et de quatre chapitres de données, notre effort de recherche international, collaboratif et transatlantique a permis de faire la lumière sur l'écologie alimentaire des orques de l'Atlantique Nord, leurs niveaux de contaminants et les risques qui y sont associés.

Le chapitre deux de cette thèse de doctorat fournit une vue d'ensemble de la littérature existante jusqu'aux contributions de cette thèse au domaine. Le chapitre trois étudie la variation de l'exposition aux contaminants en fonction des habitudes alimentaires des orques d'Islande. Ce chapitre se concentre sur la spécialisation alimentaire individuelle au sein de la population (obtenue par l'observation et les isotopes stables) et découvre une variation significative des niveaux de contaminants parmi les orques d'Islande, principalement influencée par les

préférences alimentaires (régimes alimentaires à base de poisson uniquement vs. régimes alimentaires composés à la fois de poisson et de mammifères marins). Le chapitre quatre développe une nouvelle méthode pour déterminer avec précision la composition du régime alimentaire des orques sauvages. En analysant des échantillons de graisse archivés d'orques hébergées à SeaWorld et de leurs proies, j'ai mesuré la composition en acides gras des prédateurs et des proies. J'ai ensuite calculé les coefficients de calibration pour l'analyse quantitative des signatures d'acides gras et j'ai validé la méthode en utilisant des orques du Groenland dont le contenu de l'estomac était connu. Le chapitre cinq met ensuite en application cette nouvelle méthode sur environ 200 orques de l'Atlantique Nord, de l'Arctique canadien au nord de la Norvège, et révèle d'importantes variations tant entre les populations d'orques de l'Atlantique Nord que à l'intérieur de celles-ci. Enfin, le chapitre 6 démontre que ces différences alimentaires sont le principal facteur d'accumulation des contaminants chez les orques dans cet océan. Il souligne que les orques qui se nourrissent de mammifères marins, en particulier dans l'ouest de l'Atlantique Nord, sont exposées à des risques élevés d'effets néfastes sur la santé en raison des concentrations de biphényles polychlorés. En examinant le rôle des habitudes alimentaires dans les risques associés aux contaminants chez les orques de l'Atlantique Nord, les connaissances fournies par cette thèse peuvent motiver davantage les décideurs politiques et les parties prenantes à améliorer les mesures d'élimination des déchets contaminés. Ces résultats soulignent la nécessité d'empêcher la libération de nouveaux contaminants dans l'environnement afin de protéger le plus grand prédateur océanique.

ACKNOWLEDGEMENTS

I would like first and foremost to express my most profound gratitude to my advisor, Dr. Melissa McKinney for her unfailing, invaluable support and guidance throughout my entire PhD journey. From the very beginning, Melissa showed unwavering confidence in my abilities and research, which served as a constant source of motivation. She gave me such strong support in my application to McGill University and gave me the opportunity to work on my dream PhD project. Her mentorship has been crucial in helping me reach this significant milestone, and I will never be able to thank her enough for the countless hours she has dedicated to this effort. From listening to my naïve rambling about how quickly I would graduate, to seeing me beaten down with tears in my eyes, she's always watched over me and has made a profound impact on my life and career. Melissa has inspired me to become a better researcher, to think critically, challenge myself, go beyond my limits, and strive for excellence in all aspects of my life. I firmly believe I would not be half the researcher I am today without her guidance. Melissa, thank you from the bottom of my heart for watching out for me, for being a constant source of inspiration, and an extraordinary advisor.

Next, I would like to express my most sincere gratitude for Dr. Robert Letcher, my co-advisor. Rob's support and praise in the early stages of my PhD journey have been instrumental to my success and confidence as an early-career researcher. Working by his side has been an incredible honor. He welcomed me to his lab in Ottawa with open arms and his kindness and empathy made me feel at home, far from home. Rob took me under his wing at multiple conferences, consistently introducing me to colleagues and assisting me in building my own professional network. Thank you, Rob, and thanks for the fun times in Kyoto!

I would also like to thank the third member of my committee, Dr. Nil Basu, for his guidance and invaluable feedback throughout my entire PhD journey. From my department, I

would like to thank Dr. Brian Driscoll for the good laughs and pandemic support, Dr. Jeff Cardille for the good times teaching ENV-301 together, as well as Dr. Kyle Elliott for his precious feedback on my proposal.

Next, I would like to express my warmest gratitude to Dr. Filipa Samarra and all the volunteers of the Icelandic Orca Project. Filipa has been a constant and unfailing source of inspiration, support, and kind encouragements throughout the years, and I will never thank her enough for her invaluable role in my development as a young scientist. She has turned me into a more critical thinker and a rigorous researcher. I can also never thank her enough for the opportunities to go in see my favorite animals in the field and build a serious fieldwork and outreach skillset.

I would like to sincerely thank Dr. Jean-Pierre Desforges, for believing in me, when I had nothing to show for, and for introducing me to Dr. McKinney. He was key in me securing the PhD project of my dreams, and I will cherish all his precious advice for the rest of my career. I am extremely grateful to Dr. Jeremy Kiszka for believing in me and always offering me exciting new opportunities, in science communication or in research. I would also like to thank Dr. Rune Dietz, and Dr. Christian Sonne for their constant unfailing support and words of encouragement on all my manuscripts. Thank you to Dr. Eve Jourdain, who taught me about being a strong dedicated woman in the killer whale research world. Our friendship and her precious advice will keep motivating me throughout my career. Thanks should also go to Dr. Audun Rikardsen and Lisa Kettemer for welcoming me to Tromsø in the early stages of my PhD and trusting me to take care of their precious killer whale biopsies. Big thanks to Dr. Steve Ferguson, Dr. Cory Matthews and Dr. Cortney Watt from DFO for their trust, support and their helpful advice throughout my PhD journey. I would also like to thank my additional co-authors Dr. Aqqalu Rosing-Asvid, Dr. Katrine Borgå, Dr. Anders Ruus, Dr. Sara Iverson, Dr. Denis Roy, Dr. Sandra

Grandquist, and the late Dr. Gísli Víkingsson, for their feedback and words of encouragement throughout this entire adventure. I would also like to warmly thank Jared Towers, Gary Sutton and Tasli Shaw from Bay Cetology, for their precious advice on camera equipment, science communication and future career path. Thank you to Dr. Dawn Noren, Dr. Irv Schultz, Dr. Paul Chittaro and Dr. Brad Hanson from NOAA for their support in securing funding for my postdoc. Thanks to Dr. Tanya Brown for giving me the opportunity to work with her as a postdoc in the future. Big thanks to Dr. Krishna Das, Dr. Adrian Covaci and Dr. Govindan Malarvannan, and Dr. John Wise for their support in my master's journey. I would not be writing these lines without the opportunities they gave me. I missed many names in this paragraph, and I would like to extend my thanks to all the amazing researchers I met at various events/conferences who have helped shape the scientist I am today.

A scientist is nothing without their lab, and I would like to express my most heartfelt gratitude to my dear friends and members of my lab(s): Dave Blair, Adam Pedersen, Nadia Facciola, Kailee Hopkins, Haley Land-Miller, Ambar Maldonado Rodriguez, Rose Lacombe, Megan Franz, Chantel Michelson, Keenin Coombs, Kiah Lee, Nasrin Golzadeh and our visiting undergraduate students. Through COVID, and through all the struggles, their support meant the world to me, and I am forever grateful to have such a wonderful and caring group of friends.

In addition, I would like to express my gratitude to my dear friend and colleague, Naomi Mathew, who played a pivotal role in the creation of Whale Scientists and has been instrumental in launching my science communication journey. My second life as a science communicator has helped me become a better scientist and has brought me the skills to share my research in an entertaining way. I would also like to thank all the incredible people I've met through science communication, who have motivated me to explore science outside of Academia and made me into the early-career scientist I am today.

Thanks to the Fonds the Recherche du Québec, the Natural Resource Sciences department, the CREATE Environmental Initiative program, the Quebec Center for Biodiversity Science, the Society of Environmental Toxicology and Chemistry, and ECOTOQ for funding my PhD adventures.

Thanks to all my friends from Montréal, form France, from Europe and elsewhere, as well as all my marine mammals, SETAC, and NRS friends, for keeping me somewhat sane during this grueling process, and for picking me up and giving me good pints when I needed it most. Special mention to Alexis Trinquet and Hannah Lieberman!

The biggest thanks in the world go to my loving, caring, inspiring, and supportive partner, Tristan. I am eternally grateful for his unwavering patience and constant words of affirmation, which provided me with a safe space to conduct this project. What makes his support even more remarkable is that he demonstrated such care and dedication while navigating the challenging journey of his own PhD, here at McGill University. I am immensely proud of him for his incredible ability to excel in his own projects while being a pillar of support for me. Next, I am grateful to my fur child, Nala, whom I brought back from France to Quebec one week before the pandemic lockdown. She became my source of sanity during the pandemic, especially when I found myself stranded in Quebec, far from my family. Finally, I want to express love and thanks to my family members, each of whom played a significant role in supporting me *(Enfin, je tiens à exprimer mon amour et ma gratitude envers les membres de ma famille, chacun d'entre eux ayant joué un rôle important dans cette aventure)*. I extend a special mention to my dear sister, Ambre, my amazing and strong mother, Nadine, my stepdad, Camille, my American mom, Jeaneane, my American dad, Jim, my second mom, Elizabeth, and my second dad, Kenneth, who all contributed to my strength and motivation, and showered me with love and patience *(tous ont contribué à ma force et à ma motivation, et m'ont comblé d'amour et de patience)*. I would also

like to acknowledge Mammie, Papi, Mémé, Mémé Janine, Joan, Jo, Ja, Maddie and Orie. Their support propelled me forward during the darkest times and eventually made me reach for the light at the end of the tunnel. This thesis is dedicated to all of them *(Leur soutien m'a fait avancer dans les moments les plus sombres et m'a finalement permis d'atteindre la lumière au bout du tunnel. Cette thèse leur est dédiée à tous)*.

CONTRIBUTION TO ORIGINAL KNOWLEDGE

This dissertation represents a significant milestone in research, making a substantial contribution to the field by uncovering the complex relationship between feeding ecology and the accumulation of persistent organic pollutants in killer whales across the North Atlantic Ocean. By employing cutting-edge methods and introducing novel approaches to analyze killer whale diets, this thesis stands as a remarkable achievement, showcasing innovative research and enhancing our understanding of these iconic marine predators.

Chapter Three of the thesis highlights the significance of considering interindividual variation in prey specialization when examining wildlife feeding ecology and its relationship to the accumulation of persistent organic pollutants. This aspect has been frequently overlooked in ecotoxicological research. The study is the first to report persistent organic pollutant concentration for killer whales in Iceland. I reveal widely varying polychlorinated biphenyl concentrations within the population, which are linked to the dietary preferences of each individual. Killer whales feeding on marine mammals exceed established thresholds for risks of health effects. Recognizing the ecological differences between individuals is crucial to accurately assess the threats posed by contaminants to the long-term survival of this population. By uncovering these relationships and emphasizing the importance of individual specialization in exposure to toxic contaminants, this work also highlights a knowledge gap for other populations, and the need for more research into the ecology of individuals.

Chapter Four introduces a quantitative fatty acid analysis method for estimating the diets of killer whales. Accurate diet estimates are crucial for assessing trophic interactions and understanding food web dynamics, especially for apex predators like cetaceans, which have the potential to influence entire ecosystems through cascading effects. By calculating calibration coefficients that account for the fatty acid metabolism in different layers of blubber using

managed-care killer whales, I validated the method through cross-validation simulations and applied it to subsistence-harvested killer whales from Greenland, focusing on the outer blubber layer. This innovative method, the first developed for cetaceans, addresses limitations of previous diet estimation techniques, and provides a valuable tool for studying the feeding ecology of cetaceans. It holds great promise for understanding the diet dynamics of free-ranging toothed whales and other cetacean species across the world's oceans.

Chapter Five makes a significant contribution to the field of feeding ecology by using our newly validated quantitative fatty acid signature analysis approach to wild killer whales across an ocean basin. In the first, and largest study of its kind, I examine the diet composition of killer whales across the North Atlantic, involving nearly 200 killer whales and over 900 potential prey species. The findings demonstrate regional variation in the diets of killer whales, with whales in different areas primarily consuming other whales, seals, or fish. Additionally, the study highlights substantial individual specialization within these regions. These results further emphasize the importance of considering individual dietary preferences in future ecological studies. This is the first time fatty acids are used to revel the mysterious feeding habits of Western North Atlantic killer whales. Our results will contribute to a better understanding of the impacts of killer whale predation on community and ecosystem dynamics in the changing marine environment.

Chapter Six makes a significant contribution to the field of ecotoxicology by providing unprecedented insights into the concentrations of persistent organic pollutants in killer whale populations across the North Atlantic, using the diet estimates generated in Chapter Five. My research reveals distinct differences in contaminant levels between Western and Eastern North Atlantic killer whales, with higher concentrations found in the Western locations. These variations are attributed to differences in feeding habits, not only across location but between

individuals within locations, highlighting the role of individual feeding specialization in contaminant exposure and associated health risks. My findings underscore the need for improved waste disposal practices, prevention of further contamination, and mitigation of emerging contaminants. By enhancing our understanding of persistent organic pollutant distribution and its implications for killer whale populations, this research promotes the development of effective strategies for protecting marine ecosystems and the health of these apex predators.

CONTRIBUTION OF AUTHORS

The thesis is structured as a collection of four data manuscripts (chapters Three, Four, Five & Six), with each manuscript adhering to the formatting guidelines of the respective journal in which it was published or submitted for publication. I am the sole first author of the four manuscripts.

For Chapter Three, I performed the contaminant analyses at the National Wildlife Research Center (NWRC) at Environment and Climate Change Canada (ECCC), analyzed the data and wrote the manuscript. Robert J. Letcher, Rune Dietz, Christian Sonne and Melissa A. McKinney designed the study. Filipa I.P. Samarra and Gísli Víkingsson collected the killer whale samples. Christian Sonne and Rune Dietz ensure the transfer of the samples from Iceland to Canada. David Blair trained me at NWRC and assisted me in the contaminant analyses. Jean-Pierre Desforges performed some of the data analysis. All authors discussed the results and implications and commented on the manuscript at all stages.

For Chapter Four, I developed the method, performed the data analyses, and wrote the manuscript with input from Melissa A. McKinney and Sara J. Iverson. Rune Dietz, Christian Sonne and Melissa A. McKinney designed the study. Rune Dietz, Christian Sonne and Aqqalu Rosing-Asvid provided the killer whale and fish samples from Greenland. Melissa A. McKinney oversaw the fatty acids analysis for all samples. Haley Land-Miller and Adam F. Pedersen analyzed fatty acids for Greenland narwhal and minke whale. Denis Roy calculated the calibration coefficients. All authors reviewed and edited subsequent versions of the manuscript.

For Chapter Five, I co-designed the study with Melissa A. McKinney, Rune Dietz, and Christian Sonne, with input from all co-authors. I performed the fatty acid analyses and data analysis and wrote the original draft of the manuscript with input from Melissa A. McKinney. Filipa I. P.

Samarra, Audun H. Rikardsen, Aqqalu Rosing-Asvid, Lisa E. Kettemer, Steven H. Ferguson, Cortney A. Watt, Cory J. D. Matthews; Rune Dietz, Christian Sonne, and Jeremy J. Kiszka provided the killer whale samples/data. Filipa I. P. Samarra, Sandra M. Granquist, Eve Jourdain, Katrine Borgå, and Anders Ruus provided the Icelandic and Norwegian prey samples. All authors reviewed and edited subsequent versions of the manuscript.

For Chapter Six, I co-designed the study with Melissa A. McKinney, Rune Dietz, and Christian Sonne, with input from all co-authors. I, as well as Melissa A. McKinney, Robert J. Letcher, Cory J.D. Matthews, Cortney A. Watt, Steve H. Ferguson performed or supervised the contaminant analyses. I performed the data analysis and wrote the original draft of the manuscript with input from Melissa A. McKinney. Filipa I.P. Samarra, Audun H. Rikardsen, Aqqalu Rosing-Asvid, Steven H. Ferguson, Cortney A. Watt, Cory J. D. Matthews; Rune Dietz, Christian Sonne, and Jeremy J. Kiszka provided the killer whale samples/data. All authors reviewed and edited subsequent versions of the manuscript.

1 CHAPTER ONE: GENERAL INTRODUCTION

Our world relies on a delicate balance between its systems, and billions of people rely on the oceans to live and prosper. According to the United Nations, the sustainability of our oceans is under severe threat. As long-lived cosmopolitan species, marine mammals are the sentinels of our oceans, and allow us to understand how anthropogenic effects impact the ecosystems on which we rely. Top predator marine mammals are also essential to the oceans' ecological balance, and can exert top-down effects on entire food webs (Springer et al., 2003). Today, marine mammals face various cumulative threats, including prey availability due to climate change and chemical pollution (Simmonds, 2018). As such, it is necessary to understand the threats marine mammals face to improve their conservation and allow human populations, who rely on the same ecosystems, to prosper.

As the oceans' top marine mammal predators, killer whales (*Orcinus orca*) accumulate high concentrations of anthropogenic contaminants (R. Dietz et al., 2019; Jepson et al., 2016). Some of these chemicals called persistent organic pollutants (POPs) were massproduced in the twentieth century because of their flame retarding properties and their potency as pesticides. A few POPs, like polychlorinated biphenyls (PCBs), were banned under the Stockholm Convention in 2004 because of their toxicity to humans and wildlife, while others came to replace them, like emerging flame retardants. The main concern with POPs is their persistence in the environment, high immune and reproductive toxicity, and tendency to bind to lipids, causing them to concentrate as they move up the food webs, even decades after their ban (R. Dietz et al., 2019). Consequently, killer whales are the most contaminated animals on the planet. A recent population modeling study suggested that half of killer whales' populations could disappear by 2100 because of their PCB concentrations (Desforges et al., 2018). But not all killer whales are

equally threatened. The ones that feed on marine mammals, like seals and toothed whales, are far more likely to accumulate toxic amounts of POPs (Andvik et al., 2020; Remili et al., 2021). Conversely, killer whales that rely on fish usually have POP levels under known thresholds for adverse health effects (Krahn et al., 2007). These dietary differences sometimes occur in the same population, with individuals specializing in different prey (Jourdain et al., 2020; Samarra et al., 2017c). Thus, taking individual feeding variation into account is critical to understanding killer whales' ecology and exposure to legacy and emerging POPs (Andvik et al., 2020; Remili et al., 2021). Before I embarked on my doctoral research, there was a significant gap in knowledge regarding killer whale populations in the Arctic and sub-arctic regions of the North Atlantic (NA), particularly in the Canadian Arctic.

Studying North Atlantic killer whale diets is essential to improve this charismatic top predator's conservation efforts. The study predicting killer whale population collapses by 2100 was based on a low sample size in the NA (Desforges et al., 2018). My thesis provides critical information on how diets influence NA killer whales' POPs accumulation, identify at-risk groups, and will empower decision-makers with data to support continued and improved POPs management efforts. Additionally, the QFASA approach will help investigate cetaceans' diets in-depth enabling further research on their dietary shifts caused by climate change. Our results may also explain how killer whales may impact prey availability for other charismatic and threatened predators like polar bears and indigenous communities relying on subsistence harvesting.

Ultimately, my main objective is to understand how inter-population and intra-population differences in feeding ecology impact risks related to POP exposures in these NA killer whales. To address this problem, I used multiple high-resolution chemical tracers in samples collected within similar time frames and across all NA regions to improve our understanding of killer whale feeding ecology. I gathered samples and/or data for the project (n=191) collected by our

international partners between 2010 and 2022. Local indigenous collaborators took samples (biopsies or subsistence harvests) from Pond Inlet, and Pangnirtung in Canada, and from Tasiilaq and Nuuk in Greenland. Two Faroese samples came from stranded individuals. The other samples were remote biopsied from small boats in Norway, Iceland, Newfoundland, and Saint-Pierre and Miquelon.

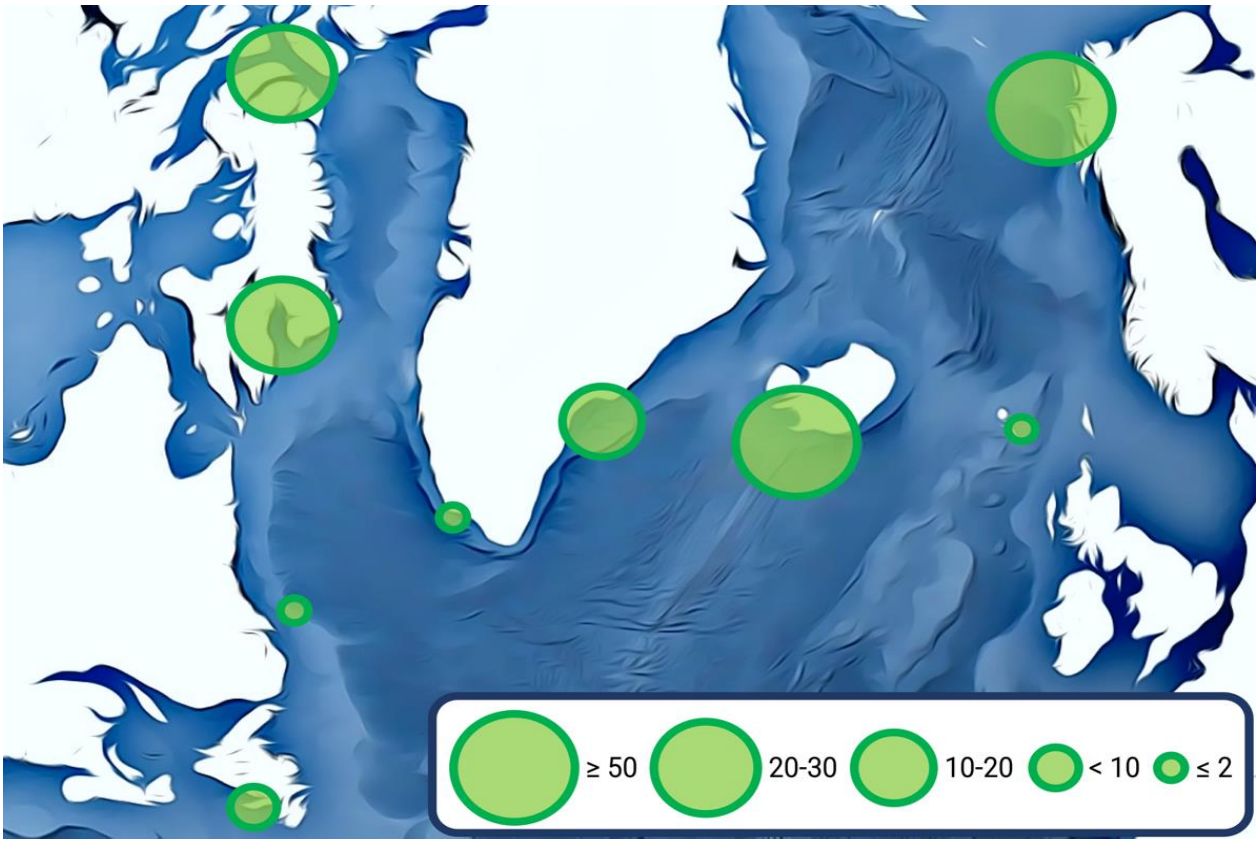

Figure 1-1:Study area covered by this doctoral thesis. Each circle represents an area and approximate number of biopsies collected for killer whales.

2 CHAPTER TWO: REVIEW OF THE RELEVANT LITERATURE

This chapter presents a comprehensive literature review addressing key aspects related to the feeding ecology and risks associated with persistent organic pollutant accumulation in North Atlantic killer whales. We examine the concept of persistent organic pollutants, their adverse effects on marine mammals, and the primary factors driving their accumulation in these animals. Additionally, we explore the pivotal role of killer whales as apex predators in the ocean and the significance of studying their diets. Specifically, we delve into the feeding ecology of North Atlantic killer whales, including intra- and inter-population variations. Through synthesizing the existing knowledge in these domains, this literature review aims to establish a comprehensive foundation for further analysis and a deeper understanding of the intricate dynamics among killer whales, their dietary habits, and persistent organic pollutant accumulations and associated health risks.

2.1 PERSISTENT ORGANIC POLLUTANTS

With at least 350 000 chemicals and mixtures of chemicals registered for use in the world, chemical pollution has been identified as one of the nine key global threats to wildlife, as well as human health, in the Anthropocene era (Steffen et al., 2011; Steffen et al., 2015; Wang et al., 2020). One major group of contaminants, known as persistent organic pollutants POPs, have in common their environmental persistence, long-range transport, ability to bioaccumulate and biomagnify through the food web, and toxicity to humans and biota. Once POPs are emitted, they are resistant to breakdown reactions and can travel long distances by atmospheric, oceanic and/or fluvial transport before being re-deposited in areas far from where they were released

(Kelly et al., 2007; Wania et al., 1996). Following deposition in the environment, POPs can enter biota through inhalation and dermal absorption, although the diet is the main route of entry of most POPs for consumers (Mackay, 1989; Mackay et al., 2000). Depending on their chemical structure, POPs can accumulate in the body and bind to lipids or proteins and biomagnify through the food web (Wania et al., 1996). In 2001, under a United Nations treaty known as the Stockholm Convention, nearly all countries agreed to reduce or eliminate the production, use, and/or release of twelve key POPs, known as "the dirty dozen", or 'legacy' POPs (*The Stockholm Convention on Persistent Organic Pollutants*, 2001). After 2004, a number of new chemicals, known as 'new and emerging' POPs, were added to the convention and others are being evaluated for possible inclusion (Table 2-1). Long-banned POPs still linger in the environment and other chemicals that may have POPs-like properties are still in use and are undergoing primary emissions (Muir et al., 2006).

The Stockholm Convention on POPs encourages parties to develop and implement national action plans for the safe disposal of POPs. These plans encompass various measures, including the development of inventories to identify the sources, quantities, and locations of POPs, enabling a better understanding of the issue, and facilitating prioritization of disposal actions. Parties are also urged to establish appropriate disposal facilities that consider the safe handling, storage, and destruction of POPs (*The Stockholm Convention on Persistent Organic Pollutants*, 2001). The convention promotes the adoption and promotion of environmentally sound technologies, such as high-temperature incineration and chemical destruction, to effectively eliminate or reduce the concentration of POPs in waste streams. Moreover, the Stockholm Convention emphasizes the significance of international cooperation in supporting the safe disposal of POPs. This involves sharing best practices, technical expertise, and financial resources to assist developing countries in establishing and operating disposal facilities. Finally,

the convention highlights the importance of minimizing unintentional releases of POPs through measures like reducing hazardous waste generation, promoting cleaner production methods, and phasing out or substituting POPs with safer alternatives. The Stockholm Convention requires Parties to phase out the use of polychlorinated biphenyls (PCBs) in equipment by 2025 and ensure elimination of PCBs by 2028 (five years from the submission date of this thesis).

Table 2-1: Persistent organic pollutants (POPs) added to the Stockholm convention from its creation in 2001 until now.

Legacy POPs "The Dirty Dozen"	Type of chemical	Year added
Aldrin	Pesticide	2001 (Effective 2004)
Chlordane (CHLs)	Pesticide	2001 (Effective 2004)
Dichlorodiphenyltrichloroethane (DDTs)	Pesticide	2001 (Effective 2004)
Dieldrin	Pesticide	2001 (Effective 2004)
Endrin	Pesticide	2001 (Effective 2004)
Heptachlor	Pesticide	2001 (Effective 2004)
Hexachlorobenzene (HCB)	Industrial chemical, By-product	2001 (Effective 2004)
Mirex	Pesticide	2001 (Effective 2004)
Polychlorinated biphenyls (PCB)	Industrial chemical, By-product	2001 (Effective 2004)
Polychlorinated dibenzo-p-dioxins (PCDD)	By-product	2001 (Effective 2004)
Polychlorinated dibenzofurans (PCDF)	By-product	2001 (Effective 2004)

Toxaphene	Pesticide	2001 (Effective 2004)
New & Emerging POPs	**Type of chemical**	**Year added**
Hexachlorocyclohexane (HCHs)	Pesticide	2009
Chlordecone	Pesticide	2009
Hexabromobiphenyl	Industrial chemical	2009
Hexa-brominated diphenyl ethers (BDE), HeptaBDE, OctaBDE	Industrial chemical	2009
Pentachlorobenzene	Industrial chemical, By-product, Pesticide	2009
Perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride (PFOSF)	Industrial chemical	2009
TetraBDE, Penta BDE	Industrial chemical	2009
Technical endosulfan and its related isomers	Pesticide	2011
Hexabromocyclododecane (HBCD)	Industrial chemical	2013
Hexachlorobutadiene	Industrial chemical, By-product	2015
Pentachlorophenol and its salts and esters	Pesticide	2015
Polychlorinated naphthalenes	Industrial chemical, By-product	2015
DecaBDE (commercial mixture, c-decaBDE)	Industrial chemical	2017
Short-chain chlorinated paraffins (SCCPs)	Industrial chemical	2017

Dicofol	Pesticide	2019
Perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds	Industrial chemical	2019
Perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds	Industrial chemical	2022

Under review as of 2023: Dechlorane Plus, Methoxychlor, Chlorpyrifos, chlorinated paraffins, long chain-perfluorocarboxylic acids, their salts and related compounds, and UV-328.

2.2 EFFECTS OF POPS ON MARINE MAMMALS

POPs can be found virtually everywhere on our planet in measurable concentrations (Ross et al., 2000b), but it is concentrations in top trophic level wildlife that are often of particular concern (Sonne et al., 2018). Marine mammals, particularly cetaceans, are one of the animal groups that accumulate high concentrations of several POPs (Jepson et al., 2016), in part because of their longevity (Desforges et al., 2016) and their thick layer of subcutaneous blubber in which these pollutants preferentially accumulate (Reijnders et al., 2009). However, the diet is the main driver of marine mammals' contamination, as they accumulate biomagnifying compounds through their top predator position in the food web (Herman et al., 2005; Kelly et al., 2007).

High levels of contaminants like PCBs, organochlorine pesticides (OCs) and flame retardants (FRs) in marine mammals have been associated with health issues that include increased risks of altered immune, endocrine, and reproductive functions, as well as carcinogenicity (Desforges et al., 2016; Mos et al., 2006). PCBs and brominated flame retardants (BFRs) have been known to interfere with the proper functioning of the immune system by

disrupting the activity of natural killer cells and T lymphocyte cells, known to play a role in the innate and adaptive immune systems respectively, making the body more susceptible to infections (Desforges et al., 2017; Ross et al., 1996). PCBs, BFRs and OCs are also endocrine disruptors, having been shown to interfere with thyroid function and vitamin A homeostasis (Boas et al., 2006; Letcher et al., 2010; Schwacke et al., 2012; Wells et al., 2005). These pollutants may affect the functioning of the reproductive system for both male and female individuals (Buckman et al., 2011), thereby reducing reproductive success (Jepson et al., 2016; Sonne et al., 2015). They have also been linked to elevated rates of gastrointestinal cancers (Martineau et al., 1994). Toxicity effects for PCBs are estimated to occur past the 10 mg/kg lipid weight (lw) threshold in marine mammals (Kannan et al., 2000). Many populations of marine mammals for which PCB levels were measured had concentrations above this threshold, rending them vulnerable to health adverse effects. Thus, there is a need to assess both the levels, toxicity, and risks caused by POPs for all marine mammal populations around the globe.

2.3 DRIVERS OF POPS IN MARINE MAMMALS

Understanding the major drivers of POP uptake and accumulation in marine mammals is essential for comprehending their exposure pathways, distribution patterns, and potential impacts on individual animals and populations. Longevity is an important driver for contaminant accumulation since marine mammals have relatively extended lifespans. As a result, these animals experience the bioaccumulation of lipophilic POPs in their tissues over time (Borgå et al., 2004; Kelly et al., 2008; Letcher et al., 2009). Older individuals generally exhibit higher POP burdens due to cumulative exposure, reduced elimination rates, and increased feeding activity throughout their lifetimes. Additionally, sex-based disparities in the accumulation of persistent organic pollutants (POPs) have been documented in numerous marine mammal species (Beck et

al., 2005; Krahn et al., 2009; Lawson et al., 2020; McKinney et al., 2010; Remili et al., 2020). Typically, females demonstrate lower levels of POPs in comparison to their male counterparts. This discrepancy can be attributed to the transmission of POPs during reproductive processes (Andvik et al., 2021; Lee et al., 2023; Tanabe et al., 1982). Throughout gestation and lactation, lipophilic substances stored within maternal tissues, particularly blubber, have the potential to mobilize and transfer to offspring, thereby engendering augmented contaminant burdens in female marine mammals (Wells et al., 2005).

Marine mammals exhibit species-specific variations in their biotransformation capacity, which influences the metabolism and elimination of POPs. POPs can be metabolized by phase I (CYP450) and phase II biotransformation enzymes, making them more excretable, but with a low efficiency in marine mammals (Houde et al., 2005; McKinney et al., 2006; Meyer et al., 2018). Cetaceans are also known to be deficient with respect to the Paraoxonase 1 (PON1) gene (Meyer et al., 2018). This PON1 enzyme plays a critical role in detoxification of Phase I products in other carnivorous species (Meyer et al., 2018), but is inactive in cetaceans, thus reducing their detoxification abilities. Ultimately, the most effective way for cetaceans to get rid of their POP burdens is through maternal transfer of pollutants to the offspring (Jeong et al., 2018).

Finally, dietary habits and trophic position play a vital role in marine mammals' exposure to POPs (Corsolini et al., 2017; Kelly et al., 2008; Letcher et al., 2010; Won et al., 2018). When these pollutants are discharged into rivers, lakes, or oceans, they are often taken up by small organisms like plankton or algae, which form the base of the marine food chain (Desforges et al., 2014; Frouin et al., 2013; Sobek et al., 2010). As larger organisms feed on these smaller ones, the pollutants they contain are transferred to their bodies through biomagnification (Bengtson Nash et al., 2018; Das et al., 2017; Remili et al., 2020). Biomagnification refers to the increasing

concentration of certain chemicals or pollutants as they move up the food chain (Borgå et al., 2004). This transfer of pollutants continues as they progress up to higher trophic levels. As predators, marine mammals consume large quantities of fish or other marine organisms that have already accumulated pollutants in their bodies (Hickie et al., 2007). Because of biomagnification, the concentration of POPs increases at each level of the food chain (Cullon et al., 2012; Letcher et al., 2009; Mackay et al., 2000; Sørmo et al., 2006). And because POPs (at least PCBs, OCs, and BFRs) are lipophilic, they have a higher affinity for fats and oils (Ewald et al., 1998). As a result, they have a greater tendency to accumulate in the adipose tissue (blubber) of marine mammals, which is their primary fat storage site.

2.4 KILLER WHALES ARE THE OCEANS' APEX PREDATORS

Killer whales, also known as orcas, are the largest species of the dolphins' family (Delphinidae). They have a cosmopolitan distribution and are most commonly encountered in coastal, temperate waters (Ford, 2009). They are easily recognizable due to their striking black and white markings and can grow up to 9 meters in males and 7 meters in females (Ford, 2009). With a mean life expectancy of approximately 50 years and a maximum longevity of 80-90 years, females have a longer lifespan than males, which attain sexual maturity at around 15 years and have a mean life expectancy of about 30 years, with maximum longevity of about 50-60 years (Foote, 2008; Foster et al., 2012; Nielsen et al., 2021). Sexual dimorphism is evident in the anatomical characteristics of male and female killer whales. One notable distinction lies in the dorsal fin morphology. Adult male killer whales, or bulls, exhibit robust and tall dorsal fins, reaching heights of up to 1.8 meters (Ford, 2009). In contrast, female killer whales, or cows, possess relatively smaller dorsal fins, typically measuring around 0.7 to 0.9 meters in height. The size disparity in dorsal fins allows the visual identification of sex and sometimes age class.

Young males' dorsal fins typically look like females until the male reaches sexual maturity. When going through the late stages of puberty, males' dorsal fins sprout and become tall and straight. On the ventral side, differences are observed in the presence of mammary slits and the genital slit. Female killer whales possess mammary slits, which are openings located near the base of the mammary glands. These slits facilitate nursing and the provision of milk to their offspring (Ford, 2009).

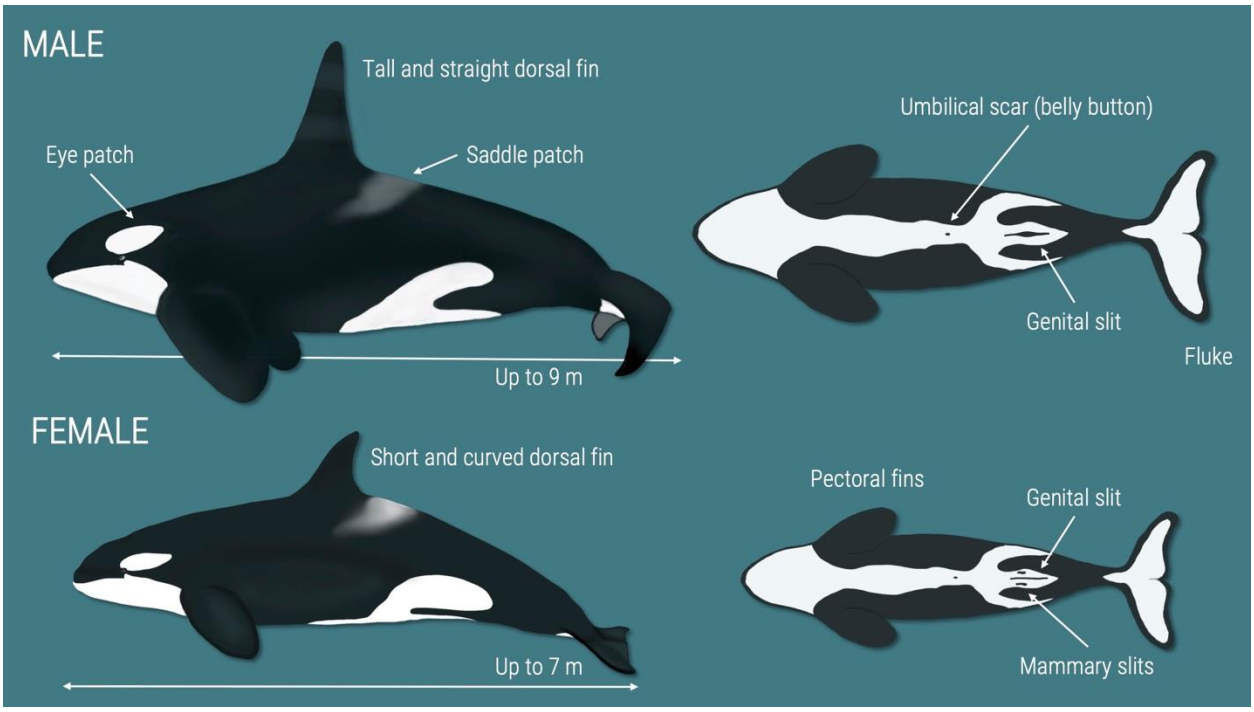

Figure 2-1: Anatomy differences between a typical adult male killer whale and a typical adult female killer whale (Illustrations by Anaïs Remili)

Killer whales are considered a charismatic species thanks to their intelligence, and distinct appearance that captures the public's attention. Additionally, they have been featured in popular media, including movies and documentaries, which has increased public interest in the species (Wearing et al., 2011). By showcasing their captivating behaviors and emphasizing their need for natural habitats, these films have helped generate empathy and concern among the

general public, thereby mobilizing support for conservation efforts (Burford et al., 2017). Tourist parks, especially those showcasing killer whale shows and educational programs, play a crucial role in conservation (Kelsey, 1994). They offer close encounters, fostering appreciation and connection with killer whales. Educational presentations and interactive experiences raise awareness about their biology and conservation challenges, garnering public support. However, due to concerns about animal welfare, marine parks' popularity has declined, with more focus on whale watching in natural habitats (Silk et al., 2018). This shift has prompted increased efforts to protect wild populations through monitoring, behavioral studies, and establishing protected areas (Jefferies et al., 2021). Public demand has pressured governments and international organizations to enact laws safeguarding killer whales and their habitats, evident in measures implemented by the United States and Canada to mitigate human activities' impact on the critically endangered Southern Resident population (Burnham et al., 2021; Pedersen, 2022).

As apex predators, killer whales play a vital role in the marine food web, regulating the populations of their prey and maintaining a healthy balance in the ocean ecosystem (Estes et al., 2016). They are known to hunt cooperatively, using sophisticated hunting techniques that involve coordination, communication, and strategic planning. For example, some killer whale populations are known to use "carousel feeding," where they work together to drive schools of fish into a tight ball, and then take turns feeding on them (Simila et al., 1996). Other feeding strategies may include stealth and utilizing rocky shores to sneak up on seals (Vongraven et al., 2014). Some groups of killer whales have been observed exhibiting more than one type of feeding strategies, known as “prey-switching”, which may occur seasonally or in response to declining rates of a specific prey (Samarra et al., 2015). This level of cooperation and communication is thought to be rare in the animal kingdom and highlights the complexity of killer whales’ feeding strategies.

Killer whale diets may be impacted by climate change, which can modify the migration of several fish stocks on which these whales prey (Fossheim et al., 2015). The changes in fish availability could disrupt the delicate equilibrium within the marine ecosystem and present challenges for killer whales, compelling them to adjust their hunting strategies, relocating North to follow the fish, or potentially explore alternative sources of prey (Nikolioudakis et al., 2019; Nøttestad et al., 2015; Olafsdottir et al., 2019). The lack of Arctic sea ice, which is diminishing each year and expected to disappear during the summer within the next decade, translates to an increase in killer whale presence in higher latitudes (Gascard et al., 2019). Indeed, while the large dorsal fin of killer whales presents navigational difficulties among sea ice, the absence of entrapment risks provides a new environment for these whales to explore. This presents an opportunity for killer whales to discover and diversify their diet by encountering new potential prey (Ferguson et al., 2010). There, experts suspect they now hunt Arctic seals or other Arctic marine mammals (Bourque et al., 2018; Ferguson et al., 2012b; Ferguson et al., 2010; Matthews et al., 2019). As a result, trophic cascades arising from changes in killer whale diets can have substantial ecological consequences (Estes et al., 2003; Estes et al., 1998). These effects can propagate both upwards and downwards, affecting other trophic levels and potentially leading to shifts in prey populations and ecosystem dynamics (Estes et al., 2016). Additionally, killer whales could affect local human populations, by competing for the same prey (Westdal et al., 2013). This competition for limited prey may have implications for the subsistence and cultural practices of indigenous communities, who have traditionally relied on these species for sustenance and cultural significance. Moreover, changes in prey availability may necessitate adaptations in hunting practices and the exploration of alternative food sources, further impacting the traditional livelihoods and food security of these communities (Ferguson et al., 2012a; Westdal et al., 2013; Young et al., 2019). Understanding these potential dietary shifts is

crucial for effective conservation and management strategies, and we need elucidate how they may affect the ecosystems in the North Atlantic. But first, we need to accurately estimate the whales' diets.

2.5 STUDYING KILLER WHALE DIETS

Studying killer whale diets can be approached through various methods, each providing valuable insights into their feeding habits. These methods include:

1. *Observation*: Direct observation of killer whales in their natural habitat allows researchers to visually identify and document their prey species. By observing hunting behaviors, prey selection, and feeding strategies, researchers can gain a comprehensive understanding of the diet composition of killer whales (Ford et al., 2006). Beyond observation, scientists may use tracking tools such as satellite tags to infer movements and association with prey stocks. Notably, recent research on the Norwegian killer whale population, exemplified by studies like, has revealed a correlation between the spatial utilization strategies of killer whales and their engagement with fishing activities, specifically in relation to herring density (Vogel et al., 2021).
2. *Stomach Contents or Fecal Remains*: Examining the stomach contents or fecal remains of killer whales provides valuable information about the specific prey items they have consumed. These can include fish otoliths or other hard tissue like bones, or body parts. Analyzing these samples thus allows researchers to identify prey species and quantify their relative abundance, shedding light on the diet preferences and feeding patterns of killer whales (Ryan et al., 2012).

3. *Metabarcoding in the Feces:* Metabarcoding involves analyzing the DNA present in fecal samples to identify the species consumed by killer whales. This method utilizes DNA sequencing techniques to identify prey DNA fragments in the feces, providing a comprehensive and detailed picture of the diet composition, including both known and unknown prey species (Hanson et al., 2021).
4. *Stable Isotopes*: Stable isotope analysis is based on the principle that the isotopic composition of an organism's tissues reflects the isotopic signatures of its diet. By analyzing stable isotopes ratios, such as carbon and nitrogen or sulfur for example, in bulk, in various tissues of killer whales, researchers can infer the trophic position of the prey species and their relative contribution to the overall diet (Newsome et al., 2010). Additionally, stable isotopes can be measured in specific amino acid or proteins to help fine tune the diet signals and remove the geographic baseline variation in C or N ratios (Matthews et al., 2020).
5. *Fatty Acids*: Fatty acid analysis involves studying the composition and ratios of fatty acids in the blubber of killer whales. Different prey species contain distinct fatty acid profiles, and by comparing these profiles with those found in the blubber of killer whales, researchers can distinguish between different feeding habits and evaluate dietary preferences (Herman et al., 2005).
6. *Bioaccumulating organic contaminants or trace element*s: Killer whales are apex predators, and as such, they are exposed to and accumulate biomagnifying organic contaminants and trace elements present in their prey species. By analyzing the levels of these substances (PCBs or mercury for example) in killer whale tissues, such as blubber or muscle samples, researchers can gain insights into the types and concentrations of contaminants present in their diet. This information can help identify the potential sources

of contamination in the marine environment and assess the overall health and ecological impacts on killer whale populations (Krahn et al., 2007).

These methods can be used individually or in combination to provide a comprehensive understanding of killer whale diets. Each approach offers unique advantages and limitations (reviewed in Table 2-2), and their combined use enhances the accuracy and reliability of the dietary assessments, contributing to our knowledge of killer whale ecology and their role in marine ecosystems. For example, assessing marine mammals' diets and related inter-and intra-population variation can be challenging, especially in isolated regions like the NA, where visual observation is limited. Visual observations of marine mammals foraging in the wild are infrequent, often seasonal, limited to surface events, and may not accurately reflect their long-term diet (Bowen et al., 2013). Thus, researchers usually have to rely on chemical tracers measured primarily from skin biopsies because they can reflect integrated diet signals over time (Krahn et al., 2007; Remili et al., 2020). Stable isotopes of carbon and nitrogen and qualitative fatty acid (FA) signature analyses can help determine a predator's feeding habits, given that enough individuals are sampled (Bourque et al., 2018; Foote et al., 2012).

We may analyze FA signatures quantitatively to estimate the diets of carnivore populations. FAs get integrated with minor and predictable modifications from the prey to the predator's fat (*e.g.*, blubber). To reconstruct their diets to the species level, scientists developed quantitative FA signature analysis (QFASA) in seals and polar bears (Iverson et al., 2004; McKinney et al., 2013). Using the statistical distance between the FA signature of potential prey species and that of the predator, this method calculates the proportion of various prey species in the predator's diet. However, this method was never successfully applied to cetacean skin biopsies due to the absence of cetacean-specific calibration coefficients (CCs) accounting for their lipid metabolism (Choy et al., 2019). CCs are computed for each FA as the ratio of the FA

proportion in the predator to the FA proportion in the prey (Iverson et al., 2004). Whale blubber is thick and highly stratified, thus preventing researchers from using other species' CCs in a cetacean model (Bourque et al., 2018). Yet, killer whales are a prime candidate for developing a QFASA method on cetaceans; they are top predators with a thick layer of fat, thus facilitating layer-specific analyses. Whilst feeding trials are challenging to implement, many captive individuals are kept in facilities worldwide, allowing access to blubber samples and diet items. Developing this method for killer whales and cetaceans depends on the calculation of cetacean-specific CCs. It would be an invaluable tool to help understand their feeding ecology on a fine scale.

Table 2-2: Strengths and limitations of methods used to estimate the diets of marine mammals (modified from Bowen and Iverson, 2013).

Method	Dietary history	Species composition	Prey size	Requirements	Strengths	Limitations
Feces, Hard parts	Last few meals	Yes	Yes	Reference collection of prey species and bones Otoliths size measurements Otoliths-prey size regression	Large sample size possible Non-lethal collection Inexpensive	Preys must have hard parts and they must be ingested Hard parts must resist digestion Correction factors to reduce bias caused by partial erosion and complete digestion must be estimated Correction factors not available for all prey species False positives and negatives possible May not be representative of species with long foraging trips Demographic traits of individuals unknown
Feces, metabarcoding	Last few meals	Yes	No	Access to sequencing and DNA extraction	Allows for a large range of potential prey	Does not allow for quantification of prey species in the diet.

						May be biased by cross-contamination
Stomachs, Hard parts	Last few meals	Yes	Yes	Reference collection of prey species and bones	Large sample size possible	Animals must be dead
				Otoliths size measurements	Non-lethal collection	Prey must have species-specific hard parts, and these must be ingested
				Otoliths-prey size regression	Inexpensive	Hard parts must resist digestion
						Correction factors to reduce bias, but these are not usually available
						False positives and negatives possible
						May not be representative of species with long foraging trips
						Often many empty stomachs
						Differential digestion may further bias results
Stable Isotopes	Days to years depending on tissue	No, But Exception For Simple Diet	No	Fractionation factors for tissues	Integrates diet over time	Trophic levels are relative to carbon source which must be measured
				Reference isotope levels from lower trophic levels	Used as independent check of trophic level	False positives and negatives possible
						Composition and size of prey not known
Fatty Acids	Days to months depending on species and life history	Yes	Some Coarse Resolution Possible	Distinguishable prey fatty acid signatures	Integrates diet over weeks-months	Detection level of rare prey still being evaluated
				Calibration coefficients (CC) to account for predator metabolism	Sampling location less likely to bias composition	False positives and negatives possible
				Prey fat content	Demographic traits of individuals known	Because of long integration time, location of foraging less well defined
				Predator adipose tissue		Only course resolution of prey size
						Estimates sensitive to CC and fatty acid set

Biomagnifying contaminants	Days to months	No	No	Contaminant levels in prey to calculate biomagnifying factor	Integrates diet over time Used as independent check of trophic level

2.6 FEEDING ECOLOGY AND ECOTOXICOLOGY OF NORTH ATLANTIC (NA) KILLER WHALES

Although killer whales' feeding ecology has been extensively studied in the North Pacific (Herman et al., 2005), much less is known about the NA killer whale's feeding habits. Killer whales in the NA are separated into different clusters of individuals (Forney et al., 2006; E. Jourdain et al., 2019). The major killer whale groupings include the Norwegian, Icelandic and Canadian Arctic groups (E. Jourdain et al., 2019). Genetic studies are still underway to characterize genetically distinct populations in the NA (Foote et al., 2019). In some remote, under-studied locations of the NA, population estimates are not available and the only information available is the minimum individual count (Table 2, Jourdain et al., 2019).

Killer whale populations in some regions, like in the North Pacific, have been classified into separate ecotypes (Ford et al., 1998). An ecotype describes a conspecific group of individuals with similar ecological adaptations regardless of genealogical relationship; that is, ecotypes are usually designated based on ecological, and not phylogenetic, criteria (Cronin et al., 2009). Each killer whale ecotype specializes on certain prey, with specific patterns of movement, behavior, and social system adaptations that are linked to this specialization (Ford et al., 1998; Whitehead et al., 2018; Yurk et al., 2015). Two NA ecotypes have been suggested in the past, based on limited, mostly observational data. Foote et al. (2009) defined the two potential ecotypes as

- Type 1: generalist killer whales are smaller, rely mostly on Atlantic herring (*Clupea harengus*) but also on some marine mammals, show heavy tooth wear, and;
- Type 2: specialist killer whales are larger, show almost no tooth wear and feed mostly on marine mammals.

At the time I am submitting this literature review, these two types have been retired by Foote, in a letter to *Marine Mammal Science* (Foote, 2022). His main argument was that type 2 was established based on five museum specimens, and that recent studies pointed toward a more complex ecology for North Atlantic killer whales, compared to a two-type separation.

Observational studies from the previous four decades have provided some insight into NA killer whale ecology. The data available for the three biggest killer whale groupings (e.g. Norway, Iceland and the Canadian Arctic + Greenland) are summarized in Table 3. Briefly, studies described the observed diet of killer whales in the NA and reported that while Norwegian and Icelandic whales appear the feed mainly on fish like herring (Foote et al., 2010; Samarra et al., 2017a; Sigurjónsson, 1988; Simila et al., 1993), Greenlandic and Canadian whales may focus feeding on marine mammals (Ferguson et al., 2012b; Ferguson et al., 2010; Higdon et al., 2012). However, isolated sightings have reported predation events on marine mammals for Norwegian and Icelandic whales (Samarra et al., 2015; Vongraven et al., 2014), and conversely, predation on fish for Canadian whales (Laidre et al., 2006; Westdal et al., 2013).

Visual observation may not provide individual-level diet information, especially in remote regions of the Western NA, which is required to quantitatively interpret intra- and inter-group variation in POPs concentrations (Annex Table 1). The use of chemical tracers has increased in recent decades because they allow for a long-term feeding ecology assessment (Bowen et al., 2013). Therefore, non-lethal, easily accessible chemical tracers analyzed in tissue

samples have been used to provide insight into feeding patterns of different groups of killer whales in the NA Ocean (Table 2-4). Briefly, tracers revealed that Icelandic and Norwegian killer whales seem to rely on fish (Foote et al., 2012; Samarra et al., 2017c; Wolkers et al., 2007) and Greenlandic and Canadian whales seem to rely to some extent on marine mammals (Bourque et al., 2018; Matthews et al., 2014; Pedro et al., 2017).

Table 2-3: Summary of published visual observations of North Atlantic killer whale feeding studies (up to 2019).

Main findings	Reference
Iceland	
Killer whales in Iceland are associated with herring stocks	Sigurjónsson et al., 1988
Some Icelandic killer whales move to Scotland and feed on marine mammals	Samarra & Foote, 2015
Some pods of killer whale follow the herring stocks along the west coast of Iceland	Samarra et al., 2017
Norway	
Norwegian killer whales feed on herring	Similä et al., 1996
Photo ID data suggests Norwegian and Icelandic groups do not overlap	Foote et al., 2010
Norwegian killer whale predation on seal	Vongraven et al., 2014
KI and K pods eat seal. Behavior similar to North Pacific transients	Jourdain et al., 2017
Greenland and Canadian Arctic	
Killer whale in the Canadian Arctic prey on marine mammals (baleen and toothed whales)	Ferguson et al., 2010; 2012
Killer whale in the Canadian Arctic may feed on narwhal, beluga and bowhead whale	Hidgon et al., 2012
Inuit reports confirm predation on marine mammals and were unable to confirm predation on fish	Westdal, 2013
Killer whale off West Greenland were sighted eating fish	Laidre et al., 2006

Table 2-4: Feeding ecology based on chemical tracers of the diet revealed a mixture of fish-feeding and marine mammal-feeding killer whales across the North Atlantic (up to 2019).

Tracer(s) used	n	Years	Main findings	Reference
Iceland				
Stable isotopes	64	2014-2016	Killer whale stable isotopes showed a herring-based diet. KW traveling to Scotland had higher $\delta^{15}N$	Samarra et al., 2017c
Stable isotopes	3	2012	Inconclusive on the diet of Icelandic killer whale. Values differed from Norwegian whales	Foote et al., 2012
Norway				

POPs	8	2002	Killer whales POP signatures reflect a diet based on herring but some individuals feeding on seals show high PCB concentrations	Wolkers et al., 2007 Andvik et al., 2020
Stable isotopes	17	2012	Stable isotope ratios consistent with herring diet, and with low variation among individuals but some individuals preying on seal have high $\delta^{15}N$ values	Foote et al., 2012 Jourdain et al., 2020
Greenland and Canadian Arctic				
POPs	18	2012-2014	POPs were elevated in Greenlandic killer whales suggesting a diet that includes marine mammals	Pedro et al., 2017
Fatty acids	18	2012-2014	Greenlandic killer whales showed fatty acid patterns that differed from killer whales known to feed on exclusively on fish	Bourque et al., 2018
Bulk and AA stable isotopes	13	1948–2011	No overlap of East Canadian Arctic and North West Atlantic niches. Diet associated with marine mammals to some extent.	Matthews & Ferguson, 2014

No study to date has assessed the feeding ecology variation between and within all North Atlantic killer whale groups. This statement holds until early 2023, before the release of the sixth chapter of this doctoral thesis. Studies using bulk stable isotopes have provided some dietary insights. However, the low variation between the samples and the overall small sample size did not help resolve killer whale feeding ecology across the NA (Foote et al., 2012). Additionally, the resolution of bulk stable isotopes studies has often been too low to be conclusive on intra-group dietary preferences (Foote et al., 2012). Studies using higher resolution tracers (fatty acids, POPs, and amino acid stable isotopes) have not been done together within a single region, nor within similar time frames to allow meaningful comparisons. In the eastern North Pacific, the combined use of POPs and fatty acids was key to delineating three distinct ecotypes (residents, transients and offshores) (Herman et al., 2005; Krahn et al., 2007; Krahn et al., 2008). However, to my knowledge, there is insufficient understanding of feeding variation within and between killer whale groups in the NA to confirm or refute specific killer whale ecotypes in this ocean.

2.7 INTRA- VS. INTER-POPULATION VARIATION

Traditional ecological studies typically disregard intra-specific variation and focus instead on means and total population measures, assuming individuals use the same resources (Bolnick et al., 2011; Bolnick et al., 2003). However, studies focusing on intra-specific feeding ecology have demonstrated the importance of individual traits in the feeding ecology of a population (Bolnick et al., 2003; Estes et al., 2003; Kernaléguen et al., 2015). Individual specialization occurs when an individual's niche is narrower than its population's niche (Bolnick et al., 2003). Although individual specialization may be explained by biological factors (age and sex), it may also be driven by factors beyond demographic variation, such as interspecific competition, intraspecific competition, ecological opportunity, and predation (Araujo et al., 2011; Bolnick et al., 2003). Thus, an observed generalist population can either be made up of individual generalists or a mixture of specialized individuals (Bolnick et al., 2003). Conspecific groups exploiting different resources can have several positive effects on the population's overall fitness and survival (Tixier et al., 2017) due to niche complementarity, which is the tendency for phenotypically divergent individuals to compete less strongly (Araujo et al., 2011; Bolnick et al., 2011). Individual specialization impacted POPs accumulation in Norwegian killer whale in a recent study where seal-feeding whales were at risk from their PCB levels, contrary to fish-feeding whales (Andvik et al., 2020).

There is a need to use higher-resolution chemical tracers (fatty acids, or POPs) in samples collected within similar time frames and across regions to improve our understanding of killer whale feeding in the NA Ocean (E. Jourdain et al., 2019). Inter-population and intra-population differences in feeding ecology may result in differential risks related to climate-driven changes in prey availability and effects related to exposures to environmental contaminants (Andvik et al., 2020; Pedro et al., 2017). Thus, improved conservation strategies should include renewed

efforts to resolve the question of the feeding ecology and differential POPs exposures of NA killer whales. Additionally, the development of novel tools to accurately predict the diets of killer whales could be use in future research assessing the potential impact of killer whale predation on Northern ecosystems and its implications for indigenous communities.

CONNECTING PARAGRAPH

In the following chapter, we shed light on the often overlooked but critical aspect of inter-individual variation in prey specialization within killer whale feeding ecology. We focus on Icelandic killer whales and investigate the levels of persistent organic pollutants, particularly PCBs, in their blubber. Our findings reveal remarkable differences in PCB concentrations among individuals. Notably, individuals with a mixed diet that included marine mammals have mean PCB concentrations six to nine times higher compared to individuals specializing in fish. These results are of great concern, as PCBs have been identified as potential threats to killer whale population growth, particularly when levels in mixed feeders surpass established thresholds. It is therefore imperative to acknowledge the ecological diversity among individuals to accurately assess the risks that contaminants pose to the long-term persistence of these magnificent marine predators. This chapter was published in *Environmental Science and Technology*, a very well-respected ecotoxicology journal. This chapter was a collaboration between nine coauthors (including myself).

3 CHAPTER THREE: INDIVIDUAL PREY SPECIALIZATION DRIVES PCBS IN ICELANDIC KILLER WHALES

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3.1 ABSTRACT

Inter-individual variation in prey specialization is an essential yet overlooked aspect of wildlife feeding ecology, especially as it relates to intra-population variation in exposure to toxic contaminants. Here, we assessed blubber concentrations of an extensive suite of persistent organic pollutants in Icelandic killer whales (*Orcinus orca*). Polychlorinated biphenyl (PCB) concentrations in blubber were >300-fold higher in the most contaminated individual relative to the least contaminated, ranging from 1.3 to 428.6 $mg.kg^{-1}$ lw. Mean PCB concentrations were six-to-nine-fold greater in individuals with a mixed diet including marine mammals than in fish specialist individuals, whereas males showed PCB concentrations four-fold higher than females. Given PCBs have been identified as potentially impacting killer whale population growth, and levels in mixed feeders specifically exceeded known thresholds, the ecology of individuals must be recognized to accurately forecast how contaminants may threaten the long-term persistence of the world's ultimate marine predator.

3.2 TABLE OF CONTENT ART

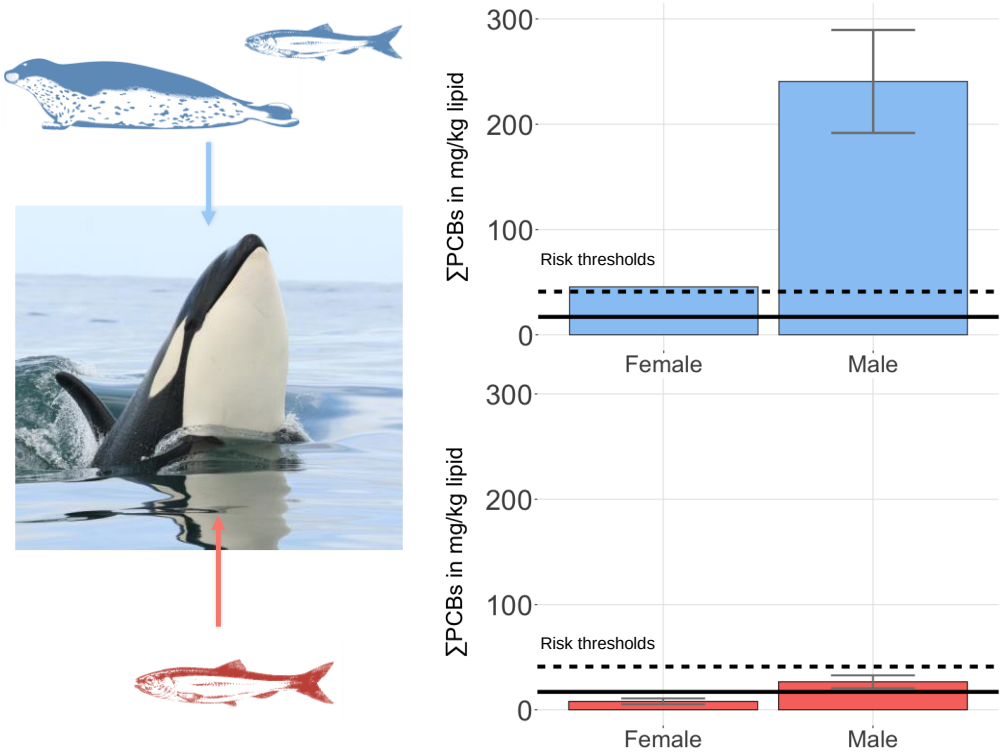

Figure 3-1: Graphical abstract

Keywords: POPs, stable isotopes, diet, intra-population prey specialization, trophic position, contaminants, risk assessment

3.3 INTRODUCTION

Environmental contaminants of toxicological concern such as persistent organic pollutants (POPs), and polychlorinated biphenyls (PCBs) in particular, biomagnify within food webs, making feeding ecology an important aspect of understanding contaminant accumulation in wildlife (Borgå et al., 2012). Yet, most feeding ecology and wildlife contaminant assessments have focussed on populations mean diets or contaminant concentrations, with limited consideration of individual variation in foraging behavior, also referred to as individual

specialization (Bolnick et al., 2003; Desforges et al., 2018). Although individual specialization may, in part, be explained by biological factors such as age and sex, it may also be driven by factors that go beyond demographic variation, including inter-individual and population variations in patterns of resource competition, predation and ecological opportunities (Bolnick et al., 2003). Studies examining variation in diet among individuals demonstrated the importance of individual traits in describing feeding ecology and associated individual specialization with better overall fitness and survival in marine mammal populations (Bolnick et al., 2003; Estes et al., 2003; Kernaléguen et al., 2015). With dietary absorption as a main route for contaminant accumulation, individuality in diet is expected to lead to variations in pollutant exposure and associated health risks, both within and among populations (Andvik et al., 2020; Borgå et al., 2012).

As a generalist apex predator with a tendency to adopt prey specializations at the individual or population level, killer whales (*Orcinus orca*) may provide critical insights into how feeding ecology may influence/drive contaminant accumulation (Ford, 2009; Herman et al., 2005; Krahn et al., 2007; Matthews et al., 2010). In well-studied regions, such as the eastern North Pacific, substantial differences in feeding ecology among killer whale populations have led to their classification into different ecotypes based largely on prey specialization (*i.e.* fish feeders vs. marine mammal feeders) (Herman et al., 2005; Krahn et al., 2007). However, far less is known about the foraging habits of killer whales in the North Atlantic (E. Jourdain et al., 2019). In the North Atlantic, killer whales have been tentatively identified as generalist and specialist feeding ecotypes (Foote et al., 2009). The supporting evidence suggests that Greenlandic and Canadian whales seem to rely mainly on marine mammals (Bourque et al., 2018; Matthews et al., 2014; Pedro et al., 2017). In contrast, whales in Norway and Iceland seem to vary in their intake of fish and marine mammals, between a diet composed predominantly of

fish on one hand to a diet including marine mammal prey to an unknown extent on the other hand (Foote et al., 2012). Nevertheless, new stable isotope analyses pointed to the possibility of individual specialization within the Icelandic and Norwegian populations (Jourdain et al., 2020; Samarra et al., 2017c).

Killer whales are among the most contaminated animals on the planet, and their exposure to high levels of contaminants like POPs has been thought to contribute to reduced reproductive success and population growth (Desforges et al., 2018; Rune Dietz et al., 2019; Jepson et al., 2016). For marine mammals specifically, POP levels have been linked to altered immune function, reduced reproductive success, endocrine disruption, and carcinogenicity (Rune Dietz et al., 2019; Fossi et al., 2018). Modeling studies have suggested that PCB contamination alone could contribute to reduced population growth in highly exposed populations of killer whales worldwide (Desforges et al., 2018; Hall et al., 2018; Hickie et al., 2007) (but see (Witting, 2019)). While the relationship between killer whale POP levels and diets is not well known across the North Atlantic, Greenlandic killer whales evaluated in recent years showed high POP concentrations that aligned with fatty acid signatures supportive of marine mammals as dietary components (Bourque et al., 2018; Desforges et al., 2018; Rune Dietz et al., 2019; Pedro et al., 2017). A recent study highlighting the role of intra-population variation in diets on contaminant accumulation and PCB patterns in Norway showed that seal-feeding killer whales were four times more contaminated than fish-feeding killer whales with PCB profiles dominated by higher chlorinated compounds (Andvik et al., 2020; Jourdain et al., 2020). This study demonstrated the need to account for intra-population fine-scale variations in feeding habits when quantifying contaminant accumulation in North Atlantic killer whale populations.

A population of North Atlantic killer whales regularly frequents Icelandic coastal waters where they seasonally congregate at wintering and spawning grounds of their assumed primary

prey, Atlantic herring (*Clupea harengus*) (Samarra et al., 2017a; Samarra et al., 2017c). This population has been reported to prey on fish, cephalopods, seabirds, and marine mammals (Samarra et al., 2018). Long-distance photographic matches have also noted several individuals traveling to Scotland to feed on high trophic prey, including seals (Samarra et al., 2015; Samarra et al., 2017c). Here we analyzed blubber samples from fifty Icelandic killer whales to quantify within-population variation in blubber concentrations of major POP groups, to identify how POPs vary with individual foraging specialization, and to assess how associated risks of health effects may vary with individual foraging specializations.

3.4 MATERIAL AND METHODS

3.4.1 Sampling

Sixty-four biopsies were collected opportunistically from 50 killer whales (35 males, 13 females, and 2 juveniles) in 2014 (n = 45 individuals) and 2016 (n = 5 individuals) in western and southern Iceland waters, where they are frequently seen feeding on herring (Table S3-2) (Samarra et al., 2017a; Samarra et al., 2017c). Biopsies comprising skin and blubber were collected using an ARTS pneumatic darting system (LKARTS-Norway, Norway) and stainless steel 25 x 7 mm (CetaDart, Denmark) biopsy tips. Biopsy tips were sterilized before use and stored in clean plastic bags. Samples were generally collected from the body's mid-lateral region, below the dorsal fin, and stored frozen in the field at -20 °C in aluminum foil. Once back at the lab, samples were stored at -80 °C until analysis. The shipment was conducted in Styrofoam boxes with dry ice until arrival in the lab at Carleton University, Ottawa. All sampled killer whales were photographically identified (Samarra et al., 2017b) to minimize the risk of re-sampling the same individuals within a single field season. Sampled individuals were sexed based on genetic analysis for whales sampled in 2014. Because genetic analyses were not

completed for the 2016 samples, sex was assigned based on morphological characteristics and sighting history, which were further relevant to determine individuals' age class (Tavares et al., 2017; Tavares et al., 2018). Age class was defined based on morphological characteristics and divided into three categories (as per Tavares et al. 2017) as follows: 1) adults were defined differently for males and females; adult males were considered individuals that have reached sexual maturity and presented a distinguishably taller dorsal fin, including individuals whose dorsal fin has started its growth spurt but is not fully grown yet; in the case of females, these were defined as mature-sized individuals, with a relatively smaller dorsal fin than adult males, seen consistently with a calf in echelon position, or without developing dorsal fin for at least three years; 2) Large juveniles – unknown sex or known males (genetically sexed) which have dorsal fins of the same apparent size as adult females but whose dorsal fin does not appear to have started its growth spurt; 3) juveniles, smaller sized individuals that have not reached mature size for which sex is unknown. No calves or young juveniles (≤ 3 years age) were sampled.

3.4.2 Contaminant analyses

Analytes monitored were as follows: 62 individually eluting or co-eluting PCB congeners; 20 individual organochlorines (OCs); 25 individual or co-eluting polybrominated diphenyl ethers (PBDEs); and 23 other non-PBDE flame retardants (FRs) (Full list of analyzed contaminants in the supporting information). Extraction and analysis of PCBs/OCs/PBDEs/non-PBDE FRs were based on methods previously described (Letcher et al., 2018; McKinney et al., 2009). Briefly, blubber biopsies were cut lengthwise into two equal depth segments, one slice (excluding skin) for analysis of POP concentrations, and the other preserved for future studies. The blubber sub-sample for POP analysis (mean weight: 0.04 g, range: 0.01 to 0.18 g) was then accurately weighed into a mortar and homogenized with pre-cleaned diatomaceous earth (DE). An aliquot was used to determine lipid content gravimetrically. After spiking with a mixture of

13C-labeled and non-labeled C/PCB/FR surrogates as internal standards, extraction was performed by accelerated solvent extraction, then extracts were subjected to clean-up by gel permeation chromatography and solid phase extraction. The final extract was separately analyzed for PCBs and OCs by gas chromatography-mass spectrometry (GC-MS) with electron ionization (EI), and then for PBDE/non-PBDE FRs by GC-MS with electron capture negative ionization (ECNI). Identification and quantification were performed using MassHunter Quantitative Analysis software (Version B.07.01, Agilent Technologies). Each batch included ten samples, a blank, and standard reference material, the National Institute of Standards and Technology pilot whale (*Globicephala melas*) blubber homogenate (NIST-1945).

3.4.3 QA/QC results

The standard reference material SRM (NIST 1945 pilot whale blubber) was run eight times and checked for precision and accuracy. The overall POP recovery was 102 % (96-109 %) for ∑OCs (fourteen compounds), 105 % (99-111%) for ∑PCBs (thirty-three congeners) and 112 % (91-135 %) for ∑PBDEs (five congeners). Internal standard recoveries were 85 % (68-95 %) for PCBs (six ^{13}C-labelled congeners), 70 % (47-106 %) for OCs (eighteen ^{13}C-labelled compounds), and 150 % (89-214 %) for FRs (five ^{13}C-labelled compounds). Method limits of detection (MLODs) and quantification (MLOQs) were defined as the minimum amount of analyte which produced a peak with a signal to noise ratio of 3 and 10, respectively (McKinney et al., 2011c). A blank was run with each batch. No contamination was present in any of the blanks.

3.4.4 Dietary indicators

A Stable Isotope Bayesian Ellipses analysis (SIBER) was performed as per (Samarra et al., 2017c) on already published δ^{13}C and δ^{15}N data from the 45 whales sampled in 2014 to determine diet types (Jackson et al., 2011). Based on this analysis, a δ^{15}N cut-off was established

to delineate a diet-type, which was further validated using observational or photographic evidence of movement patterns and feeding habits based on predation events for these same whales, whenever available. Five whales sampled in 2016 were not assigned to a diet-type because the samples were not analyzed yet for $\delta^{15}N$ values. *Diet-type* was classified as 'mixed-diet' for whales that appear to feed on both fish and higher trophic level prey (including seals and small cetaceans) and 'fish-feeder' for whales believed to predominantly prey on herring.

3.4.5 Data analysis

The five main contaminant classes, *i.e.*: ∑PCBs, dichlorodiphenyltrichloroethane (∑DDTs), chlordane (∑CHLs), ∑PBDEs and hexachorobenzene (HCB) were quantified in >70% of all samples. Hexachlorocyclohexanes ∑HCHs and ∑non-PBDE FRs were only quantified in 10% and 17% of the samples, respectively, and most concentrations were <LOD. Thus, these compounds were reported in the results, but were not included in further statistical analyses. HCB was the only chlorobenzene detected. We henceforth refer to HCB instead of ∑ClBz. Three individuals were sampled in both 2014 and 2016 (IS018, IS067, and IS046, all males). We used a Student's t-test to determine if there were differences between years (Supporting information). For the other whales sampled more than once within the same year (n = 9 re-sampled individuals), we used a Student's t-test to determine if there were differences in contaminant classes between the samples. Because duplicate and triplicate biopsies from the same individual showed similar congener concentrations and profiles, their concentrations were averaged (Supporting information).

All POP concentrations were lipid corrected and expressed in $mg.kg^{-1}$ lipid weight (lw). As lipid content was missing for three individuals (IS243, IS229 and IS174), we estimated the values by interpolation from a regression of contaminant concentration with lipid content. We examined CB153, as it is one of the most recalcitrant PCBs in marine mammals, as well as

∑PCB (Mackay, 2006). The latter showed a stronger correlation with lipid content ($R^2 = 0.42$, $p < 0.001$ versus $R^2 = 0.36$, $p = 0.004$) and was thus used to approximate lipid content for the missing samples. Two outliers (IS069 and IS229) were removed from the contaminant dataset due to their high standardized residual values (>3) (Table S3-2). Prior to statistical analysis, concentrations were log-transformed (log x+1) to approximate normal distribution, which was evaluated and confirmed with qqplots on residuals and/or Shapiro tests.

We used a generalized linear modeling approach (GLM) to explore contaminant variation. The effect of three independent variables, *sex* (male, female), *diet-type* (fish, mixed), and *sampling-season* (2014-winter, 2014-summer), were tested for the concentrations of significant contaminant classes: ∑PCB, ∑DDT, ∑CHL, ∑PBDE, and HCB. The whales sampled in more than one season (e.g., winter and summer) were randomly assigned to either winter or summer. Individuals only sampled in 2016 (n = 5) were excluded from the models due to the lack of $\delta^{15}N$ values. Age class was not included in the model due to low sample size (Table S3-2). The three large male juveniles from the mixed-diet type were pooled with adult males as they were close to adulthood. We ran each possible model combination of the three independent variables for each contaminant class. We used Akaike's information criterion corrected for small sample size (AIC_c) and considered smaller AIC_c values to be indicative of better models. We also used the variance explained (1-(residual deviance/null deviance)), also known as McFadden's pseudo R^2, to determine how well the different models explained variation in pollutant classes within the population. A *sex* × *diet-type* interaction could not be included in the models due to the low sample size for marine mixed-diet females (n = 1) (Table S2). However, to further investigate the relationship between diet-types and POP concentrations, Pearson correlation tests were performed on the log-transformed contaminant classes and non-transformed $\delta^{15}N$ values for males and females separately (data available only for 2014 samples; (Samarra et al., 2017c)). To

assess how the individuals grouped in terms of contaminant concentration similarities, we performed a hierarchical agglomerative cluster analysis on log-transformed individual compound concentration when detected in more than 70 % of the samples using the Euclidean distance, and Ward's D2 method, bootstrapped 1000 times (eclust function of the factoextra package in R).

The compounds detected in more than 70 % of the samples were *cis*-chlordane, *p,p'*-DDD, *p,p'*-DDE, *p,p'*-DDT, dieldrin, heptachlor epoxide, hexachlorobenzene, Mirex, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, and CBs 52, 74, 95, 99, 101, 105, 118, 128, 132, 138/163, 146, 149, 150, 153, 170, 174, 177, 179, 180, 183, 187, BDE 47 and 100 (Table S2). For these, any non-detects (N.D.) were assigned a random value between 0 and the MLOD of the compound before inferential statistical analysis. Compounds detected, but below the MLOQ, were assigned a random value between MLOD and MLOQ. Overall, 18% of the dataset corresponded to values below MLOQ or MLOD. Finally, to further explore the intra-population variability in PCB patterns, we performed a PCA on arcsine-transformed CB congener percentages when they were detected in more than 70% of the samples.

3.5 RESULTS AND DISCUSSION

Concentrations of PCBs, which were the highest among all POP classes studied, showed a 300-fold difference among individuals within the population, ranging from 1.3 to 428.6 $mg.kg^{-1}$ lw (Fig. 3-2A). The next highest contaminant classes included DDTs and CHLs, which varied by up to 200- and 150-fold, respectively, among individuals (0.9 to 183.8 $mg.kg^{-1}$ lw for ∑DDTs and 0.4 to 61.2 $mg.kg^{-1}$ lw, for ∑CHLs) (Fig. 3-2B). For comparison, a similar 300-fold difference in PCB concentrations was documented between different populations of fish vs mammal-specialists in the eastern North Pacific (range from 1.7 $mg.kg^{-1}$ lw for the northern residents to 574 $mg.kg^{-1}$ lw for the North Pacific transients) (Buckman et al., 2011; Krahn et al., 2007). This similar variation within Icelandic killer whales compared to distinct ecotypes in

other areas is unexpected, given the killer whales in our study belong to the same population according to recent studies (Tavares et al., 2017; Tavares et al., 2018). This suggests that the ecology of individuals plays a critical and previously overlooked role in population exposures to PCBs and other biomagnifying contaminants.

We tested the effects of *diet-type, sex,* and *sampling-season* for each major POP class. To do so, individuals were first assigned to a diet-type. We determined diet-types by re-conducting a SIBER analysis on the published stable isotope data from Samarra et al. 2017c, using updated observed movement, and feeding behavior data (Samarra et al., 2017c) (See supporting information for the full analysis). Fish-feeding killer whales were characterized by low $\delta^{15}N$ values (<14‰) and/or followed herring closely around Iceland (Fig. 3-2B). Mixed-diet killer whales were frequently observed in herring grounds feeding on herring, but had also been observed to prey on marine mammals (*i.e.* seals or small cetaceans), had elevated $\delta^{15}N$ values (>14‰), and/or traveled to Scotland where they target marine mammals (Fig. 3-2B) (Supporting information). Five individuals could not be assigned to a diet-type due to a lack of $\delta^{15}N$ values for these whales. The clear distinction between the isotopic niches of individuals strongly suggests intra-population variation in foraging behavior (Samarra et al., 2017c) (Fig. 3-2B).

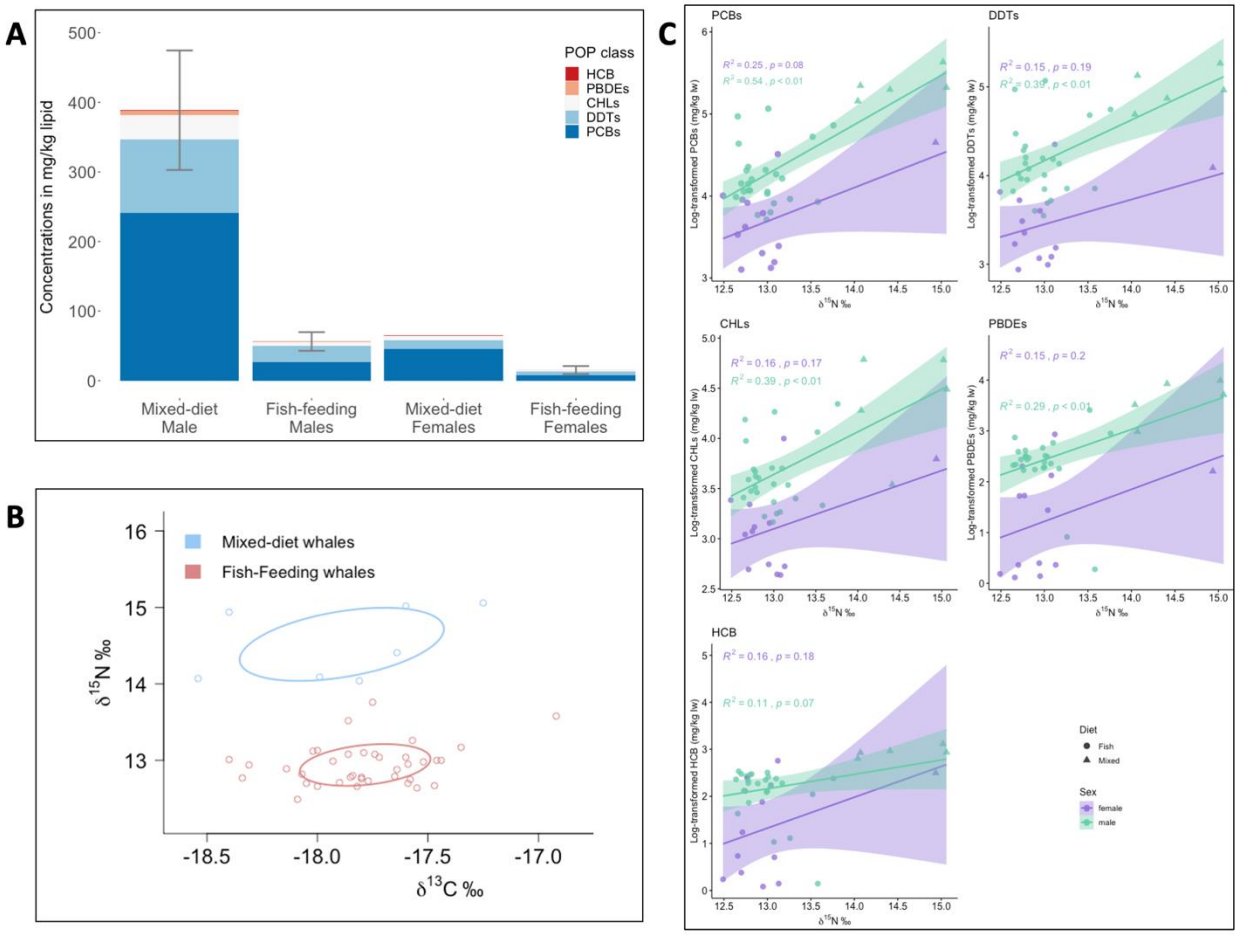

Figure 3-2: A) Concentrations of POP classes (mg.kg^{-1} lipid weight) in Icelandic killer whale blubber biopsies (results are expressed as arithmetic mean ± SE (except for the single mixed-diet female) ; B) Isotopic biplot from the SIBER (Jackson et al., 2011) stable isotope analysis conducted on $\delta^{13}C$ and $\delta^{15}N$ between the two diet types, modified from Fig. S1 to include the two defined diet-types (Samarra et al., 2017c); C) Correlations of log-transformed concentrations of ∑PCB, ∑DDT and ∑CHL, ∑PBDEs and HCB with $\delta^{15}N$ values for males and females.

Diet-type and *sex* were the two factors that most strongly explained concentrations of POPs within the Icelandic killer whale population based on our GLM modeling approach. Best fit models (Table 3-1) including these two predictors explained >50% of the variance among individuals in concentrations of PCBs, DDTs, CHLs, HCB, and PBDEs. The second-best or third-best models for all POP classes included *sampling season*. However, sampling season and its 95 % confidence intervals overlapped zero in all POP classes, and thus was considered not

significant in the POPs variation. *Diet-type* had a stronger effect than *sex* on contaminant variation for all POP classes, with estimates for *diet-type* predictors being on average 0.5 higher than estimates for *sex* predictors (Table 3-1). As a result, mixed-diet males and females had mean PCB concentrations 9.0- and 5.7-fold higher than fish-feeding males and females, respectively. Conversely, mean PCB concentrations for mixed-diet and fish-feeding males were just 5.3- and 3.3-fold higher than the mixed-diet and fish-feeding females, respectively (Table S3-1). PBDEs and hexabromocyclododecane (HBCDD) behaved similarly to legacy POPs, being detected more frequently and in higher concentrations in mixed-diet versus fish-feeding whales (Table S3-1). Furthermore, the effect of diet was evident from the positive linear association of POPs with trophic position ($\delta^{15}N$) across males and females, based on Pearson correlation tests (Fig. 3-2C). Indeed, a moderate positive correlation was found for ∑PCBs, weak positive correlations were found for ∑DDTs, ∑CHLs and ∑PBDEs, and a positive, but not significant, correlation was found for HCB, in males. For females, correlations were positive between the major POP classes and $\delta^{15}N$, although none of these relationships were significant. This similar association between $\delta^{15}N$ values and POP concentrations suggests an absence of a *sex* x *diet-type* interaction. Moreover, the difference in POP concentrations in Icelandic killer whales was larger between fish-feeding and mixed-diet whales than what was reported for Norwegian killer whales, where mixed-diet whales had PCB concentrations four times higher than fish-feeding whales (Andvik et al., 2020). This difference is consistent with a more pronounced dietary segregation between feeding types in Iceland.

Table 3-1:Summary results from the generalized linear modelling approach that tested the effects of three independent variables (sex, diet-type and sampling-season) on the log-transformed concentrations of ∑PCBs, ∑DDTs, ∑CHLs, HCB and ∑PBDEs in the blubber biopsies of the Icelandic killer whales sampled in 2014 and 2016.

Models	AIC_c	ΔAIC_c	AIC_c Wt	Variance Explained	Intercept	CI 95%	Predictor: Sex	CI (95%)	Predictor: Diet-type	CI 95%

∑PCBs ~ sex + diet-type	41.8	0	0.78	0.67	3.61	3.41 - 3.81	**0.61**	0.37 - 0.85	**1.11**	0.79 - 1.43
∑PCBs ~ sex + diet-type + sampling-season	44.4	2.54	0.22	0.67						
∑DDTs ~ sex + diet-type	46.2	0	0.78	0.61	3.39	3.18 - 3.60	**0.74**	0.49 – 1.00	**0.82**	0.48 - 1.15
∑DDTs ~ sex + diet-type + sampling-season	48.8	2.57	0.22	0.61						
∑CHLs ~ sex + diet-type	41.6	0	0.78	0.55	3.05	2.85 - 3.25	**0.57**	0.33 - 0.81	**0.76**	0.44 - 1.08
∑CHLs ~ sex + diet-type + sampling-season	44.2	2.56	0.22	0.55						
HCB ~ sex + diet-type	559.2	0	0.56	0.31	4.64	4.15 - 5.23	**0.8**	0.13 - 1.42	**1.33**	0.55 - 2.28
HCB ~ diet-type	561.2	2.03	0.20	0.23						
HCB ~ sex + diet-type + sampling-season	561.7	2.53	0.16	0.31						
HCB ~ diet-type + sampling-season	563.6	4.46	0.06	0.23						
HCB ~ sex	566.6	7.39	0.01	0.13						
HCB ~ sex + sampling-season	568.0	8.85	0.01	0.15						
∑PBDEs ~ sex + diet-type	99.0	0	0.76	0.54	1.14	0.75 – 1.53	**1.22**	0.76 – 1.70	**1.22**	0.60 – 1.84
∑PBDEs ~ sex + diet-type + sampling-season	101.3	2.36	0.24	0.54						

AIC_c: Akaike's Information Criterion corrected for small sample size. Only models with a ΔAIC_c below 10 are shown. AIC_c Wt represents the discrete probability of each model. Variance Explained was calculated for each model : 1-(Residual Deviance/Null Deviance).

Icelandic whales appear to manifest a long-term individual specialization on different prey rather than a generalist feeding behavior. Fish-feeding killer whales in our study had lower overall contaminant concentrations and a contaminant composition characteristic of fish-eating mammals (Andvik et al., 2020; Herman et al., 2005) (Fig. 3-3B, 3-3C and 3-3C). Indeed, the C2 cluster "fish-feeding males" (Fig. 3-3B) included most of the fish-feeding males, which were separated by low Euclidian distances, suggesting little differences in contaminant concentrations among them. Specifically, fish-feeding males were associated with less chlorinated and/or less persistent congeners in the PCA analysis (CBs 52, 95, 105, 101, 118). Limited variation in POP concentrations and patterns within males of this diet-type is consistent with reports of long-term

dietary specialization on herring (Samarra et al., 2017a; Samarra et al., 2017c) (Fig. 3-3A and 3-3B). Our cluster and PCA analyses revealed that POP profiles are similar among mixed-diet individuals but differed markedly from fish-feeding individuals (Fig. 3-3A, 3-3B and 3-3C). All mixed-diet individuals were grouped in the C3 cluster "mixed-diet" and PCB congener profiles were characterized by a large proportion of highly chlorinated and persistent congeners (CBs 153, 180, 170, 177 and 183), a characteristic of a marine mammal-based diet (Andvik et al., 2020; Herman et al., 2005) (Fig. 3-3A and 3-3C).

The results from this study provide new insights into the complexity of feeding habits adopted by Icelandic killer whales. Firstly, the whales categorized as mixed-diet were frequently sighted feeding on herring in Iceland (Samarra et al., 2017c), but exhibited contaminant profiles consistent with a diet that includes marine mammals. Secondly, our pattern analyses (Fig. 3-3A, 3-3B and 3-3C) suggest that the diets of some non-mixed-diet whales may, in fact, contain some higher trophic level prey (IS003, IS251 and IS136). This suggestion is supported by the large variability of both PCB and $\delta^{15}N$ values across individuals (Fig. 3-2C) and previous opportunistic observations of different prey events in Icelandic waters that involved mammals, birds, and other prey species (Samarra et al., 2018). Indeed, while contaminant loads reflect the whales' long-term feeding habits, at least for males, isotopic ratios only are indicative of feeding habits for the few weeks prior to sampling (based on studies performed on other cetacean species) (Aubin et al., 1990; Hicks et al., 1985). As a result, whales with lower $\delta^{15}N$ values could seasonally or occasionally prey on marine mammals and these events may only be detected in the whales' POP concentrations (Fig 3-2C). Some individuals in the population might thus be true generalists, occasionally preying on marine mammals, and show elevated POP concentrations. Higher-resolution dietary tracers like fatty acid signatures are needed to shed further light on individual-level prey composition (Bourque et al., 2018; Budge et al., 2006).

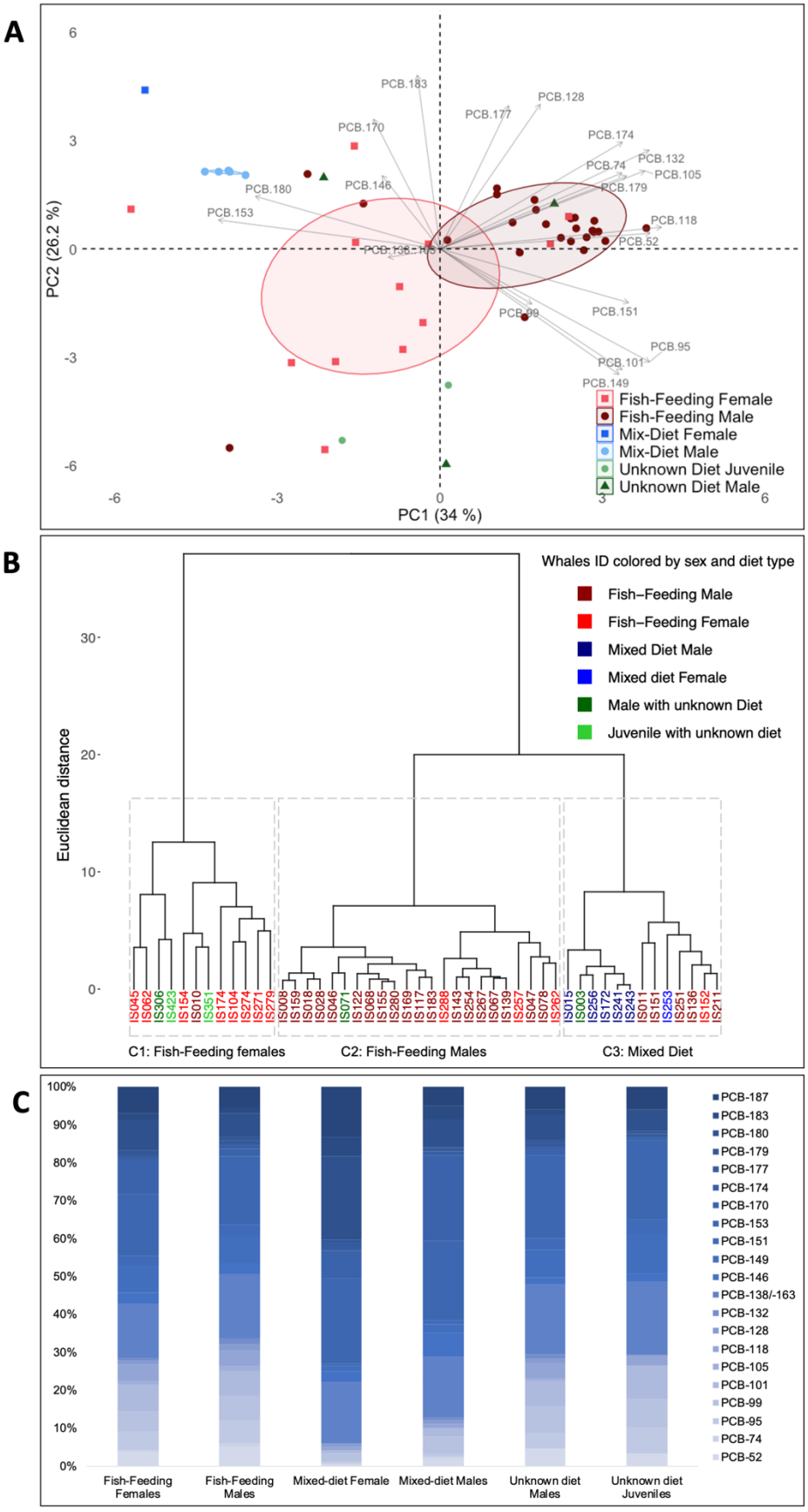
A
PC2 (26.2 %)
PC1 (34 %)
PCB.183
PCB.128
PCB.177
PCB.170
PCB.174
PCB.132
PCB.74
PCB.105
PCB.180
PCB.146
PCB.179
PCB.153
PCB.118
PCB.52
PCB.99
PCB.151
PCB.95
PCB.101
PCB.149
Fish-Feeding Female
Fish-Feeding Male
Mix-Diet Female
Mix-Diet Male
Unknown Diet Juvenile
Unknown Diet Male
B
Whales ID colored by sex and diet type
Fish–Feeding Male
Fish–Feeding Female
Mixed Diet Male
Mixed diet Female
Male with unknown Diet
Juvenile with unknown diet
Euclidean distance
C1: Fish-Feeding females
C2: Fish-Feeding Males
C3: Mixed Diet
C
PCB-187
PCB-183
PCB-180
PCB-179
PCB-177
PCB-174
PCB-170
PCB-153
PCB-151
PCB-149
PCB-146
PCB-138/-163
PCB-132
PCB-128
PCB-118
PCB-105
PCB-101
PCB-99
PCB-95
PCB-74
PCB-52
Fish-Feeding Females
Fish-Feeding Males
Mixed-diet Female
Mixed-diet Males
Unknown diet Males
Unknown diet Juveniles

Figure 3-3: Pattern analysis of contaminants in Icelandic killer whales. A) Principal component analysis on PCB congener composition among Icelandic killer whales. B) Hierarchical agglomerative cluster analysis based on log-transformed individual POP concentrations showing three clusters: C1: Fish-feeding females, C2: Fish-feeding males and C3: Mixed-diet. C) PCB congener composition among Icelandic killer whales. Each bar represents the percentage of each PCB congener in ∑PCBs.

Sex was the second most important factor contributing to contaminant variation in the Icelandic killer whale population. PCB concentrations were ~4-fold higher in males than in females for each diet-type, consistent with previous findings across reproducing killer whale (Table S3-1, S3-2) (Buckman et al., 2011; Ross et al., 2000a). Fish-feeding females had the largest within-group variation in contaminant concentrations and profiles (Fig. 3-3, 3-4). Indeed, most fish-feeding females grouped in the C1 cluster "fish-feeding females" showed a large inter-individual variation in POP concentration and their ellipse was the largest in our PCA, reflecting different PCB profiles. Adult female cetaceans are known to transfer ~10 % and 60 % of their body burdens to their offspring during gestation and lactation, respectively (Borrell et al., 1995; Ross et al., 2000a; Tanabe et al., 1982). As the largest portion of these burdens are offloaded during the first pregnancy (and nursing), contaminant levels may also vary with the number of births and thus, with age (Ross et al., 2000a; Wells et al., 2005). This could explain why three fish-feeding females clustered with the 25 fish-feeding males, and one clustered with the mixed-diet whales, possibly reflecting females that have not yet (successfully) reproduced (Fig. 3-3C). An exception to the established gender effect for contaminant transfer occurs in populations where contaminant loads impair reproduction, eliminating the primary excretion route for these compounds in females, leading to elevated tissue concentrations (Jepson et al., 2016).

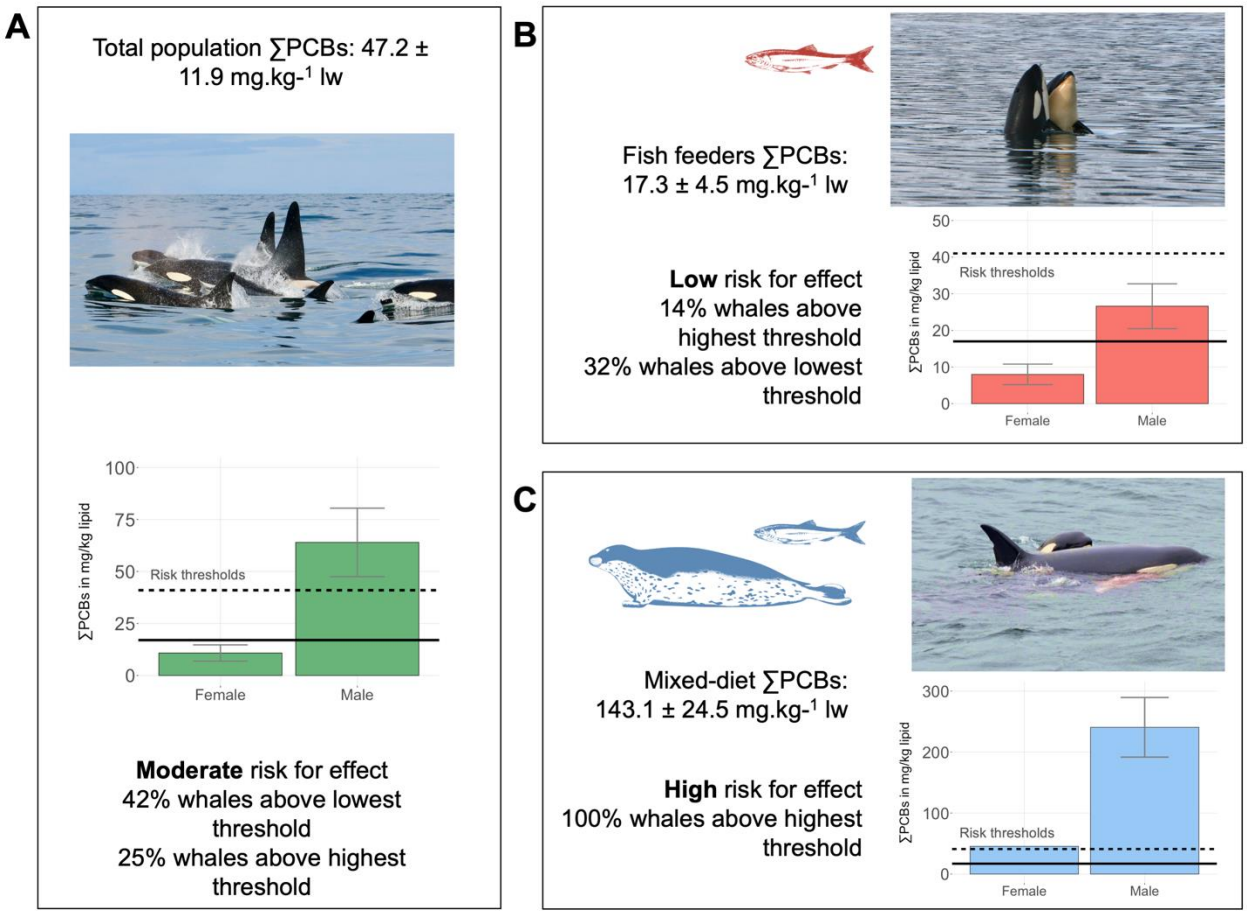

Figure 3-4:Risk assessment for Icelandic killer whales with respect to PCB exposure for A) the population as a whole, B) fish-feeding killer whales (25 males and 12 females) and C) mixed-diet killer whales (5 males and 1 female).

The striking differences in contaminant concentrations among Icelandic killer whales suggest that individual prey specialization and associated intra-population variation in POP loads should be considered in risk assessments going forward. A commonly used PCB concentration threshold for immunotoxic effects in marine mammals is 17.0 mg.kg^{-1} lw while the highest PCB toxicity threshold for impaired reproduction calculated for marine mammals is 41.0 mg.kg^{-1} lw (Fig. 3-4) (Helle et al., 1976; Kannan et al., 2000). In the North-Atlantic, some mixed-diet killer whales from Norway were recently reported to have PCBs concentrations above thresholds for

health effects (Andvik et al., 2020). In this study, out of the seven whales preying on seal to a certain extent, four had levels above the 41 $mg.kg^{-1}$ lw threshold. In Greenland, mean PCB concentrations for mammal-feeding sub-adult and adult killer whales were also above the 41 $mg.kg^{-1}$ lw (Pedro et al., 2017). A recent global killer whale modeling study predicted based on available blubber PCB data that the Icelandic killer whale population was not likely to face meaningful risk to population growth (Desforges et al., 2018). The model in this study used PCB concentrations based on five females (range: 14 to 41 $mg.kg^{-1}$ lw) from Iceland, showing concentrations similar to the fish-feeding killer whales in our study. However, these concentrations did not account for intra-population variation in feeding ecology and higher contaminant concentrations in mixed-diet individuals, particularly the males, as found in this study. All Icelandic mixed-diet killer whales from our study had PCB concentrations above the highest toxicity threshold, suggesting that these whales face increased risks of adverse reproductive and immune effects from PCB exposure alone, with potential consequences on population growth. Thus, the ecology of individuals must be understood to accurately forecast how environmental contaminants of toxicological concern may threaten the long-term persistence of the world's ultimate marine predator.

3.6 ACKNOWLEDGEMENTS:

Thank you to the Fair Isle Bird Observatory, particularly Sam Hood and David Parnaby, for kindly providing photographs on the seal predation event observed off Fair Isle. Thank you to Marie-Thérèse Mrusczok, Andrew Scullion and Hugh Harrop for their personal communication on a marine mammal predation event in Iceland. We also thank Sara Tavares for help in processing individual photo-identifications and for providing the genetic results for sex determination. Biopsy sampling methods were approved by the School of Biology Ethics

Committee of the University of St. Andrews. All research and sample collection was carried out in compliance with local regulations and under an institutional permit for the Marine Research Institute, Reykjavík. **Funding**: This work was funded by the Canada Research Chairs Program (to M.A.M., 950-232183) and the Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grants Program (to M.A.M., RGPIN-2019-05330). Supplemental support was provided by the Ecotoxicology and Wildlife Health Division, Environment and Climate Change Canada (to R.J.L.). Additional funding was provided by an Icelandic Research Fund START Postdoctoral Fellowship (grant number 120248042) and Project Grant (grant number 163060-051) to F.I.P.S. **Authors information:** R.J.L., M.A.M, R.D. and C.S. designed the study. F.I.P.S and G.V. collected the samples in Iceland. R.D. and C.S. ensured the transfer of the samples to Canada. A.R. and D.B. performed the lab experiment. A.R. and J.P.D. analyzed the data. A.R. wrote the manuscript. All authors discussed the results and implications and commented on the manuscript at all stages. **Competing interests**: Authors declare no competing interests.

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3.8 SUPPLEMENTARY INFORMATION

List of contaminants targeted in the POP analysis

PCBs: CB 16, 18, 19, 22, 25, 28, 31, 33, 44, 49, 52, 56, 60, 66, 67, 70, 71, 74, 77, 81, 82, 87, 95, 97, 99, 101, 105, 110, 114, 118, 123, 126, 128, 132, 138/163, 141, 146, 147, 153, 156, 157, 158, 167, 169, 170, 173, 174, 177, 179, 180, 183, 185, 187, 189, 194, 195, 199, 203, 206 and 209

OCs: chlorobenzenes (ClBzs: 1,2,4,5-tetraClBz/1,2,3,5-tetraClBz, 1,2,3,4-tetraClBz, pentaClBz, hexaClBz), hexachlorocyclohexanes (HCHs: α-HCH, β-HCH, γ-HCH), octachlorostyrene (OCS), chlordanes (CHLs: heptachlor epoxide, oxychlorodane, trans-chlordane, cis-chlordane, trans-nonachlor, cis-nonachlor), Dieldrin, DDTs (p,p'-DDE, p,p'-DDD, p,p'-DDT), and Mirex (photo-Mirex, Mirex)

PBDEs: BDE 3, 7, 15, 17, 28, 47, 49, 66, 71, 77, 85/155, 99, 100, 119, 138, 153, 154, 181, 183, 203, 205, 206, 207, 209

FRs: 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), pentabromoethyl benzene (PBEB), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), decabromo-diphenyl ethane (DBDPE), 2,4,6-tribromophenyl allyl ether (TBP-AE), tetrabromo-o-chlorotoluene (TBCT), pentabromotoluene (PBT), hexabromobenzene (HBB), pentabromobenzyl acrylate (PBB-Acr), pentabromo-p-xylene (TBX), 1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane (α-DBE-DBCH, β-DBE-DBCH), α-hexabromocyclododecane (α-HBCDD), octabromo-1,3,3-trimethyl-1-phenyl indane (OBTMPI), polybrominated biphenyls (BB 101 and 153), pentabromophenyl allyl ether (PBP-AE), 5,6-dibromo-1,10,11,12,13,13-hexachloro-11-tricyclotridecene (DBHCTD), 2,4,6-tribromophenyl-2,3-dibromopropyl ether (TBP-DPTE), 2,3-dibromopropyl pentabromophenyl ether (PBP-dbpe), bis(2-ethylhexyl)-tetrabromophthalate (BEH-TEBP), syn-Dechlorane Plus (syn-DDC-CO), anti-Dechlorane Plus (anti-DCC-CO).

Supplementary Text

SIBER analysis and diet-types

The killer whales in this SIBER analysis (Jackson et al., 2011) only included whales from 2014 and groups based on movement patterns established by Samarra et al. (2017) (Samarra et al., 2017) were updated based on new photo-ID data. Movement types included whales known to follow the herring stock between its spawning and overwintering grounds in Iceland, whales known to travel between Iceland and Scotland and known or believed to target both fish and marine mammals and whales sighted for one season only and for which the year-round movements and year-round feeding preferences are generally unknown. Two non-parametric Kruskal-Wallis ANOVA were used to test for differences in stable isotope values between the groups. The δ13C values did not differ ($p > 0.05$) between the "follows herring all year" group, the "travels to Scotland" group and the "not seasonally spotted" group. δ15N values for the "follows herring all year" group were significantly lower ($p = 0.021$) than the "not seasonally spotted" group and significantly lower ($p = 0.005$) than the "travels to Scotland" group. The "not seasonally spotted" group also had lower values than the "travels to Scotland" group ($p = 0.004$) (Figure S3-1).

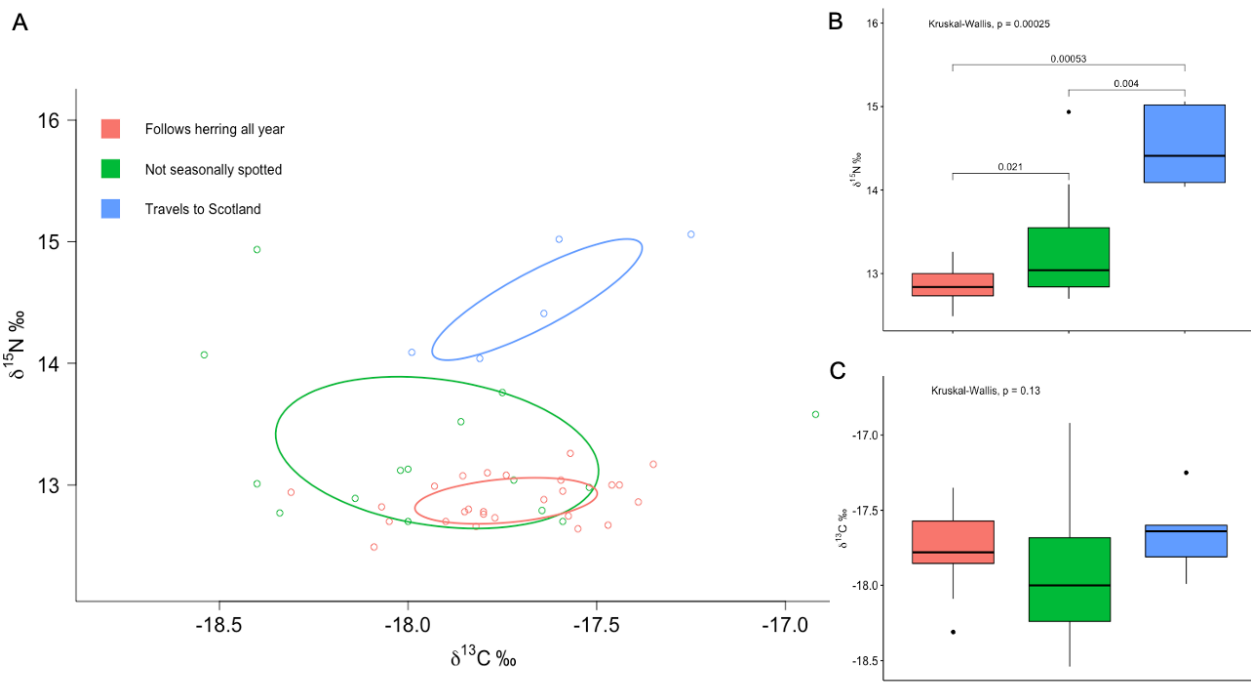

Figure S3-1: Results from the Stable Isotope Analysis. A) Isotopic biplot from the SIBER analysis conducted on δ13C and δ15N in 2014 whales (as per Samarra et al. 2017(Samarra et al., 2017)). The ellipses represent the core isotopic niches of each group (40%) B) δ15N (‰) in killer whale groups. C) δ13C (‰) in killer whale groups. The notation "follows herring all year" correspond to the whales known to follow herring stocks between their spawning and overwintering grounds in Iceland. The "travels to Scotland" correspond to the whales known to travel between Iceland and Scotland. The "not seasonally spotted" correspond to the whales sighted for one season only and for which the year-round movements are generally unknown.

The SIBER analysis showed no niche overlap for δ15N between the whales travelling to Scotland that are known or believed to target both fish and marine mammals ("travels to Scotland" group) and whales that follow herring all year ("follows herring all year" group), nor between whales travelling to Scotland and the whales only sighted seasonally in Iceland ("not seasonally spotted" group), with traveling individuals characterized by greater δ15N (Figure S3-1). There was an overlap in the isotopic niche of the whales only sighted seasonally in Iceland ("not seasonally spotted" group) and the whales that follow herring all year ("follows herring all

year" group); the calculated overlap was 0.138 ‰2, which accounted for 16 % of the "not seasonally spotted" ellipse area and 99 % of the "follows herring all year" ellipse area. The standard ellipse area corrected for a small sample size (SEAc) were 0.139 ‰2, 0.295 ‰2, and 0.858 ‰2 for "follows herring all year", "travels to Scotland", and "not seasonally spotted" groups, respectively. All whales from the "follows herring all year" group had δ15N values >14 ‰, thus this threshold was used to differentiate between diet types.

Based on δ15N values and photo-identification data, we identified two primary diet-types: fish-feeding and mixed-diet whales. All whales from the "follows herring all year" group were assigned to the fish-feeding group based on photo-identification records that suggest strong association to Icelandic herring coupled with low (<14 ‰) δ15N ratios (Samarra et al., 2017). Individuals from the "not seasonally spotted" group that had δ15N values below 14 ‰ were categorized as fish-feeders, as they overlapped with the "follows herring all year" group and were observed at least during part of the year in herring grounds in Iceland. The mixed-diet whales included whales that have been frequently observed feeding on herring but have also been confirmed or are suspected to feed at least to a certain extent on marine mammals. Two individuals from the "not seasonally spotted" group were assigned to the mixed-diet group (IS253 and IS256) because of their elevated δ15N values (above 14‰), lack of association of isotopic niche with other individuals in that group, and field observations of predation on a small cetacean (Mrusczok, pers. comm.). The five individuals from the "travels to Scotland" group were assigned to the mixed-diet group based on elevated δ15N, opportunistic observations of seal predation (individual ID #IS172, IS015 and IS229) in Scotland (Scullion, pers. comm., Harrop, pers. comm.), and/or confirmed travel to Scotland (individual IDs #IS241 and IS243) (Mrusczok & Scullion, 2019) where they are believed to target marine mammals. Individuals

sampled in 2016 (three adult males and two juveniles) were assigned to an "Unknown Diet" category due to lack of supporting information.

Contaminant results

Table S3-1: Concentrations of polychlorinated biphenyls, organochlorine pesticides and flame retardants (mg.kg−1 lipid weight) and lipid content (%) in Icelandic killer whale blubber biopsies sampled in 2014 and 2016. Results are expressed as arithmetic mean ± SE (min – max). Total includes 33 males, 13 females and two juveniles (two outliers were removed from the dataset).

	N	Lipid %	∑PCBs	∑DDTs	∑CHLs	HCB	Dieldrin	Mirex	∑HCHs	∑PBDEs	Non-PBDE FRs
All individuals	48	9.7 ± 1.2 (1.0 - 48)	47.2 ± 11.9 (1.3 - 428.6)	28.3 ± 6.1 (0.9 - 183.8)	8.6 ± 2.1 (0.4 - 61.2)	0.3 ± 0.04 (N.D. - 1.3)	1.4 ± 0.2 (N.D. - 6.2)	0.4 ± 0.1 (N.D. - 4.4)	– (N.D. - 0.5)	0.9 ± 0.3 (N.D. - 9.8)	– (N.D. - 0.3)
Male Fish-feeding	25	10.1 ± 1.2 (1.0 - 24)	26.6 ± 6.1 (5.2 - 116.3)	23.2 ± 6.0 (3.5 - 117.18)	5.86 ± 1.2 (1.4 - 22)	0.2 ± 0.02 (N.D. - 0.83)	1 ± 0.1 (N.D. – 3.0)	0.2 ± 0.1 (N.D. – 1.0)	– (N.D. - 0.5)	0.4 ± 0.1 (N.D. - 2.6)	– (N.D. - 0.1)
Female Fish-feeding	12	12.0 ± 3.9 (2.9 – 48.0)	8.0 ± 2.8 (1.3 - 32.4)	5.0 ± 1.9 (0.9 - 22.5)	2.1 ± 0.8 (0.4 – 10.0)	0.1 ± 0.04 (N.D. - 0.6)	0.4 ± 0.2 (N.D. - 0.3)	0.1 ± 0.03 (N.D. - 0.3)	– (N.D. - 0.2)	0.1 ± 0.1 (N.D. - 0.9)	– (N.D. - 0.07)
Male Mixed-diet	5	8.7 ± 3.2 (1.0 - 16)	240.6 ± 48.9 (143.6 - 428.6)	106.6 ± 23.8 (49.0 - 183.8)	35 ± 11.4 (3.5 - 61.2)	1.0 ± 0.1 (0.6 - 1.3)	5.3 ± 0.5 (3.5 - 6.2)	2.3 ± 0.6 (1.3 - 4.5)	– (N.D. - 0.5)	5.5 ± 1.6 (1.0 - 9.8)	– (N.D. - 2.6)
Female Mixed-diet	1	11.6 –	45.5 –	12.5 –	6.3 –	0.3 –	0.7 –	1.8 –	0.1 –	0.2 –	0.3 –
Male Unknown diet	3	3.0 ± 1.0 (2.0 – 5.0)	81.8 ± 66.4 (10.1 - 214.4)	54.7 ± 41.6 (8.9 - 137.8)	18.9 ± 15.1 (2.8 - 49.1)	0.5 ± 0.2 (0.2 - 0.8)	2.6 ± 1.4 (1.2 - 5.4)	0.7 ± 0.5 (1.2 - 5.4)	– (N.D. - N.D.)	1.3 ± 0.9 (0.3 - 3.1)	– (N.D. - 0.2)
Juvenile Unknown diet	2	3.2 ± 0.7 (2.5 - 3.9)	5.3 ± 1.9 (3.4 - 7.2)	4.7 ± 0.9 (3.8 - 5.5)	1.6 ± 0.8 (0.8 - 2.4)	0.2 ± 0.2 (<0.01 - 0.3)	0.9 ± 0.9 (<0.01 - 1.7)	<0.01 (<0.01 - <0.01)	– (N.D. - N.D.)	0.2 ± 0.2 (<0.01 - 0.5)	– (N.D. - N.D.)

Polychlorinated biphenyls are noted ∑PCBs, dichlorodiphenyltrichloroethanes : ∑DDTs, chlordanes : ∑CHLs, hexachlorobenzene : HCB, hexachlorocyclohexanes : ∑HCHs, polybrominated diphenyl ethers : PBDEs, Non-PBDE FRs mainly contain hexabromocyclododecane (α-HBCDD). N.D. indicates a value below the detection limit.

Table S3-2:Concentrations of polychlorinated biphenyls, organochlorine pesticides and flame retardants (ng.g−1 lipid weight), δ15N + δ13C (‰) and lipid percentage (%) in blubber biopsies of Icelandic killer whales. IS229 and IS069 were outliers and thus removed from the dataset.

Season	Year	ID	Sex	Age Class	*Diet-type*	Lipid %	∑PCBs	∑DDTs	∑CHLs	HCB	Dieldrin	Mirex	∑HCHs	∑PBDEs	∑non PBDE FRs	$\delta^{15}N$	$\delta^{13}C$
Summer	2016	IS003 (0108)	Male	Adult	Unknown	2	214440	137758	49054	840	5448	1743	N.D.	3123	159	NA	NA
Summer	2014	IS008 (0603)	Male	Adult	Fish	9	14254	10559	3887	299	925	130	N.D.	390	N.D.	12.70	-17.59
Summer	2014	IS010	Male	Adult	Fish	1	8519	7171	2149	1	0	0	N.D.	2	N.D.	13.58	-16.92
Winter	2014+2016	IS011 (0701)	Male	Adult	Fish	2	104787	106030	17460	302	1514	204	N.D.	742	N.D.	12.66	-17.82
Winter	2014	IS015 (993)	Male	Adult	Mixed	1	428580	183750	60540	1299	6239	4483	N.D.	9789	2581	15.02	-17.60
Summer	2014+2016	IS018 (9021)	Male	Adult	Fish	13	21544	16378	5192	228	781	191	N.D.	501	N.D.	12.98	-17.52
Summer	2014	IS028	Male	Adult	Fish	18	20599	19226	4918	268	906	110	167	168	N.D.	12.76	-17.80
Winter	2014	IS045	Female	Adult	Fish	3	6168	4001	1433	4	4	122	N.D.	1	N.D.	12.95	-17.59
Summer	2014+2016	IS046 (9706)	Male	Adult	Fish	4	17903	16027	4229	206	1299	129	N.D.	336	N.D.	12.79	-17.65
Winter	2014	IS047	Male	Adult	Fish	8	6455	4976	1781	167	223	61	N.D.	322	N.D.	13.04	-17.60
Summer	2014	IS062	Female	Adult	Fish	3	10146	6560	2427	6	16	2	N.D.	2	N.D.	12.49	-18.09
Summer	2014+2016	IS067	Male	Adult	Fish	14	9982	6956	2625	236	678	95	176	271	N.D.	12.64	-17.55
Summer	2014	IS068	Male	Adult	Fish	12	16393	13629	3437	190	585	77	55	181	N.D.	13.17	-17.35
Summer	2014	IS069	Female	Adult	Fish	15	11	120	N.D.	177	N.D.	N.D.	177	N.D.	N.D.	12.88	-17.64
Summer	2016	IS071	Male	Adult	Unknown	5	20549	17455	4726	226	1162	349	N.D.	443	N.D.	NA	NA
Winter	2014	IS078	Male	Adult	Fish	8	5152	3533	1465	122	547	2	N.D.	188	N.D.	12.99	-17.93
Winter	2014	IS104	Female	Adult	Fish	13	1267	871	494	4	256	35	N.D.	2	N.D.	12.70	-18.05
Summer	2014	IS117	Male	Adult	Fish	9	10722	10146	2324	128	619	48	N.D.	193	N.D.	13.00	-17.44

Summer	2014	IS122	Male	Adult	Fish	8	22655	21389	4653	190	774	369	285	308	N.D.	12.78	-17.85
Winter	2014	IS136	Male	Adult	Fish	10	72806	55644	22046	239	3053	995	N.D.	892	293	13.76	-17.75
Summer	2014	IS139	Male	Adult	Fish	16	11723	9019	2890	287	1079	76	N.D.	299	N.D.	12.80	-17.84
Winter+Summer	2014	IS143	Male	Adult	Fish	12	8240	5317	1940	119	1129	17	64	229	N.D.	13.08	-17.86
Winter	2014	IS151	Male	Adult	Fish	1	66866	58890	14901	831	4	299	N.D.	2578	N.D.	13.52	-17.86
Winter	2014	IS152	Female	Adult	Fish	4	32411	22514	9959	570	3046	292	N.D.	858	N.D.	13.12	-18.02
Summer	2014	IS154	Female	Adult	Fish	4	1561	1217	435	6	5	1	N.D.	133	N.D.	13.08	-17.74
Summer	2014	IS155	Male	Adult	Fish	9	14151	13763	3251	213	820	87	N.D.	411	N.D.	12.78	-17.80
Summer	2014	IS159	Male	Adult	Fish	11	16041	11911	4021	297	1175	87	N.D.	295	N.D.	12.82	-18.07
Winter	2014	IS169	Male	Adult	Fish	7	11381	9221	2988	133	823	96	N.D.	273	N.D.	12.73	-17.77
Winter	2014	IS172	Male	Large juvenile	Mixed	11	143581	48994	18990	634	3486	1314	N.D.	3265	1244	14.04	-17.81
Winter	2014	IS174	Female	Adult	Fish	NA	3388	1697	1102	8	3	135	N.D.	1	51	12.66	-18.00
Summer	2014	IS183	Male	Adult	Fish	9	11284	7047	3500	321	1017	111	332	244	N.D.	13.00	-17.46
Summer	2014	IS211	Male	Adult	Fish	25	43478	29704	9432	339	2807	275	N.D.	218	14	12.67	-17.47
Winter	2014	IS229	Male	Large juvenile	Mixed	NA	1691	757	176	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	14.09	-17.99
Winter	2014	IS241	Male	Adult	Mixed	7	210156	91838	30778	862	5129	1865	N.D.	5153	1478	15.06	-17.25
Winter	2014	IS243	Male	Large juvenile	Mixed	NA	198768	74267	3472	924	5693	2094	N.D.	8453	1518	14.41	-17.64
Summer	2014	IS251	Male	Adult	Fish	6	116294	117185	18391	261	1901	397	N.D.	324	N.D.	13.01	-18.40
Summer	2014	IS253	Female	Adult	Mixed	12	45479	12458	6280	316	694	1817	72	171	317	14.94	-18.40
Summer	2014	IS254	Male	Adult	Fish	22	5874	3991	1662	187	674	41	N.D.	174	N.D.	12.89	-18.14
Summer	2014	IS256	Male	Adult	Mixed	16	222089	134175	61228	846	5728	1859	456	964	531	14.07	-18.54
Summer	2014	IS257	Female	Adult	Fish	9	22248	14097	5125	65	236	221	N.D.	337	N.D.	12.71	-17.90
Summer	2014	IS262	Female	Adult	Fish	14	4202	3066	1184	130	369	37	N.D.	208	15	12.75	-17.58
Winter+Summer	2014	IS267	Male	Adult	Fish	14	9421	7249	2550	126	1000	81	N.D.	85	N.D.	13.26	-17.57
Summer	2014	IS271	Female	Adult	Fish	14	1331	987	441	174	297	148	180	28	N.D.	13.04	-17.72
Winter	2014	IS274	Female	Adult	Fish	4	2457	1538	530	0	20	2	N.D.	2	N.D.	13.13	-18.00
Winter	2014	IS279	Female	Adult	Fish	16	2014	1172	555	76	163	59	N.D.	2	N.D.	12.94	-18.31

Summer	2014	IS280	Male	Adult	Fish	8	18485	15281	5027	237	795	92	N.D.	582	N.D.	13.10	-17.79
Summer	2014	IS288	Female	Adult	Fish	48	8292	2262	1306	244	468	240	153	53	72	12.77	-18.34
Summer	2016	IS306	Male	Adult	Unknown	2	10114	8854	2811	418	1165	2	N.D.	332	N.D.	NA	NA
Summer	2016	IS351	NA	Large Juvenile	Unknown	3	3454	3779	766	5	2	2	N.D.	1	N.D.	NA	NA
Summer	2016	IS423	NA	Juvenile	Unknown	4	7192	5521	2339	316	1708	2	N.D.	458	N.D.	NA	NA

We used a Student's t-test to assess the variation of contaminant classes between years. For the other whales sampled more than once within the same year, we used a Student's t-test to determine if there were differences in contaminant classes between the samples. Sampling location/year was not identified as an informative variable in the model selection. Consistent with this, we found no differences for individuals (n = 3) who were sampled in both in 2014 and 2016 (p = 0.40 for PCBs; 0.45 for DDTs; 0.40 for CHLs; 0.10 for PBDEs; and 0.10 for HCB). The results indicate that the changes in POPs burdens in these killer whales was likely negligible on a two year-scale. The same way, no differences were found between samples of 9 whales sampled more than once in the same year (p = 0.92 for PCBs; 0.93 for DDTs; 0.92 for CHLs; 0.65 for PBDEs; and 0.84 for HCB).

Emerging contaminants

Emerging non-PBDE FRs were mostly found in mixed-diet whales (86 % of individuals), with less detections in fish-feeding whales (14 % of individuals). The main non-PBDE FR detected was HBCDD, with concentrations ranging from N.D. to 2.2 mg.kg-1 lw. When detected, mean HBCDD concentrations in mixed-diet whales were 1.0 mg.kg-1 lw while they were 0.16 mg.kg-1 lw in fish-feeding whales. To our knowledge, only one study in the literature that have reported on FR contaminants in killer whales. Fish-feeding southern resident killer whales, had HBCDD

concentrations of 0.08 mg.kg-1 lw (Jayda, 2018). It seems that at least some emerging FRs follow the same pattern as legacy POPs across diet-types.

Lipid percentages

Low lipid percentages were calculated for our study and could be the result of small biopsies. Our sample weight for our analyses was 0.04 g, range: 0.01 g to 0.18 g. Some samples were too small, and a gravimetric lipid determination could not be used to determine the lipid percentages. We were able to use the methods from a previous study to lipid normalize our data for missing lipid values, as lipid normalization of fatty tissue POP concentrations is the norm in wildlife ecotoxicology (Hebert & Keenleyside, 1995). Multiple studies on contaminant accumulation in killer whales reported low lipid percentages (< 12%) but these low percentages did not seem to affect the results, so long as they were reported on lipid weight basis (Krahn et al., 2004; Herman et al., 2005; Krahn et al., 2009; Lawson et al., 2020).

Geographical differences in POPs: Iceland vs. Scotland

The mixed-diet whales grouped in the cluster and PCA analyses, suggesting the geographic variation of POPs between killer whales supposedly feeding in Scotland and killer whales supposedly feeding in Iceland is negligible. Thus, the high POP concentrations and highly chlorinated PCB congener profiles suggest that these whales consistently predate on marine mammals.

CONNECTING PARAGRAPH

After observing variations in contaminant concentrations and profiles among Icelandic killer whales, I became curious about how we could accurately determine the diet of North Atlantic killer whales. These whales typically reside farther from shore and human settlements, except for Norway and Iceland. Consequently, traditional monitoring programs based on photo identification and long-term observations present challenges, particularly in the Western North Atlantic region above Newfoundland. To address this issue, I dedicated over a year to developing Quantitative Fatty Acid Signature Analysis for killer whales. This method shows great potential in accurately predicting the diet of killer whales by analyzing the fatty acid proportions in their blubber and that of their potential prey. However, developing this technique was no easy feat. Prior to our contributions, an essential factor was missing from the analysis: calibration coefficients that consider the predator's metabolism. Furthermore, the composition of killer whale blubber varies between its inner and outer layers, adding complexity to the method. I thus relied on archived samples from SeaWorld, that included full blubber samples for killer whales and whole fish that the whales ate while living at SeaWorld. Despite the considerable effort required, successfully developing this method stands as one of my proudest achievements during my PhD journey. The research was published in *Scientific Reports* and involved nine co-authors, including myself.

4 CHAPTER FOUR: VALIDATION OF QUANTITATIVE FATTY ACID SIGNATURE ANALYSIS FOR ESTIMATING THE DIET COMPOSITION OF FREE-RANGING CETACEANS

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Keywords: whales, diet estimation, fatty acids, QFASA, QFASAR, blubber, *Orcinus orca*, calibration coefficients

4.1 ABSTRACT

Accurate diet estimates are necessary to assess trophic interactions and food web dynamics of ecosystems, particularly for apex predators like cetaceans, which regulate entire

food webs. Quantitative fatty acid analysis (QFASA) has been used to estimate the diets of marine predators in the last decade but has yet to be implemented on free-ranging cetaceans, from which typically only biopsy samples containing outer blubber are available, due to a lack of empirically determined calibration coefficients (CCs), accounting for cetacean fatty acid (FA) metabolism. Here, we develop and validate QFASA for killer whales using full blubber from managed-care and free-ranging individuals. First, we compute full, inner, and outer blubber killer whale CCs from the FA signatures across the blubber layers of managed-care killer whales and their long-term diet items. We then run cross-validating simulations on the managed-care individuals to evaluate the accuracy of the estimates by comparing full-depth and depth-specific estimates to true diets. Finally, we apply these approaches to harvested killer whales from Greenland to test the utility of the method for free-ranging killer whales, particularly for the outer blubber. Accurate diet estimates for the managed-care killer whales are only achieved using killer whale-specific and blubber-layer-specific CCs. Modeled diets for the Greenlandic killer whales largely consisted of seals (75.9 ± 4.7%) and/or fish (20.4 ± 2.4%), mainly mackerel, which was in accordance with stomach content data and limited literature on this population. Given the remote habitats and below surface feeding of most cetacean species, this newly developed cetacean-specific QFASA method, which can be applied to biopsies, offers promise to provide a significant new understanding of diet dynamics of free-ranging cetacean species throughout the world's oceans.

4.2 INTRODUCTION

Accurate diet estimates are necessary to assess trophic interactions and food web dynamics of ecosystems, particularly in the case of apex predators, which can regulate entire food webs through trophic cascades (Springer et al., 2003). Cetaceans, especially toothed-whales, are at the top of the oceans' food webs and their effects on ecosystems have been

documented for decades (Estes et al., 2016). Nonetheless, their diets and related inter- and intra-population variation are not often well known, especially in remote regions where visual observation can be challenging (Newsome et al., 2010). While visual observations of cetaceans foraging in the wild can provide precious information on feeding ecology, data acquisition through observation of predation events is infrequent, limited to surface-events, often seasonal, and may not accurately reflect the long-term diet of a population (Bowen et al., 2013). Similarly, stomach contents and fecal samples are both challenging to obtain, only represent recent feeding patterns and, in the case of stomach contents, can only be obtained from deceased individuals, which may not represent the healthy part of the populations (Bowen et al., 2013). Thus, the use of chemical tracers measured largely from the blubber of biopsies collected remotely from cetaceans has increased in recent decades due to relative ease of sampling and ability to reflect integrated diet signals over time (Krahn et al., 2007; Remili et al., 2021). Stable isotopes of carbon and nitrogen in the skin have revealed some inter and intra-population variation in the feeding patterns in cetaceans, providing diet composition estimates mostly at the trophic level, although species-level precision from stable isotopes remains challenging (Foote et al., 2012; Pinzone et al., 2019; Remili et al., 2020). And while higher-resolution fatty acid (FA) signature analysis has also been applied to a couple of cetacean populations to infer dietary patterns, quantitative estimates of prey species in their diet using FA-based approaches from biopsies of free-ranging individuals have yet to be achieved (Bourque et al., 2018; Groß et al., 2020; Jory et al., 2021; Krahn et al., 2008).

FA signatures can be analyzed quantitatively to estimate the diets of predator populations, based on the knowledge that certain FAs are integrated with minor and predictable modification from the prey to the predator's fat storage tissues (e.g., blubber). The quantitative FA signature analysis (QFASA) model was developed to estimate the combination of prey FA

signatures that comes closest to matching that observed in the predator, after accounting for predator FA metabolism (Iverson et al., 2004). This model requires representative FA signatures of all major potential prey species, FA signatures of the predator, selection of an appropriate subset of diet-derived FAs to include from the total FAs monitored, species-specific calibration coefficients (CCs) to account for the predator FA metabolism, and a statistical model that minimizes the distance between the predator and the mixture of prey species representing the diet (Iverson et al., 2004). The QFASA method has been applied to grey seals (*Halichoerus grypus*), harbour seals (*Phoca vitulina*), and polar bears (*Ursus maritimus*), providing proportional estimates of the prey species composition of these predators' diets (Bourque et al., 2020; Iverson et al., 2004; McKinney et al., 2013; Nordstrom et al., 2008; Thiemann et al., 2008a). This type of analyses has not yet been applied to cetacean biopsies because CCs have not yet been determined for any cetacean species, due to the challenging aspects of managed-care feeding trials and the issue of FA stratification across cetacean blubber deposition (Bourque et al., 2018; Choy et al., 2019).

To apply QFASA to cetaceans, cetacean-specific (or even species-specific) CCs are likely required; one reason for this is that CCs allow the model to account for differences in the proportion of a given FA between the prey and predator due to predator-specific metabolism (Iverson et al., 2004; Kirsch et al., 2000). CCs are computed for each FA as the ratio of the FA proportion in the predator to the FA proportion in the prey. Although simple in nature, CCs have been shown to improve dietary estimates substantially (Iverson et al., 2004). Feeding trials were used in previous studies to successfully compute CCs for pinnipeds and mustelids (Iverson et al., 2004; Thiemann et al., 2008a). While feeding trials are logistically and financially difficult to implement for cetaceans, CCs could be computed from managed-care individuals fed a constant

diet over an extensive period to ensure proper and complete integration of the prey FA into the blubber.

Another challenge with applying QFASA to cetaceans, and particularly odontocetes, comes from the stratification of FAs throughout blubber layers (Bourque et al., 2018; Koopman, 2007). Dietary FA are more represented within the inner layer (closer to capillary and muscle layers) of the blubber and thus inner blubber has been the preferred sample for studying feeding patterns using FAs (Strandberg et al., 2008). CCs are likely to vary between layers; and thus blubber-layer specific CCs are likely required for cetaceans to avoid biased diet estimates. A recent study used CCs from mink (*Neovison vison*) to compare the known diets of two captive beluga whales (*Delphinapterus leucas*) (Choy et al., 2019). Although results were promising for estimating the diets of wild belugas (Choy et al., 2020), the potential for more accurate diet estimates using CCs developed specifically for cetaceans remains unmet. In addition, these previous beluga studies focussed only on inner blubber tissues (collected by subsistence-harvest), whereas most free-ranging cetaceans are remote biopsy darted, which only collects outer blubber (and skin). Since FAs are stratified throughout the blubber of cetaceans (Koopman, 2007), effectively applying QFASA to wild cetaceans would require investigating the predator FA signatures and determining CCs across blubber layers. Additionally, the inner and outer layers could be ecologically relevant as they may represent different feeding windows since ingested FAs are preferentially deposited in the inner blubber (Koopman et al., 1996). A recent study reported that the inner blubber of cetaceans (belugas) represents the diet two-to-five weeks prior to sampling (Choy et al., 2019). If deposition follows a pattern from inner layers to outer ones, FA signatures in the outer layers could then represent a diet integrated over a longer period, and potentially over multiple seasons (Iverson et al., 2004; Koopman, 2007).

As the oceans' top cetacean predator capable of highlighting strong individual feeding specializations, killer whales (*Orcinus orca*), are a prime candidate for the development of a QFASA method for cetaceans. They also have a thick layer of blubber, thus facilitating layer-specific analyses. Additionally, while feeding trials are difficult to implement, many managed-care killer whales are kept in facilities around the world, allowing access to both archived full-depth blubber samples and diet items. In this paper, we develop and validate QFASA for cetaceans, using killer whales. First, we compute both full-depth and depth-specific CCs to account for FA metabolism in killer whales using full blubber depth FA signatures from managed-care killer whales and FA signatures from four prey species representative of their known long-term (multiple years) diets. We then run cross-validating simulations on these managed-care animals to evaluate the accuracy of the estimates by comparing the full-depth and depth-specific estimates to their known long-term diets. Finally, we apply full, inner, and outer depth approaches to samples of harvested free-ranging killer whales, to further test the applicability of the method, not just for animals with full or inner blubber available, but also from animals for which only outer blubber is collected, thus maximizing the utility of this method for feeding ecology studies on free-ranging cetaceans.

4.3 MATERIAL AND METHODS

Full depth blubber samples were collected from four managed-care and 18 free-ranging whales, and the samples were divided into ten equal length sections as described in Bourque et al., 2018. Additional information regarding sample collections and FA analyses can be found in the supplementary text and Table S4-1 and S4-2.

4.3.1 Development of QFASA using managed-care whales

In addition to FA signatures of the predator and of all potential major prey, the QFASA model requires (1) designating a particular set of FAs to use from the ~70 routinely monitored, (2) the development of species-specific CCs, and (3) a statistical model that estimates the proportional prey composition by minimizing the statistical distance between the CC-corrected predator signatures and the average prey signatures (Iverson et al., 2004). The approach for each of these steps is detailed below.

4.3.1.1 Fatty acid sets

To develop the model, we tested two sets of FAs, dietary fatty acids, which only arise from the diet, and extended dietary fatty acids that also include fatty acids partially biosynthesized by the predator (Iverson et al., 2004). The first set included every dietary FA (Iverson et al., 2004) that was above 0.1% of the total FA signature to minimize analytical variation associated with small peaks on the GC-FID; this set consisted of 21 FAs (Table S4-2). The second set included every extended dietary FA (Iverson et al., 2004), but again only those exceeding 0.1% of the total FA; this second set amounted to 30 FAs.

4.3.1.2 Calibration Coefficients

The CCs were generated from the FA signatures of the four manage-cared killer whales and the FA signatures from the four species that formed their constant, long-term (multiple years) diets. As Bourque et al. (2018) found no statistically significant differences between layers 1-4 and between layers 6-10 for the killer whale samples, throughout the current study, "full blubber FA" refers to the lipid-weighted average of all 10 layers, while "inner blubber FA" refers to the lipid-weighted average of layers 1-4 and "outer blubber FA" refers to the lipid-weighted average of layers 6-10. Three CC sets were estimated using the lipid-weighted FA signatures averaged across layers for: full blubber CCs, inner blubber CCs, and outer blubber

CCs. The CCs were calculated as the ratio of a given FA in each blubber layer of a killer whale to the ratio of that FA in its diet, weighted by the proportions of each diet item, as follows

$$CC_{FA_{i,KW_j}} = \frac{FA_{i,KW_j}}{(FA_i,\ Herring_j) \times 0.6 + (FA_i,\ Capelin_j) \times 0.32 + (FA_i,\ Mackerel_j) \times 0.04 + (FA_i,\ Salmon_j) \times 0.04}$$

where FA_i is the percent of a FA *i* in whale or prey item *j*, and 0.60, 0.32, 0.04, and 0.04 in the denominator are the weight fraction of the respective prey in the whales' diets. Given that there were four killer whales, ten capelin *(Mallotus villosus)*, ten Pacific herring (*Clupea pallasii)*, ten Pacific mackerel *(Scomber japonicus)*, and four sockeye salmon *(Oncorhynchus nerka)*, we ran all possible combinations of killer whales and diet items, which generated 4000 CCs per FA per killer whale for each blubber layer. From this, we calculated the 10% trimmed mean for each killer whale. The final CC for each blubber layer was computed as the mean of the four trimmed means. Full blubber CCs correspond to the average of the ten layers (with the FA signatures of the killer whales weighted by the lipid content of each layer), while the inner blubber CCs corresponds to layers 1-4 and outer blubber corresponds to layers 6-10, both also consisting of FA signatures weighed by the lipid content of each layer. In addition to using our own killer whale-derived CCs and to determine the sensitivity of the method to different CC sets, we also ran QFASA with previously published mink CCs (herring-fed) and grey seal CCs (Iverson et al., 2004; Thiemann et al., 2008a), as they were the most used CCs in other QFASA studies on marine mammals. The list of CCs generated in this study can be found in Table S3.

4.3.1.3 The QFASA statistical method

The QFASA model was run in R, version 3.6.1. (R Core Development Team 2019) using the QFASAR package (Bromaghin, 2017). Diet estimations in QFASA rely on multiple assumptions. First, QFASA relies on the principle that predator FA signatures can be modeled as a linear mixture of the prey FA signatures (Iverson et al., 2004). Thus, we expect the predator FA signature to be within the prey FA range; and not meeting these criteria indicates poor CCs or an

incomplete prey library (Bromaghin, 2017). We tested our data using the function *pred_beyond_prey* in QFASAR to estimate the proportion of the predator's FA values that are outside the range of their prey values. To visually test for the improvement of the datasets when full blubber CCs were applied to the predator full blubber FA signatures, we also performed a Principal Component Analysis (PCA) using the FactomineR package with the FAs (both with and without CCs applied) and the prey signature to visualize whether use of the CCs brought the FA signatures of the predator closer to the prey FA space. A second QFASA assumption is limited overlap among the prey species' FA signatures (Iverson et al., 2004). To test for this assumption, we used the *leave_one_prey_out* (LOPO) function which removes one prey signature from the library at a time and recomputes the mean prey-type and then estimates the diet of the removed prey signature. The analysis performs this computation on each prey signature, one at a time. The final output indicates the proportion of samples attributed to the correct species. We chose the best FA set to use based on their performance in the LOPO and *pred_beyond_prey* analyses.

After choosing the best FA set, we ran multiple simulations using different sets of CCs. Each set of simulations was run using both the Kullback-Leibler (KL) distance (Iverson et al., 2004) and the Aitchison distance (Stewart et al., 2014), as the literature has not yet settled on the best distance to use (Zhang et al., 2020). To evaluate which distance performed best, we determined how both scored in both the QFASA diagnostics (assumptions tests) and on the accuracy of the diet estimates relative to the true diet of the managed-care killer whales.

First, to quantitatively test the need for CCs, we ran QFASA without CCs since some studies have suggested that QFASA might not need CCs to provide correct estimates (Budge et al., 2012; Happel et al., 2016). Two simulations were run on full blubber FAs with no CCs: one with KL distance and the other with Aitchison distance. Each simulation resulted in different

proportions of prey species in the diet, referred to hereafter as "diet estimates". The means and SEs for the diet estimates were computed using bootstrap sampling (n=100), as previously described (Bromaghin, 2015; Bromaghin et al., 2017; Choy et al., 2020). The accuracy of each diet estimate was inferred from the % error (absolute value (true diet estimate - modeled diet estimate)/ true diet estimate *100) to assess whether accurate diet estimates could be achieved without the use of CCs. Next, we compared the accuracies of the diet estimates using CCs developed for phocids and mustelids on different FA layers to assess the need for a cetacean-specific CC set (Iverson et al., 2004; Thiemann et al., 2008a). Three simulations were run on the dietary FA set with either mink or grey seal CCs using the KL distance (on full blubber, inner blubber, and outer blubber) and three on the same CCs using the Aitchison distance. We then tested the accuracy of the diet estimates using the CCs we developed from the managed-care whales and their diets. Here, it would have been circular to estimate the diets of the same killer whales that were used to generate the CCs; instead, we cross-validated the CCs, by estimating the diet of two of the four killer whales (either full, inner, or outer blubber FA signatures) using the mean full, inner or outer blubber CCs of the two other killer whales. We performed this for all possible combinations of killer whales (six) for each part of the blubber. We then determined the mean diet estimates generated from these iterative analyses and calculated the % error relative to the true diet. This allowed us to test the robustness of these CCs and ensure that the individual CC variation did not impact the overall diet estimates across the layers. Next, we ran simulations with blubber layer-specific CCs (the average of the four whales for each layer) and layer-specific FA signatures to test the need for layer-specific CCs. To do this, we ran twenty simulations in total on the managed-care killer whales (ten with the KL distance, and ten with the Aitchison distance). Out of the ten simulations for each distance, five matched the CC layer to the FA layer testing the accuracy of the diets and five were mismatched testing whether

matching CCs to their respective FA mattered, and especially for the outer layers representing a biopsy to see which CC set worked best on these outer blubber FA. The options were: 1) full blubber FA and full blubber CCs, 2) layer 1 FA and layer 1 CCs, 3) layer 1 FA and inner blubber CCs, 4) inner blubber FA and layer 1 CCs, 5) inner blubber FA and inner blubber CC, 6) outer blubber FA and full blubber CCs, 7) outer blubber FA and outer blubber CCs, 8) outer blubber FA and layer 10 CCS, 9) Layer 10 FA and outer blubber CCs and 10) Layer 10 FA and Layer 10 CCs.

4.3.2 Diet estimations in the free-ranging killer whales and application of the model to remote biopsies

After choosing the FA sets, CC sets and the statistical distance that yielded the most accurate estimates, we applied QFASA to the free-ranging killer whales using our prey library consisting of various fish and marine mammal species. Due to the large difference in fat percentage between the potential prey types (marine mammal vs fish), we adjusted the dietary estimates to prey lipid percentages using the *adj_diet_fat* function. For fish, we used the whole-body fat % calculated during the lipid extraction (mean: 16% for herring, 22% for mackerel). Because killer whales tend to not just eat the blubber when they consume marine mammals, we used a fat percentage of 30%, which was the average fat percentage calculated for the whole body of harbor seals (Burns et al., 2005). The lack of whole-body fat percentage for other marine mammal species prompted us to use 30% for all seals and whales. Remote biopsies from free-ranging cetaceans (usually between 10mm and 40mm depth (Noren et al., 2012)) typically only collect the outer blubber layers , e.g., some combination of layers 10 through possibly 6, as defined in our analysis. To confirm that the newly developed QFASA model on killer whales can accurately predict diets of free-ranging individuals, and to determine which CC should be used on biopsies, we ran QFASA on the outer layers (6-10) and layer 10 FA using the outer layer CCs

or layer 10 CCs (four combinations total) and compared the resulting estimates with the ones generated using full blubber FA and full blubber CCs.

4.4 RESULTS

4.4.1 Calibration coefficients

CCs, which were generated from the FA signatures of the managed-care killer whales and from the FA signatures of their known diet items (Table S4-2), showed differences between the inner and outer blubber of the managed-care killer whales (Fig. 4-1, Fig. S4-1). The CCs calculated for the full and outer blubber of the managed-care killer whales were distinct from those previously generated from captive feeding trials on mink and grey seal CCs, especially for 18:3n3 (Fig. 4-1, Table S4-3). Some of the inner layer killer whale CCs, however, appeared to be more similar to the CCs from mink and grey seals.

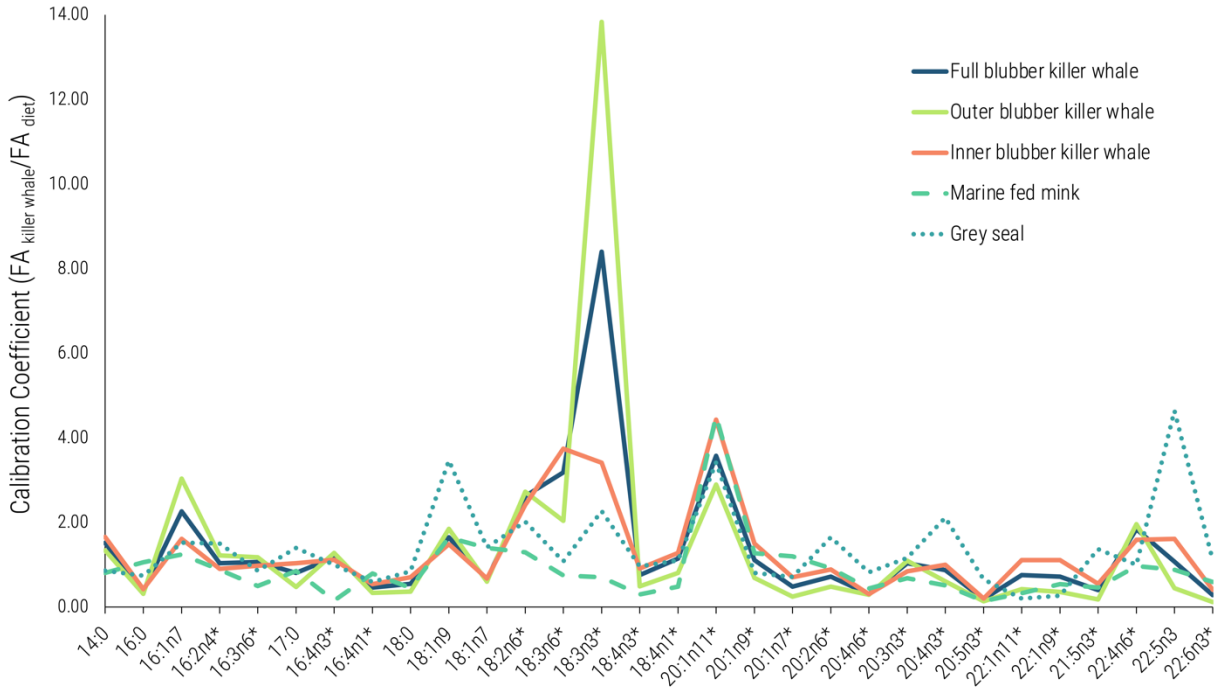

Figure 4-1:Calibration coefficients (CCs; ratio of each fatty acid in the predator to that in its diet) used in the QFASA simulations for cetaceans. Killer whale CCs were calculated from four managed-care killer whales and their known diet species in the current study, while the grey seal

CCs and the marine-fed mink CCs were reported previously (Iverson et al. 2004; Thiemann et al. 2008). CCs showed here include all dietary (with asterisk) and extended dietary FAs that were above 0.1% of total FAs. Dietary FAs only arise from the diet, while extended dietary FAs also include FAs partially biosynthesized by the predator.

4.4.2 QFASA on the managed-care killer whales

The goodness-of-fit check for the model for the full depth blubber using the function *pred_beyond_prey* showed that 39.3% of the predator FA were outside the range of prey FA without CCs. In contrast, with CCs derived from the managed-care killer whales on the dietary FA set, only 11.9% of the predator FA were outside the range of prey FA, indicating an appropriate set of CCs and prey library. The other assumption of QFASA (*i.e.*: quality of the prey library) was tested using the *leave_one_prey_out* (LOPO) function on the dietary FA sets and resulted in between 84.4% and 91.2% of correct species attribution on average (KL and Aitchison distance, respectively). The extended dietary FA set (n=30) scored similarly in the LOPO analysis, between 87.2% and 91.8% on average (Kullback-Leibler (KL) and Aitchison distance, respectively) (Table S4-4). The extended dietary set scored higher in the *pred_beyond_prey* (17% of predator FA outside the prey FA range), which indicates a poorer fit. Thus, although relatively similar, we selected the dietary FA set (n=21) for subsequent analyses.

Killer whale CCs were necessary to estimate the managed-care killer whales' diet accurately. The need for these CCs was visually supported by a PCA run on the dietary FA set for the managed-care killer whales and their prey, using various CCs (including no CCs) applied to the full blubber FA signatures (Fig. 4-2). Figure 2 demonstrates that applying full depth CCs to the full depth FA signatures puts the predator FA signatures into the prey FA range, while no CCs or CCs from seals or mink leaves the full depth FA signatures well outside the prey range. Moreover, QFASA simulations on the four managed-care killer whales without any CCs resulted

in a large overestimation of herring in their diets (Table S4-5) as herring reached a mean of 100% in the modeled diet for each killer whale, compared to 60% in the real diet. CCs calculated for non-cetacean species were also not successful at estimating the diets of the managed-care killer whales. The mink and grey seal-derived CCs showed, on average, 80.7% error compared to the true diet when the full blubber FA signatures were used, and 102.8% error using outer blubber FA signatures. Mink and grey seal CC were only somewhat better at estimating the diets based on the inner blubber FA signatures (55.3% error) (Table S4-6). The CCs from these other species overestimated the proportion of herring (76.1% herring on average) and underestimated the proportion of capelin (16.6% on average) in the diet of the managed-care whales, especially when used on the full blubber and outer blubber FA signatures. Thus, we decided to only use killer whale CCs (derived from killer whales) for remainder of the study.

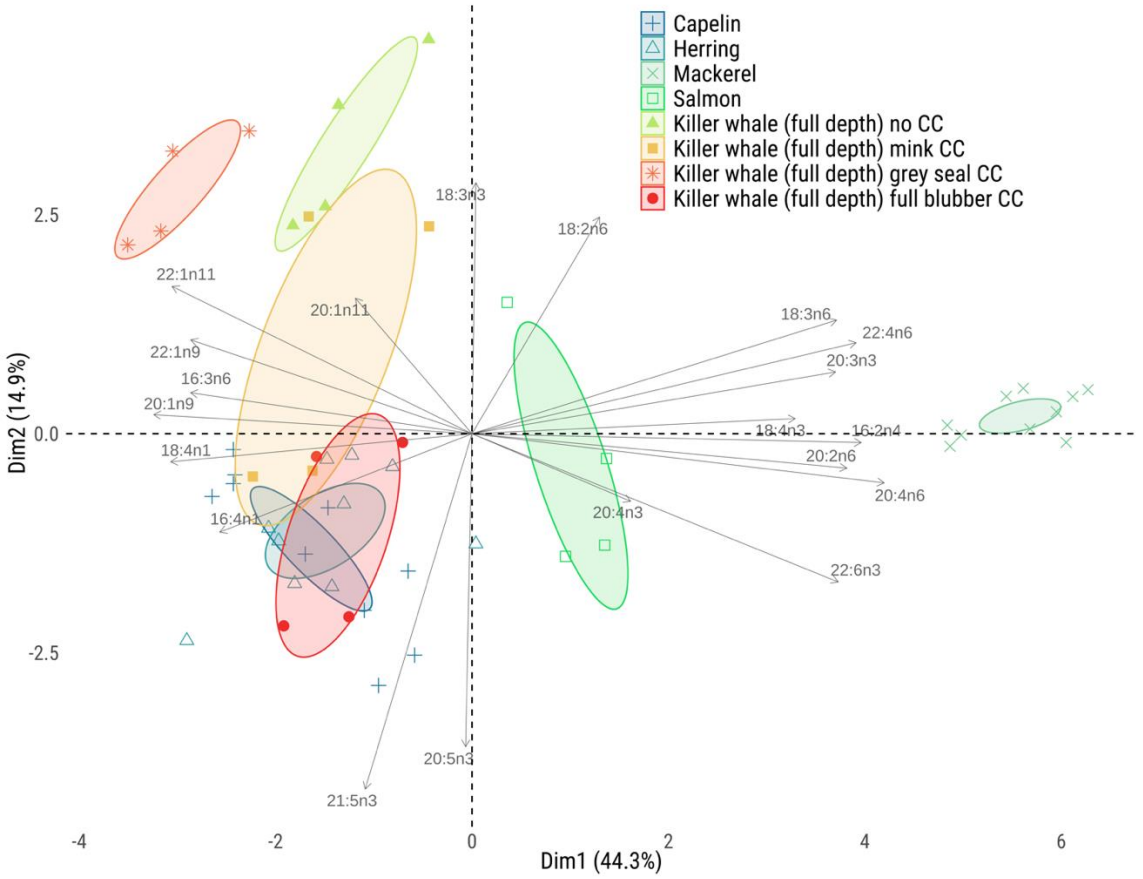

Figure 4-2: Principal component analysis of dietary fatty acid (FA) signatures in prey species, as well as in managed-care killer whales without calibration coefficients (CCs), with previously published mink and grey seal CCs, and with killer whale CCs generated for this study. Applying killer whale CCs to the predator moved their FA signatures within the ranges of the prey FA signatures (i.e.: capelin and herring mainly)

Diet estimates showed good accuracy in the cross-validation tests. (*i.e.*: running the model on two of the four managed-care killer whales, using the average CCs from the two other whales, thus running six different simulations per layer) (Table S4-7). The average diet estimates were highly accurate (18.0% error with the KL distance, and 25.1% error with the Aitchison distance), with estimates for capelin and herring being very close to the true diet: 35.2 ± 7.3 for capelin and 59.0 ± 8.9 for herring (KL distance). Two-by-two comparisons for the inner blubber yielded accurate estimates (6.3 % error with the KL distance vs. 39.6% with the Aitchison distance). Similarly, two-by-two comparisons for the outer blubber resulted in more accurate estimates with the KL distance (23.3% error with the KL distance vs. 30.2% with the Aitchison distance) (Fig. 4-3, Table S4-7)

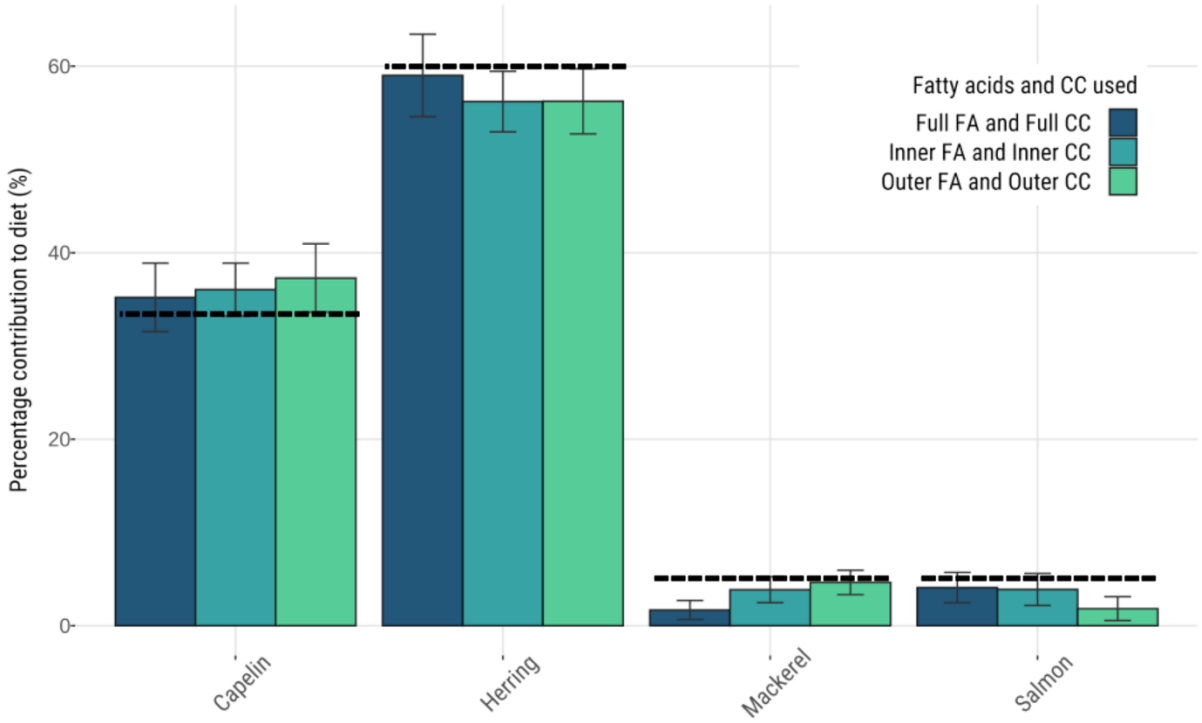

Figure 4-3: Mean diet estimates (in %) for the four managed-care killer whales obtained for the cross-validation analyses (estimating the diet of two whales using the CCs of the two other whales) and based on the prey library consisting of capelin (n = 10), herring (n = 10), mackerel (n = 10) and salmon (n = 4). The Kullback-Leibler distance was used with the dietary FA set. The true diet (dash line) fed to the managed-care killer whales consisted of 32% capelin, 60% herring, 4% mackerel and 4% salmon.

Although both statistical distances resulted in accurate diet estimates, using the KL distance resulted in more accurate estimates than the Aitchison distance when used with killer whale CCs. Additionally, diet estimates computed using the KL and CC means from the four whales were closer to the real diet than when using the Aitchison distance: 21.7% error (KL) vs. 22.1% (Aitchison) for full blubber FA signatures with full blubber CCs; 16.8% error vs. 29.83% error for the inner blubber FA signatures with inner blubber CCs; and 25.7% error vs. 32.1% error for outer blubber FA signatures with outer layer CCs (Table S4-8 and S4-9). Both the Aitchison and KL distances scored a high correct attribution rate of the prey to its species (91.0

% on average for the Aitchison distance vs. 84.4% on average for the KL distance), and results were too close to determine which LOPO analysis was better (Table S4-4). Thus, as the performance overall was somewhat better with the KL distance, it was selected to estimate the diets of the free-ranging killer whales.

Killer whale CCs accurately estimated the diet of the managed-care killer whales, provided that the appropriate layer-specific CC was used (e.g., inner blubber CCs to model the diet based on inner blubber FA signatures) (Fig. 4-3 and S4-3). The accuracy was lower when we did not use the CC set corresponding to the FA layer (Table S4-8 and S4-9). Indeed, while using full blubber CCs on full blubber FA signatures resulted in 21.7% error compared to the true diet, using full blubber CCs on outer blubber FA signatures resulted in 86.0 % error with a large overestimation of herring (89.6% estimated vs. the true diet of 60%). Our results with the KL distance showed that the inner layer average CCs yielded more accurate estimates (16.78 % error) on inner blubber FA signatures compared to layer 1 CCs (50.7% error). In the same way, layer 1 CCs produced the most accurate estimates when used on layer 1 FA signatures (18.1% error vs. 48.4% error on inner blubber FA signatures), and layer 10 CCs yielded more accurate estimates when used on layer 10 FA signatures (20.2% error) than when used on outer blubber FA signatures (67.8% error). Finally, outer blubber CCs yielded a good accuracy when used on the outer blubber FA signatures and on layer 10 FA signatures (25.7% error for outer blubber FA, and 33.5% error for layer 10 FA).

4.4.3 Diet estimations in the free-ranging killer whales and application of the model to remote biopsies

With the determination of the dietary FA set, the KL distance, and the layer-specific killer whale CCs being the optimal parameters for estimating managed-care killer whale diets, we applied QFASA to estimate the diets of free-ranging killer whales from Greenland and the

Faroe Islands. Since these killer whales were harvested or stranded, we had access to full blubber samples and were able to estimate the diets in a layer-specific manner. First, we verified our prey-library by running the LOPO analysis on the prey library and found that harp seals *Pagophilus groenlandicus*) and hooded seals (*Cystophora cristata*) overlapped. Therefore, harp seals and hooded seals were grouped into one category for diet estimation purposes. Additional QFASA method checks on the free-ranging killer whales (including the justification for using the KL distance over the Aitchison distance) are available in the supplementary text, and Table S4-10.

Based on our dietary estimates, free-ranging killer whales in Greenland (n=16) fed mainly on all species of seal present in our prey library, as well as on mackerel (Fig. 4-4). We ran QFASA models separately on the full depth, inner blubber, and outer blubber FA signatures, using the full depth CCs, inner blubber CCs and outer blubber CCs, respectively. The proportion of total seals in the Greenlandic killer whales' diets was estimated to be 82.56 ± 5.93% in the inner blubber, 67.0 ± 5.2% in the outer blubber and 75.9 ± 4.7% in the full blubber, and consisted of bearded seal (*Erignatus barbatus*), harp/hooded seal, and ringed seal (*Pusa hispida*) (Fig. 4-4). The proportion of total fish was estimated at 20.1 ± 3.4% in the inner blubber, 15.5 ± 1.6% in the outer blubber and 20.4 ± 2.4% in the full blubber. This consisted nearly entirely of mackerel (*Scomber scombrus*), with almost no herring (*Clupea harengus*) estimated in the diets. Diet estimates were low for baleen whales in all layers except the outer blubber, where bowhead whales were estimated to minimally contribute to the killer whales' diet (7.0 ± 2.5%). We reported the individual estimates for the inner layer for each free-ranging killer whale in Table S4-11 with stomach content data, when available. Whales with harp and/or hooded seals reported in the stomachs, often had a high dietary estimate for harp and hooded seals, and for seals in general (between 51.7% and 100% for total seal percentage). Both killer whales from the Faroe

Islands (n=2), conversely, showed higher proportions of fish (herring and mackerel) in their dietary estimates (27.9% and 80.3%) than all the Greenland whales. One of the two Faroese killer whales was estimated to have fed nearly exclusively (72.6%) on herring, while the whale had an estimate of mackerel (27.9%) and ringed and bearded seals (Fig. 4-4).

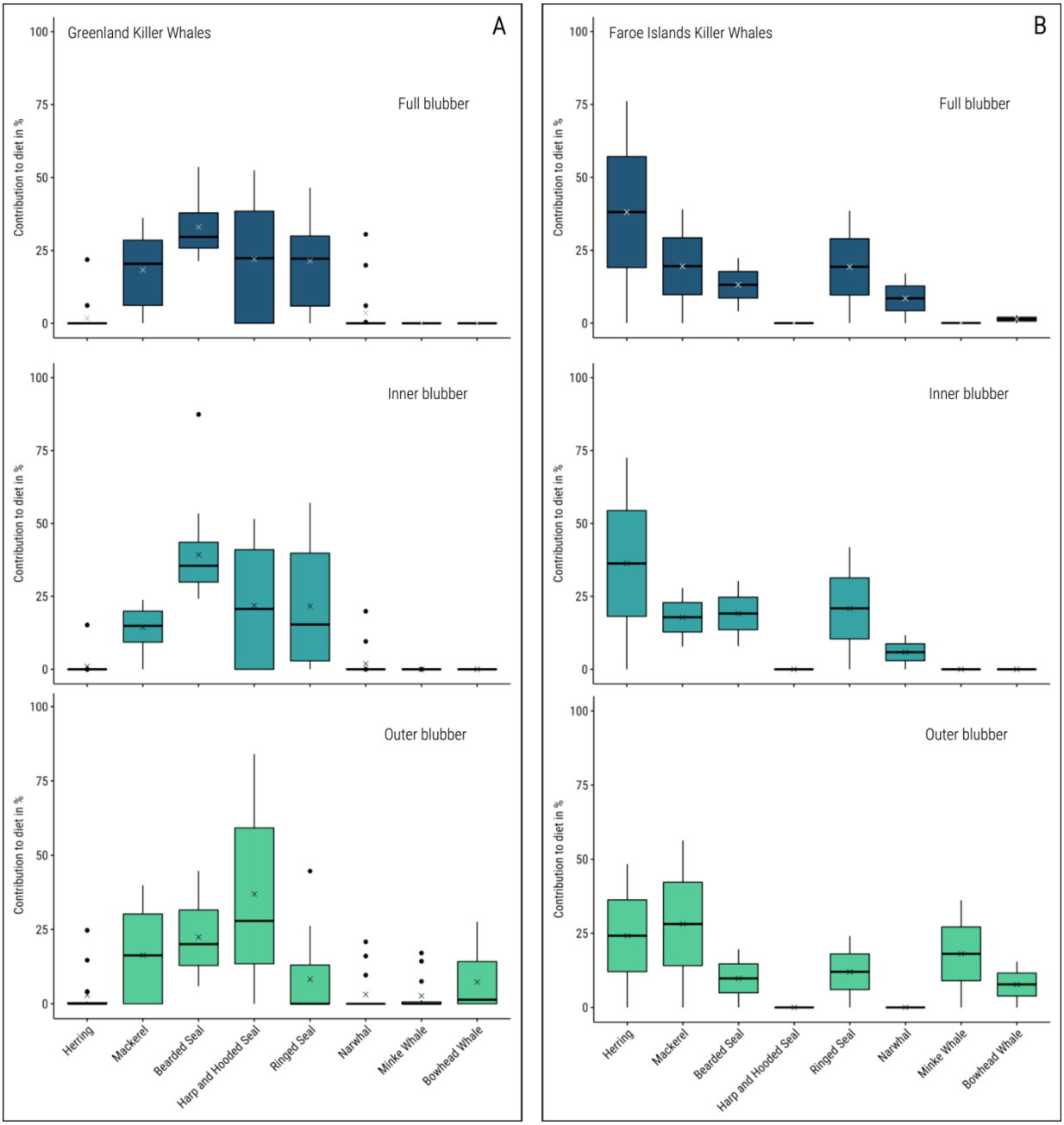

Figure 4-4: (A) Proportions of different prey species estimated in the diets of Greenland (n=16) and (B) Faroe Islands (n=2) killer whales based on the validated quantitative fatty acid signature analysis (QFASA) approach for killer whales. For outer layer, inner layer, and full depth FA signatures, outer layer, inner layer, and full depth calibration coefficients (CCs) were used, respectively. The estimates were lipid-corrected to account for differences in lipid between the prey items. The crosses show the mean, the thick bar shows the median, and the box extremities show the lower and upper quartiles.

Remote biopsies from free-ranging cetaceans typically only collect the outer blubber layers, e.g., layer 10 and likely layer 9, 8, 7 and/or 6. To test if the newly developed QFASA model for killer whales can accurately predict the diet of free-ranging individuals from biopsy samples, we ran QFASA on the outer blubber and layer 10 FA signatures using the outer blubber CCs or layer 10 CCs (four combinations total) and found the estimates were similar, and close to the diet estimates using full blubber FAs signatures and full blubber CCs (Fig. 4-5), with the four main prey species being harp and hooded seal, mackerel, bearded seal, and ringed seal.

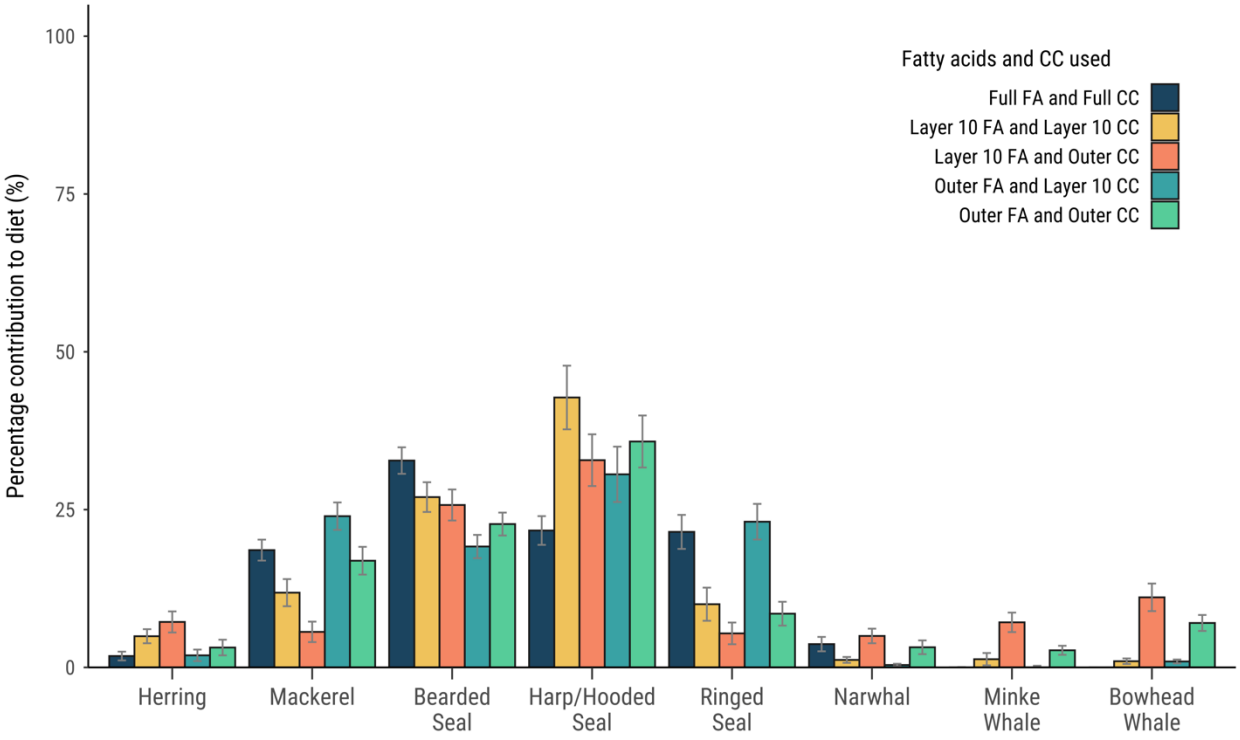

Figure 4-5:Diet estimates for Greenlandic killer whales (n=16) for full blubber FAs signatures and full blubber CCs, and four combinations of outer blubber and layer 10 (the outermost layer)

FA signatures and CCs. The simulated diets resulted in a similar percentage of prey species, with the four main species being harp and hooded seal, mackerel, bearded seal, and ringed seal.

4.5 DISCUSSION

Our results show that with the appropriate CCs, the diets of killer whales can be accurately estimated using QFASA. To use this approach, cetacean-specific CCs are essential; we demonstrated this for killer whales, but it is likely also the case for other cetaceans. A lack of CCs resulted in highly inaccurate estimates for the managed-care whales, based on their known diet. Mustelid and pinniped CCs were also unable to produce accurate estimates for the managed-care killer whales. These findings were perhaps not surprising, given differences in metabolic capacity between the order Artiodactyla (even-toed ungulates, which includes cetaceans) and Carnivora (which includes pinnipeds and mustelids) (Budge et al., 2006), as well as the high degree of stratification of FAs in cetacean blubber compared to mustelid adipose or pinniped blubber (Bourque et al., 2018; Iverson et al., 2004; Koopman, 2007).

We were able to calculate and validate CCs both for full depth blubber, as well as for inner and outer blubber. Our cross-validation simulations showed a high average accuracy using the full blubber CCs and full blubber FA signatures. Thus, when full blubber FA signatures are available, such as for stranded or harvested cetaceans (Krahn et al., 2004; Loseto et al., 2009), full blubber CCs should be used. For most free-ranging cetaceans, however, only small biopsies containing skin and partial (outer) blubber depth profiles are typically available. Due to FA stratification through blubber depths, we found that the full blubber CCs do not provide reliable diet estimates for partial depth blubber FA signatures. Critically, we were able to overcome this issue to accurately predict the diets of managed-care individuals by generating and applying layer-specific CCs. Using the layer ten (the outermost layer) or the outer blubber CCs, with

either the layer 10 or outer blubber FA signatures, produced similar dietary estimates in the free-ranging cetaceans, which could be explained by a low difference in the FA signatures within the outer layers (6-10) in these individuals (Bourque et al., 2018). Nonetheless, outer blubber CCs yielded more accurate estimates on the managed-care killer whales when applied to either outer blubber or layer 10 FA, and the outer blubber CCs account for potential small differences between the layers in the outer blubber. Therefore, we recommend using outer blubber CCs to estimate the diet of cetaceans from biopsy-derived FA signatures. Although layer 1 CCs did not perform as well as the inner blubber CCs on the inner blubber FA signatures, which can be explained by FA signature varying slightly from layer 1 to 4 (Fig. S4-1, Bourque et al. 2018), the diet estimates were accurate when using the average inner layers CCs on either layer 1 or inner blubber FA signatures. Therefore, if researchers focus on using inner blubber FA signatures to model recent blubber deposition for example, we recommend using the inner blubber CCs for more accurate dietary estimations.

In addition to validating QFASA for killer whales based on comparison for the managed-care killer whales to their true diet, further support for the approach comes from consistency in the diet estimates in free-ranging killer whales with stomach contents and available literature on their feeding habits. The QFASA method, using full, inner, and outer blubber, estimated harp/hooded seal as one of the top three diet items for Greenlandic killer whales which is corroborated by stomach contents. Stomach contents were reported for seven killer whales, and all included harp and/or hooded seal remains. Killer whales in West Greenland waters have also been reported to feed heavily on marine mammals based on visual observations (Heide-Jørgensen, 1988). Nonetheless, the LOPO analysis revealed that only 58% of the harp and hooded seal were identified correctly, with some overlap with ringed seals, thus the species-specific seal consumption estimates may be less robust than for the other prey items.

In addition to seals, Atlantic mackerel were also an important part of killer whale diets in Greenland killer whales; however, almost no predation was estimated on herring. Observations in Norway have shown an increasing association between killer whales that forage offshore and mackerel (Nikolioudakis et al., 2019; Nøttestad et al., 2014; Olafsdottir et al., 2019) and an increase in mackerel biomass has been reported in the Irminger Current, off Tasiilaq, where the Greenlandic killer whales were harvested (Jansen et al., 2016). Increases in mackerel in Greenland may be explained by the warming temperatures in the Arctic, particularly in East Greenland, that changed the migrating pattern of mackerel from the Norwegian and North Seas towards Greenland over the past decade (Jansen et al., 2016). Previous studies suggested that some populations of North Atlantic killer whales are strongly associated with mackerel stocks (Foote et al., 2012). Since killer whales are opportunistic hunters capable of switching prey (Remili et al., 2021), one could easily imagine killer whales in Greenland having a mixed diet of both mackerel and seal species.

Unlike seals and fish, almost no consumption of any whale species was estimated by QFASA, except for some consumption of narwhal (*Monodon monoceros)* (9.59-19.87%) for two individuals. One of these individuals was also the only killer whale to have whale reported in the stomach contents, although the identified species was minke whale (*Balaenoptera acutorostrata*). The only exception was for bowhead whale estimates in the outer blubber of the Greenlandic killer whales, which could indicate that bowheads are part of an occasional feeding. However, local reports off Tasiilaq suggested that killer whales occasionally prey on humpback whales or fin whales (Rosing-Asvid, pers. comm.). We attempted to include humpback whales in our analysis but had to drop it because their FA signatures overlapped with the other prey signatures (Supplementary text). The higher proportion of bowhead whales in the outer blubber could indicate occasional feeding on baleen whales in Eastern Greenland, although the estimates

are still quite low compared to the other species (like fish and seal). Thus, both short-term (stomach contents) and longer-term (QFASA) estimates were similar in suggesting a limited importance of whale species in the diets of these killer whales in Greenland. This feeding appears to differ from killer whales feeding in other Arctic environments; specifically, the killer whales in West Greenland-eastern Canadian Arctic are known to target narwhals and other whales including bowheads (*Balaena mysticetus*) (Ferguson et al., 2012a; Laidre et al., 2006; Willoughby et al., 2020).

The two killer whales from the Faroe Islands showed different diet estimates based on QFASA than the Greenland killer whales. One ate mainly herring according to QFASA, which aligns with reports stating that herring is the preferred prey of killer whales in the Faroe Islands (Bloch et al., 1988). The other had a high proportion of mackerel, but also ringed seal, and bearded seal in its diet estimates. Both Faroese whales were expected to have high proportions of fish in their diet estimates, given the lower concentrations of biomagnifying contaminants in their blubber, compared the Greenlandic whales (Pedro et al., 2017). The estimate of the two Arctic seal species in this whale's diet seems unlikely and may reflect another type of pinniped prey of some Faroese, Icelandic, and Norwegian killer whales, like grey or harbor seals (Bloch et al., 1988; Jourdain et al., 2020; Samarra et al., 2018). The prey library available for this study did not include grey and/or, and harbor seals, but future work could likely better identify seal species by including more appropriate additional seal prey in the library.

Future research may use QFASA on other cetacean species to shed light on inter-and intrapopulation dietary variations. Stratification indices (SI) which represent the differences in concentration of the main FA in the outer vs. inner blubber were calculated as the summed absolute values of the outer vs. inner blubber differences in the 16 main FA (Koopman, 2007). It seems likely that our QFASA approach could be applied to other cetaceans with stratification

indices similar to killer whales (SI = 31.43), which includes for example pilot whales (*Globicephala melas*), beluga whales, and narwhals (*Monodon monoceros*) (Koopman, 2007). To use QFASA on cetaceans with unknown stratification indices, samples obtained on stranded animals can be used to calculate the stratification index for the species. In case of a significant difference of stratification, the method would probably need to be fine-tuned by maybe using killer whale CCs from lower layers in the blubber.

This study demonstrates the utility of QFASA in estimating the diets of killer whales and likely other cetaceans, including biopsy samples; nonetheless, accurate diet estimates using this approach require certain conditions be met. In particular, QFASA relies on a complete prey library (Bromaghin et al., 2016). While our prey library contained prey species that are believed to be part of the Greenlandic killer whales' diets, some potentially important prey species could be missing from the library. For example, squid was occasionally previously reported in the whales' stomachs (Heide-Jørgensen, 1988), but was not included in the current library. The stomach contents of a killer whale harvested in September 2021 also included redfish (*Sebastes marinus*), Greenland halibut (*Reinhardtius hippoglossoides*), and Atlantic cod (*Gadus morhua*) (Rosing-Asvid, pers. comm.). Our results showing Arctic seals in the dietary estimates of one Faroese whale seem unrealistic and could have represented another species of pinniped(s). The high estimate for Arctic seals in this killer whale was likely the model's best attempt to estimate its diet using only prey signatures available to it. Thus, a carefully conceived prey library, based on prior feeding knowledge, is an important consideration. Additionally, while we used the FA signatures from the blubber of the marine mammal prey, other parts or the even whole bodies (Jefferson et al., 1991) may be consumed, which may have somewhat different FA signatures or lipid content than the full body lipid percentage we used. Finally, while previous studies have shown that fatty acids deposit in the inner blubber within a couple of weeks, we cannot yet

estimate how long it may take for dietary fatty acids to deposit within the outer blubber. Our managed-care individuals were fed a constant diet over many years, thus not enabling us to determine an exact time frame for outer blubber fatty acid deposition. With these key considerations in mind, this new QFASA approach should nevertheless provide important new insight into the feeding ecology of free-ranging killer whales and other cetaceans.

4.6 ACKNOWLEDGEMENTS

Thank you to Judy St Leger for sending the killer whale and fish samples from SeaWorld. Thank you to Erika Nilson, Alexandria Mena, and William Winhall for assistance with the SeaWorld sample shipments and information. Thanks to Kaliana Tom, Jennifer Bourque, and Sara Pedro for assistance with the fatty acid analysis. Thanks to the late Dorethe Bloch for providing samples from the two killer whales from the Faroese Islands. Thanks to Anna Roos for providing the Greenland minke whale samples. This work was funded by the Canada Research Chairs Program (to M.A.M., 950−232183), the Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grants Program (to M.A.M., RGPIN-2019−05330), and a Canada Foundation for Innovation Grant (to M.A.M., #37873). Additional earlier support came from the SeaWorld & Busch Gardens Conservation Fund (to M.A.M.) and a University of Connecticut, Institute of Biological Risk Summer Grants Program (to M.A.M.) Additional funds for sample collection in Greenland came from The Danish Cooperation for Environment in the Arctic (DANCEA) Programme (to R.D., C.S.). Thanks for the support in the form of a scholarship from the FRQNT EcotoQ Strategic Cluster and a Graduate Excellence Award from the Department of Natural Resource Sciences at McGill University (to A.R.).

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4.8 SUPPLEMENTARY INFORMATION

Additional Methods

Samples from managed-care and free-ranging killer whales and their prey

For the managed-care killer whales, we used existing FA signature data from archived blubber samples of four previously deceased (one in 2008 and three in 2010) individuals from SeaWorld (Bourque et al., 2018). The killer whale samples were frozen (−20 °C) after collection and during transport and stored at −80 °C from August 2015 until analysis in November 2015 through March 2016. Samples from these individuals were in the form of full depth blubber pieces with skin attached. The blubber was divided into ten equal-length pieces, from adjacent to muscle (layer 1) to adjacent to skin (layer 10). The managed-care killer whales were fed a constant diet consisting of roughly the same proportions of Pacific herring (*Clupea pallasii)*, capelin *(Mallotus villosus*), Pacific mackerel *(Scomber japonicus)*, and sockeye salmon *(Oncorhynchus nerka)*. To create the appropriate prey library, SeaWorld provided frozen whole fish samples from the supply they feed to the killer whales (n=10 herring, n=10 capelin, n=10 mackerel and n=4 salmon). Fish samples were received in May and June 2016 and analyzed in Sept 2016 through March 2017.

We used published FA signature data from 18 free-ranging killer whales, from Greenland (n =16) and the Faroe Islands (n = 2) (Bourque et al., 2018) to test the QFASA model on wild individuals. These samples were stored frozen (−20 °C) after collection in 2008-2014 and during transport and at −80 °C once received in May 2015. The samples were analyzed in Nov 2015 to July 2016. The full depth blubber pieces with skin attached were divided into 10 equal length sections, exactly as per the managed-care individuals. The prey FA library consisted of 535 samples from 9 different species: 55 bearded seals (*Erignathus barbatus*), 62 bowhead whales (*Balaena mysticetus*), 239 harp seals (*Pagophilus groenlandicus*), 32 hooded seals (*Cystophora*

cristata), 10 Atlantic herring (*Clupea harengus)*, 10 Atlantic mackerel (*Scomber scombrus*), 5 minke whales (*Balaenoptera acutorostrata*), 16 narwhals (*Monodon monoceros*) and 106 ringed seals (*Pusa hispida*). The bearded seal, harp seal, hooded seal, and ringed seal samples were from Greenland and the Davis strait and were published previously (McKinney et al., 2013; Thiemann et al., 2008a; Thiemann et al., 2008b). The bowhead were from Alaska, with FA data provided by Dr. Suzanne Budge (Budge et al., 2008). The Atlantic herring, Atlantic mackerel, narwhal and minke whale samples were from Greenland and the FA data were generated as part of the current study. Herring and mackerel were received with the Greenland killer whale samples and analyzed along with the SeaWorld fish from Sept 2016 to March 2017. Collection details for all managed-care and free-ranging killer whales and prey samples are provided in Table S4-1.

Table S4-1:List of sample and collection details for the managed-care killer whales and their prey, as well as the free-ranging killer whales and their prey.

Species	ID	Geography	Date	Paper originally published
Killer Whale	35143	East Greenland	Summer 2013	Bourque et al. 2018
Killer Whale	38340	East Greenland	Summer 2012	Bourque et al. 2018
Killer Whale	48335	East Greenland	Summer 2012	Bourque et al. 2018
Killer Whale	48336	East Greenland	Summer 2012	Bourque et al. 2018
Killer Whale	48337	East Greenland	Summer 2012	Bourque et al. 2018
Killer Whale	48338	East Greenland	Summer 2012	Bourque et al. 2018
Killer Whale	48339	East Greenland	Summer 2012	Bourque et al. 2018
Killer Whale	48732	East Greenland	Summer 2013	Bourque et al. 2018
Killer Whale	48733	East Greenland	Summer 2013	Bourque et al. 2018
Killer Whale	48735	East Greenland	Summer 2013	Bourque et al. 2018
Killer Whale	48736	East Greenland	Summer 2013	Bourque et al. 2018
Killer Whale	51601	East Greenland	Summer 2014	Bourque et al. 2018
Killer Whale	51606	East Greenland	Summer 2014	Bourque et al. 2018
Killer Whale	51607	East Greenland	Summer 2014	Bourque et al. 2018
Killer Whale	51610	East Greenland	Summer 2014	Bourque et al. 2018
Killer Whale	51613	East Greenland	Summer 2014	Bourque et al. 2018

Killer Whale	40888	Faroe Islands	Winter 2008	Bourque et al. 2018
Killer Whale	40889	Faroe Islands	Winter 2008	Bourque et al. 2018
Killer Whale	SW080429	SeaWorld	2008	Bourque et al. 2018
Killer Whale	SW100500	SeaWorld	2010	Bourque et al. 2018
Killer Whale	SW100743	SeaWorld	2010	Bourque et al. 2018
Killer Whale	SW100830	SeaWorld	2010	Bourque et al. 2018
Prey for the QFASA library on the managed-care whales				
Capelin (n=10)	SWCapelin-1-10	SeaWorld		This paper
Pacific Herring (n=10)	SWHerring-11-20	SeaWorld		This paper
Mackerel (n=10)	SWMackerel-21-30	SeaWorld		This paper
Sockeye Salmon (n=4)	SWSalmon-51-54	SeaWorld		This paper
Prey for the QFASA library on the free-ranging killer whales				
Atlantic Herring (n=10)	GLHerring-31-40	East Greenland		This paper
Atlantic Mackerel (n=10)	GLMackerel-41-50	East Greenland		This paper
Bearded Seal				
n=8	SGBS1-SGBS8	South Greenland		McKinney et al. 2013
n=19	SIP107-SIP317	Davis Strait		Thiemann et al. 2008
n=28	UMP197-UMP480	Davis Strait		Thiemann et al. 2008
Harp Seal				
n=135	SIP001-SIP283	Davis Strait		Thiemann et al. 2008
n=104	Tucker001-Tucker 114	Davis Strait		Thiemann et al. 2008
Hooded Seal				
n=17	SIP098-SIP272	Davis Strait		Thiemann et al. 2008
n=15	Tucker115-Tucker133	Davis Strait		Thiemann et al. 2008
Ringed Seal				
n=50	EGRS24931-EGRS34946	East Greenland		Mckinney et al. 2013
n=54	SIP227-SIP319	Davis Strait		Thiemann et al. 2008
n=28	UMP332-333	Davis Strait		Thiemann et al. 2008
Narwhal (n=16)	53801-53846	East Greenland		Unpublished
Bowhead Whale (n=62)	01KK3-01SA1	Alaska	1997-2002	Budge et al. 2008
Minke Whale (n=5)	Ba_001-2017/0001	East Greenland	2000-2017	Unpublished

Fatty acid analyses

FAs were extracted and quantified as described in previous studies (Budge et al., 2006; McKinney et al., 2013). In brief, for marine mammals, lipids were extracted from blubber using the Folch method (Folch et al., 1957). Whole fish were first homogenized in a food processor, extracted via a modified Folch and filtered (Budge et al 2006). All marine mammal and fish FA extracts were *trans*-esterified using the Hilditch reagent to produce fatty acid methyl esters (FAMEs). The FAMEs were separated, identified, and the mass percentage of each of 69 FAs was quantified by gas chromatography on an Agilent 8860 system (Santa Clara, CA, USA) with flame ionization detection (GC-FID). Each FA is named using the nomenclature x:y*n*z, where x is the length of the carbon chain, y is the number of double bonds, and z is the position of the first double bond from the methyl ('n') end of the chain. As Bourque et al. (2018) found no statistically significant differences between layers 1-4 and between layers 6-10 for the killer whale samples, throughout the current study, "full blubber FA" refers to the lipid-weighted average of all 10 layers, while "inner blubber FA" refers to the lipid-weighted average of layers 1-4 and "outer blubber FA" refers to the lipid-weighted average of layers 6-10.

Quality Control:

Quality control, as previously described (Bourque et al., 2018), included the extraction and analysis of a standard reference material, SRM1945 pilot whale blubber, from the US National Institute of Standards and Technology (NIST), with each batch of 11 samples. The SRM was run 16 times, and the relative standard deviation of the FA values averaged 16% compared to the published 27 individual FA values (Bourque et al., 2018; Kucklick et al., 2010). All fish samples were extracted and analyzed in duplicate. The average FA values of duplicates was used, and the percent difference of the retained duplicates averaged 20%. A 18 FAME mixed standard (68B; Nu-Chek Prep, Elysian, MN, USA) was run for additional quality control for the fish samples; the average relative error was 5.6% (n = 5).

Table S4-2:FA percentages (mean ± SE) in the managed-care killer whales (n=4) and their prey items. Only FA above 0.1% are shown. The prey species were: Pacific herring (Clupea pallasii), capelin (Mallotus villosus), Pacific mackerel (Scomber japonicus), and sockeye salmon (Oncorhynchus nerka). Italicized fatty acids are the extended dietary set and bold fatty acids are the dietary set. Normal font represents non-dietary fatty acids (which were not included in the QFASA analyses).

	Capelin (n=10)	Pacific herring (n=10)	Pacific mackerel (n=10)	Sockeye salmon (n=4)	Killer whale (n=4)
Saturated FA					
14:0	3.21 ± 0.29	4.75 ± 0.23	1.93 ± 0.12	2.08 ± 0.06	6.01 ± 0.24
iso15:0	0.06 ± 0.01	0.14 ± 0.01	0.13 ± 0.01	0.12 ± 0.01	0.45 ± 0.05
15:0	0.20 ± 0.01	0.41 ± 0.02	0.63 ± 0.03	0.33 ± 0.02	0.69 ± 0.09
16:0	15.47 ± 0.99	19.02 ± 0.75	15.77 ± 0.17	12.08 ± 0.39	6.39 ± 0.36
17:0	0.09 ± 0.01	0.22 ± 0.01	0.74 ± 0.01	0.24 ± 0.01	0.14 ± 0.02
18:0	1.89 ± 0.18	2.59 ± 0.11	7.80 ± 0.19	3.59 ± 0.16	1.29 ± 0.15
∑SFA	20.93 ± 0.12	27.13 ± 0.09	27.01 ± 0.03	18.44 ± 0.07	14.97 ± 0.06
Mono-Unsaturated FA					
16:1n11	0.29 ± 0.02	0.44 ± 0.02	0.59 ± 0.02	0.34 ± 0.03	1.57 ± 0.20
16:1n9	0.17 ± 0.01	0.16 ± 0.01	0.35 ± 0.02	0.38 ± 0.04	1.58 ± 0.26
16:1n7	5.01 ± 0.36	6.84 ± 0.18	2.31 ± 0.12	3.27 ± 0.33	13.33 ± 0.35
7Me16:0	0.30 ± 0.01	0.23 ± 0.03	0.16 ± 0.01	0.30 ± 0.04	0.24 ± 0.01
16:2n4	0.33 ± 0.02	0.23 ± 0.03	0.75 ± 0.03	0.32 ± 0.06	0.28 ± 0.02
16:3n6	0.22 ± 0.02	0.77 ± 0.06	0.07 ± <0.01	0.10 ± 0.02	0.53 ± 0.03
17:1	0.06 ± <0.01	0.28 ± 0.01	0.28 ± 0.01	0.31 ± 0.04	0.40 ± 0.02
16:4n3	0.07 ± 0.01	0.11 ± <0.01	0.16 ± 0.01	0.19 ± 0.01	0.11 ± 0.01
16:4n1	0.21 ± 0.03	0.68 ± 0.08	0.05 ± <0.01	0.04 ± 0.01	0.20 ± 0.02
18:1n11	0.77 ± 0.02	0.29 ± 0.05	0.06 ± 0.01	1.34 ± 0.18	7.29 ± 0.60
18:1n9	5.13 ± 0.19	15.83 ± 1.66	6.51 ± 0.19	16.04 ± 0.83	17.74 ± 0.63
18:1n7	2.67 ± 0.12	4.67 ± 0.31	2.82 ± 0.04	2.92 ± 0.33	2.38 ± 0.14
18:1n5	0.57 ± 0.02	0.32 ± 0.06	0.13 ± 0.01	0.61 ± 0.05	0.36 ± 0.02
20:1n11	0.74 ± 0.07	1.86 ± 0.41	0.20 ± 0.05	4.79 ± 1.28	4.63 ± 0.43
20:1n9	9.85 ± 1.11	3.71 ± 0.63	0.57 ± 0.06	2.13 ± 0.38	6.37 ± 0.28
20:1n7	1.00 ± 0.13	0.45 ± 0.03	0.23 ± 0.03	0.31 ± 0.10	0.30 ± 0.02
22:1n11	9.84 ± 1.42	6.73 ± 1.17	0.20 ± 0.04	3.87 ± 1.45	5.46 ± 0.13
22:1n9	1.30 ± 0.17	0.44 ± 0.05	0.19 ± 0.03	0.43 ± 0.17	0.52 ± 0.02
24:1n9	0.75 ± 0.02	0.55 ± 0.07	0.50 ± 0.03	0.47 ± 0.14	0.18 ± 0.01
∑MUFA	39.29 ± 0.12	44.58 ± 0.14	16.14 ± 0.01	38.17 ± 0.21	63.46 ± 0.10
Poly-Unsaturated FA					

18:2n6	0.82 ± 0.02	0.89 ± 0.05	1.56 ± 0.07	1.46 ± 0.09	2.33 ± 0.89
18:3n6	0.01 ± <0.01	0.05 ± <0.01	0.28 ± 0.01	0.03 ± <0.01	0.10 ± 0.01
18:3n3	0.27 ± 0.02	0.51 ± 0.03	1.42 ± 0.09	0.88 ± 0.06	3.59 ± 0.72
18:4n3	0.55 ± 0.03	0.83 ± 0.05	1.95 ± 0.16	0.83 ± 0.07	0.54 ± 0.04
18:4n1	0.16 ± 0.01	0.14 ± 0.01	0.01 ± <0.01	0.11 ± 0.03	0.16 ± 0.02
20:2n6	0.21 ± 0.05	0.19 ± 0.04	0.60 ± 0.06	0.34 ± 0.03	0.13 ± 0.01
20:4n6	0.52 ± 0.03	0.95 ± 0.04	2.78 ± 0.13	1.28 ± 0.09	0.24 ± 0.04
20:3n3	0.09 ± 0.02	0.10 ± 0.01	0.28 ± 0.02	0.16 ± 0.01	0.10 ± 0.03
20:4n3	0.35 ± 0.01	0.34 ± 0.01	0.58 ± 0.02	1.49 ± 0.24	0.34 ± 0.04
20:5n3	13.34 ± 0.78	11.35 ± 0.29	9.65 ± 0.28	12.12 ± 1.87	2.25 ± 0.29
21:5n3	0.34 ± 0.02	0.35 ± 0.02	0.22 ± 0.01	0.26 ± 0.03	0.14 ± 0.02
22:4n6	0.03 ± 0.01	0.09 ± 0.01	0.40 ± 0.03	0.12 ± 0.01	0.12 ± 0.02
22:5n3	1.79 ± 0.13	1.03 ± 0.06	1.78 ± 0.06	4.14 ± 0.57	1.53 ± 0.30
22:6n3	19.83 ± 1.42	9.66 ± 0.41	32.73 ± 0.67	18.58 ± 0.67	3.87 ± 0.59
∑PUFA	38.30 ± 0.13	26.47 ± 0.04	54.25 ± 0.06	41.80 ± 0.25	15.44 ± 0.15
∑21 Dietary	60.07 ± 5.39	40.41 ± 3.45	54.90 ± 1.81	49.84 ± 6.66	32.31 ± 3.68
∑30 Dietary extended	95.34 ± 1.64	95.36 ± 1.55	94.57 ± 2.08	94.22 ± 2.46	81.11 ± 2.06

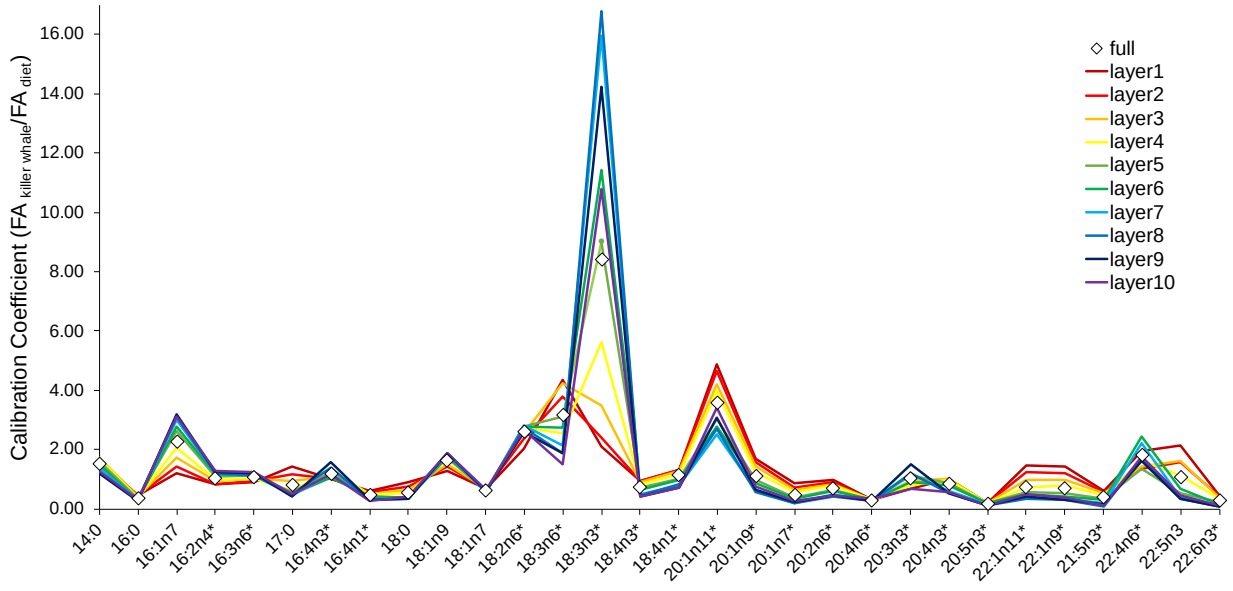

Figure S4-1:Blubber depth variation in the fatty acid calibration coefficients calculated from the managed-care killer whales and their known diet items for the dietary (with asterisk) and

extended fatty acids above 0.1% of total FA. Layer 1 is closest to muscle, while layer 10 is closest to skin.

Table S4-3:Calibration coefficients used in the QFASA simulations. Killer whale CCs were calculated for the managed-care killer whales from this study. CC shown here are for the dietary (bold) and extended dietary FA that were above 0.1% total FA. Layer 1 is closest to muscle, while layer 10 is closest to skin.

Fatty Acid	Full blubber	Layer 1	Inner blubber (Layers 1– 4)	Outer blubber (Layers 6–10)	Layer 10
14:0	1.516	1.535	1.654	1.339	1.300
16:0	0.377	0.460	0.423	0.310	0.319
16:1n7	2.261	1.210	1.607	3.034	3.104
16:2n4	1.034	0.843	0.904	1.219	1.264
16:3n6	1.067	0.908	0.976	1.176	1.234
17:0	0.807	1.423	1.032	0.476	0.499
16:4n3	1.177	1.025	1.115	1.279	1.171
16:4n1	0.463	0.595	0.542	0.336	0.267
18:0	0.551	0.898	0.706	0.366	0.420
18:1n9	1.646	1.293	1.484	1.844	1.875
18:1n7	0.644	0.685	0.677	0.601	0.628
18:2n6	2.623	2.025	2.418	2.722	2.682
18:3n6	3.180	4.355	3.742	2.038	1.516
18:3n3	8.398	2.113	3.411	13.831	10.771
18:4n3	0.752	0.965	0.907	0.490	0.406
18:4n1	1.143	1.330	1.280	0.810	0.729
20:1n11	3.571	4.860	4.428	2.895	3.416
20:1n9	1.111	1.713	1.502	0.692	0.747
20:1n7	0.481	0.873	0.702	0.246	0.265
20:2n6	0.716	0.993	0.886	0.481	0.477
20:4n6	0.307	0.323	0.308	0.292	0.349
20:3n3	1.034	0.670	0.841	1.091	0.667
20:4n3	0.871	1.003	1.000	0.609	0.574
20:5n3	0.187	0.200	0.201	0.137	0.125
22:1n11	0.753	1.465	1.106	0.423	0.507
22:1n9	0.718	1.445	1.107	0.354	0.393
21:5n3	0.399	0.620	0.545	0.180	0.130
22:4n6	1.841	1.940	1.590	1.954	1.787
22:5n3	1.073	2.140	1.614	0.444	0.450

22:6n3	0.280	0.468	0.408	0.118	0.118

Table S4-4:Leave-one-prey-out (LOPO) simulations on the prey library of the managed-care killer whales to establish a potential overlap of prey signatures. Numbers in bold represent the percentage of prey correctly identified.

Dietary FA (21) Aitchison distance				
	Capelin	Herring	Mackerel	Salmon
Capelin	**94.63%**	2.20%	0.32%	2.85%
Herring	6.57%	**84.70%**	3.73%	5.01%
Mackerel	0.72%	0.18%	**98.78%**	0.33%
Salmon	6.30%	2.22%	4.63%	**86.85%**

Dietary FA (21) KL distance				
	Capelin	Herring	Mackerel	Salmon
Capelin	**83.85%**	4.20%	11.65%	0.30%
Herring	11.10%	**82.88%**	3.45%	2.56%
Mackerel	0.52%	0.06%	**98.90%**	0.51%
Salmon	4.95%	8.15%	14.78%	**72.12%**

Dietary Extended FA (30) Aitchison Distance				
	Capelin	Herring	Mackerel	Salmon
Capelin	**94.12%**	2.23%	0.23%	3.42%
Herring	7.18%	**86.36%**	3.23%	3.22%
Mackerel	0.71%	0.19%	**98.78%**	0.32%
Salmon	5.90%	2.01%	4.31%	**87.78%**

Dietary Extended FA (30) KL Distance				
	Capelin	Herring	Mackerel	Salmon
Capelin	**85.88%**	3.03%	10.08%	1.01%
Herring	13.25%	**82.32%**	2.05%	2.38%
Mackerel	0.46%	0.15%	**98.95%**	0.43%
Salmon	3.75%	5.38%	9.27%	**81.60%**

Table S4-5:QFASA diet estimates for managed care killer whales (n=4) on the full blubber FA signatures using no CC. The actual diet of the managed care killer whales consisted of 32% capelin, 60% herring, 4% mackerel and 4% salmon.

Full Blubber - No CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	0.00	0.00	0.00	0.00	<0.01 ± 0.29	100.00	91.67
Herring	100.00	100.00	100.00	100.00	100 ± 4.11	66.67	
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± <0.01	100.00	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 4.10	100.00	
Full Blubber - No CCs - Dietary FA (21) - Aitchison distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	7.41	7.14	10.00	6.35	7.73 ± 4.85	75.86	182.50
Herring	52.90	69.82	49.19	61.00	58.23 ± 11.45	2.96	
Mackerel	11.53	4.87	7.00	10.80	8.55 ± 2.18	113.74	

Salmon	28.16	18.17	33.81	21.85	25.50 ± 9.72	537.46	

Table S4-6:QFASA diet estimates for managed care killer whales (n=4) using marine-fed mink and grey seal CC. Simulations resulted in inaccurate estimates compared to the killer whales' actual diet consisting of 32% capelin, 60% herring, 4% mackerel and 4% salmon. The mean and SE were bootstrapped 100 times.

Full Blubber - Marine-fed Mink CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	4.30	15.79	19.25	1.05	10.10 ± 5.81	68.44	
Herring	95.70	84.21	80.62	98.95	89.87 ± 6.12	49.78	79.35
Mackerel	0.00	0.00	0.13	0.00	0.03 ± 1.28	99.18	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 0.41	100.00	
Full Blubber - Marine-fed Mink CCs - Dietary FA (21) - Aitchison distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	4.41	2.52	5.69	2.72	3.83 ± 4.65	88.02	
Herring	68.85	85.83	63.83	79.32	74.46 ± 8.32	24.10	113.70
Mackerel	14.29	4.78	7.16	13.12	9.83 ± 2.77	145.95	
Salmon	12.46	6.87	23.32	4.83	11.87 ± 5.28	196.75	
Full Blubber - Gray seal CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	0.09	12.69	18.87	0.00	7.91 ± 6.72	75.27	
Herring	99.91	87.31	81.13	100.00	92.09 ± 7.30	53.48	82.19
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± <0.01	100.00	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 1.30	100.00	
Full Blubber - Gray seal CCs - Dietary FA (21) - Aitchison distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	31.38	30.23	33.26	26.97	30.46 ± 6.93	4.81	
Herring	58.20	66.08	60.42	64.28	62.25 ± 8.70	3.75	47.72
Mackerel	10.42	3.69	6.32	8.75	7.29 ± 2.38	82.31	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 1.36	100.00	
Inner Blubber - Marine-fed Mink CCs - Dietary FA (21) - KL distance							

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	29.80	40.63	35.87	38.32	36.16	12.99	
Herring	70.20	59.37	64.13	61.68	63.84	6.41	54.85
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± 0.17	100.00	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 1.04	100.00	

Inner Blubber - Marine-fed Mink CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	25.44	15.53	13.51	23.40	19.47 ± 6.60	39.15	
Herring	50.73	76.50	60.86	67.18	63.82 ± 8.20	6.36	65.83
Mackerel	12.77	4.89	6.42	6.42	7.62 ± 2.04	90.61	
Salmon	11.06	3.08	19.21	3.01	9.09 ± 3.83	127.21	

Inner Blubber - Marine-fed Mink CCs - Dietary FA (21) - KL distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	29.80	40.63	35.87	38.32	36.16	12.99	
Herring	70.20	59.37	64.13	61.68	63.84	6.41	54.85
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± 0.17	100.00	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 1.04	100.00	

Inner Blubber - Marine-fed Mink CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	25.44	15.53	13.51	23.40	19.47 ± 6.60	39.15	
Herring	50.73	76.50	60.86	67.18	63.82 ± 8.20	6.36	65.83
Mackerel	12.77	4.89	6.42	6.42	7.62 ± 2.04	90.61	
Salmon	11.06	3.08	19.21	3.01	9.09 ± 3.83	127.21	

Inner Blubber - Gray seal CCs - Dietary FA (21) - KL distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	25.33	39.49	37.73	33.58	34.03 ± 10.38	6.35	
Herring	74.67	60.51	61.80	66.42	65.85 ± 11.98	9.75	53.29
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± <0.01	100.00	
Salmon	0.00	0.00	0.47	0.00	<0.01 ± 3.62	97.06	

Inner Blubber - Gray seal CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	47.48	41.22	40.43	44.08	43.30 ± 7.13	35.33	47.07

Herring	44.03	54.95	54.16	51.52	51.17 ± 8.87	14.72	
Mackerel	8.49	3.83	5.41	4.39	5.53 ± 2.00	38.24	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± <0.01	100.00	
Outer Blubber - Marine-fed Mink CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	0.00	0.00	0.00	0.00	<0.01 ± 0.34	100.00	
Herring	100.00	100.00	100.00	100.00	100 ± 1.42	66.67	91.67
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± 1.29	100.00	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± <0.01	100.00	
Outer Blubber - Marine-fed Mink CCs - Dietary FA (21) - Aitchison distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	0.00	0.00	0.00	0.00	<0.01 ± 0.42	100.00	
Herring	70.12	78.68	63.62	73.89	71.58 ± 5.95	19.30	157.47
Mackerel	17.78	7.38	10.25	22.28	14.42 ± 3.64	260.49	
Salmon	12.11	13.94	26.13	3.83	14.00 ± 7.07	250.08	
Outer Blubber - Gray seal CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	0.00	0.00	0.00	0.00	<0.01 ± 0.45	100.00	
Herring	100.00	100.00	100.00	100.00	100 ± 0.45	66.67	91.67
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± <0.01	100.00	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 0.03	100.00	
Outer Blubber - Gray seal CCs - Dietary FA (21) - Aitchison distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	18.09	11.23	14.53	10.10	13.49 ± 6.954	57.85	
Herring	70.73	86.65	78.98	78.86	78.81 ± 9.02	31.35	70.45
Mackerel	11.18	2.12	6.48	11.04	7.70 ± 2.84	92.60	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 0.95	100.00	

Table S4-7: Two-by-two comparison of the diet estimates (in %) using the FA from two managed-care killer whales and the calibration coefficients (CCs) from the other two remaining killer whales (n=4 total) on the inner and outer blubber FA signatures and the inner and outer blubber CCs.

Full Blubber - Full CCs - Dietary (21) Fas - KL Distance

Fatty acids used	CC used	Capelin mean	Capelin SE	Herring mean	Herring SE	Mackerel mean	Mackerel SE	Salmon mean	Salmon SE
SW080429 + SW100830	SW100500 + SW100743	24.99	5.87	72.15	6.37	2.85	2.78	0.00	1.21
SW100500 + SW100743	SW080429 + SW100830	52.60	5.47	38.35	8.97	0.00	0.19	9.05	5.19
SW080429 + SW100500	SW100830 + SW100743	34.22	6.86	65.70	6.94	0.07	2.07	0.00	0.35
SW100830 + SW100743	SW080429 + SW100500	33.23	9.95	53.16	13.24	3.84	2.80	9.78	6.28
SW080429 + SW100743	SW100830 + SW100500	37.25	8.25	56.48	10.68	0.55	1.80	5.73	4.36
SW100830 + SW100500	SW080429 + SW100743	29.00	7.56	68.24	6.89	2.76	2.61	0.00	2.17
Actual diet		32.00	–	60.00	–	4.00	–	4.00	–
Average simulations		35.21	7.33	59.01	8.85	1.68	2.04	4.09	3.26
%Error		10.04		1.64		58.03		2.31	
Total % Error		18.01							

Full Blubber - Full CCs - Dietary (21) Fas - Aitchison Distance									
Fatty acids used	CC used	Capelin mean	Capelin SE	Herring mean	Herring SE	Mackerel mean	Mackerel SE	Salmon mean	Salmon SE
SW080429 + SW100830	SW100500 + SW100743	37.36	3.21	60.29	3.62	0.00	0.18	2.35	2.36
SW100500 + SW100743	SW080429 + SW100830	34.55	3.66	58.48	5.57	2.90	1.26	4.07	2.65
SW080429 + SW100500	SW100830 + SW100743	33.67	4.16	57.65	6.28	1.75	1.34	6.94	4.09
SW100830 + SW100743	SW080429 + SW100500	38.77	3.37	58.30	4.13	1.27	1.17	1.66	2.20
SW080429 + SW100743	SW100830 + SW100500	32.76	3.26	66.05	2.73	1.19	0.91	0.00	0.53
SW100830 + SW100500	SW080429 + SW100743	35.94	3.44	59.62	4.13	1.28	1.13	3.16	2.27
Actual diet		32.00	–	60.00	–	4.00	–	4.00	–
Average simulations		35.51	3.52	60.06	4.41	1.40	1.00	3.03	2.35
% Error		10.96		0.11		65.04		24.24	
Total % Error		25.09							

Inner Blubber - Inner CCs - Dietary (21) Fas - KL Distance									
Fatty acids used	CC used	Capelin mean	Capelin SE	Herring mean	Herring SE	Mackerel mean	Mackerel SE	Salmon mean	Salmon SE

SW080429 + SW100830	SW100500 + SW100743	32.95	5.54	65.97	5.60	1.08	2.17	0.00	1.54
SW100500 + SW100743	SW080429 + SW100830	38.41	5.72	49.05	5.95	5.56	3.40	6.98	5.49
SW080429 + SW100500	SW100830 + SW100743	33.44	7.00	62.75	6.47	3.82	3.26	0.00	0.75
SW100830 + SW100743	SW080429 + SW100500	39.67	5.71	46.38	7.94	2.58	2.42	11.37	5.40
SW080429 + SW100743	SW100830 + SW100500	27.56	5.52	58.20	6.77	10.13	4.07	4.11	4.39
SW100830 + SW100500	SW080429 + SW100743	44.18	4.55	54.93	6.29	0.00	0.78	0.89	2.97
Actual diet		32.00	–	60.00	–	4.00	–	4.00	–
Average simulations		36.04	5.67	56.21	6.50	3.86	2.68	3.89	3.42
%Error		12.61		6.31		3.45		2.71	
Total % Error		6.27							

Inner Blubber - Inner CCs - Dietary (21) Fas - Aitchison Distance

Fatty acids used	CC used	Capelin mean	Capelin SE	Herring mean	Herring SE	Mackerel mean	Mackerel SE	Salmon mean	Salmon SE
SW080429 + SW100830	SW100500 + SW100743	43.77	2.79	50.49	5.87	2.35	1.43	3.38	3.20
SW100500 + SW100743	SW080429 + SW100830	31.93	5.27	62.04	4.70	0.00	0.15	6.03	5.17
SW080429 + SW100500	SW100830 + SW100743	39.54	3.93	55.80	6.43	2.03	1.65	2.63	2.51
SW100830 + SW100743	SW080429 + SW100500	35.70	3.64	57.25	4.27	0.00	0.36	7.04	4.03
SW080429 + SW100743	SW100830 + SW100500	34.99	4.46	46.30	5.27	2.21	1.41	16.50	3.43
SW100830 + SW100500	SW080429 + SW100743	38.55	2.49	61.45	2.55	0.00	0.13	0.00	0.37
Actual diet		32.00	–	60.00	–	4.00	–	4.00	–
Average simulations		36.14	3.96	56.57	4.65	0.85	0.74	6.44	3.10
%Error		12.95		5.72		78.82		61.01	
Total % Error		39.62							

Outer Blubber - Outer CCs - Dietary (21) Fas - KL Distance

Fatty acids used	CC used	Capelin mean	Capelin SE	Herring mean	Herring SE	Mackerel mean	Mackerel SE	Salmon mean	Salmon SE
SW080429 + SW100830	SW100500 + SW100743	40.05	10.10	51.28	6.48	8.67	5.13	0.00	1.01

SW100500 + SW100743	SW080429 + SW100830	20.73	3.80	78.02	4.78	0.00	0.00	1.25	3.19
SW080429 + SW100500	SW100830 + SW100743	59.18	9.17	40.82	9.18	0.00	0.00	0.00	0.10
SW100830 + SW100743	SW080429 + SW100500	23.11	6.22	60.47	6.68	11.15	5.63	5.27	5.79
SW080429 + SW100743	SW100830 + SW100500	57.71	8.37	37.83	8.05	0.00	0.00	4.46	3.82
SW100830 + SW100500	SW080429 + SW100743	22.95	6.33	69.02	6.75	8.03	4.97	0.00	1.46
Actual diet		32.00	–	60.00	–	4.00	–	4.00	–
Average simulations		37.29	7.33	56.24	6.99	4.64	2.62	1.83	2.56
%Error		16.53		6.27		16.03		54.25	
Total % Error		23.27							

Outer Blubber - Outer CCs - Dietary (21) Fas - Aitchison Distance									
Fatty acids used	CC used	Capelin mean	Capelin SE	Herring mean	Herring SE	Mackerel mean	Mackerel SE	Salmon mean	Salmon SE
SW080429 + SW100830	SW100500 + SW100743	37.39	7.19	54.12	7.78	8.49	1.45	0.00	1.23
SW100500 + SW100743	SW080429 + SW100830	54.53	4.39	45.47	4.71	0.00	0.00	0.00	2.06
SW080429 + SW100500	SW100830 + SW100743	50.57	3.72	47.83	4.63	1.60	1.18	0.00	1.29
SW100830 + SW100743	SW080429 + SW100500	29.77	3.40	60.93	5.02	3.90	2.14	5.40	3.99
SW080429 + SW100743	SW100830 + SW100500	48.27	4.41	43.62	4.15	3.01	1.91	5.09	3.92
SW100830 + SW100500	SW080429 + SW100743	32.42	5.03	65.64	3.79	1.94	1.53	0.00	0.68
Actual diet		32.00	–	60.00	–	4.00	–	4.00	–
Average simulations		42.16	4.69	52.94	5.01	3.16	1.37	1.75	2.19
%Error		31.74		11.77		21.05		56.26	
Total % Error		30.21							

Table S4-8:QFASA diet estimates for managed care killer whales (n=4) layer-specific CC derived from the same managed care killer whales (average of the four individual CCs) with the Aitchison distance. Diet estimates were more consistent and accurate with the Aitchison distance compared to the KL distance. The managed care killer whales' actual diet consisted of 32% capelin, 60% herring, 4% mackerel and 4% salmon. The mean and SE were bootstrapped 100 times. Layer 1 is closest to muscle, while layer 10 is closest to skin.

Full Blubber - Full CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	35.86	39.94	40.01	30.72	36.63 ± 3.58	14.48	
Herring	55.81	60.06	51.32	65.61	58.20 ± 4.10	3.00	22.07
Mackerel	2.85	0.00	0.00	2.54	1.35 ± 0.87	66.32	
Salmon	5.48	0.00	8.67	1.13	3.58 ± 2.45	4.47	

Layer 1 blubber - Layer 1 blubber CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	47.62	35.21	30.23	45.57	39.66 ± 3.89	23.93	
Herring	41.52	64.79	56.14	52.16	53.65 ± 4.69	10.58	37.67
Mackerel	1.70	0.00	2.39	0.00	1.01 ± 0.69	74.46	
Salmon	9.16	0.00	11.25	2.27	5.67 ± 2.73	41.72	

Layer 1 blubber - Inner blubber CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	54.79	41.54	39.50	50.46	46.57 ± 3.31	45.54	
Herring	38.70	58.28	51.99	49.54	49.63 ± 4.05	17.29	41.94
Mackerel	2.53	0.19	3.28	0.00	1.50 ± 0.83	62.55	
Salmon	3.99	0.00	5.23	0.00	2.30 ± 1.89	42.39	

Inner blubber - Inner blubber CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	41.19	36.49	32.32	39.99	37.50 ± 2.88	17.18	
Herring	48.60	63.51	55.35	59.71	56.79 ± 3.91	5.35	29.83
Mackerel	3.14	0.00	0.23	0.30	0.92 ± 0.88	77.01	
Salmon	7.07	0.00	12.10	0.00	4.79 ± 2.96	19.79	

Inner blubber - layer 1 CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	32.15	29.14	23.76	33.61	29.66 ± 3.30	7.30	
Herring	51.89	67.70	57.02	62.66	59.82 ± 4.47	0.30	61.17
Mackerel	2.07	0.00	0.00	0.00	0.52 ± 0.73	87.07	
Salmon	13.89	3.16	19.22	3.73	10.00 ± 4.37	149.99	

Outer layer blubber - Full CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	17.83	17.20	16.43	8.23	14.92 ± 3.35	53.36	
Herring	69.69	77.43	66.98	82.26	74.09 ± 4.79	23.48	
Mackerel	3.84	0.00	0.00	5.70	2.38 ± 1.52	40.40	58.07
Salmon	8.64	5.36	16.59	3.81	8.60 ± 3.65	115.03	

Outer layer blubber - Outer layer CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	46.75	41.80	40.05	31.79	40.10 ± 3.90	25.30	
Herring	48.45	58.20	52.74	63.61	55.75 ± 4.01	7.08	
Mackerel	4.07	0.00	0.00	4.60	2.17 ± 1.20	45.77	32.13
Salmon	0.73	0.00	7.21	0.00	1.98 ± 1.96	50.39	

Outer layer blubber - layer 10 CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	41.03	34.09	34.62	26.07	33.95 ± 4.30	6.10	
Herring	52.27	65.91	57.84	66.48	60.62 ± 4.45	1.04	
Mackerel	6.70	0.00	1.83	7.45	4.00 ± 1.80	0.12	17.89
Salmon	0.00	0.00	5.71	0.00	1.43 ± 1.78	64.30	

Layer 10 blubber - layer 10 CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429-10	Killer whale - SW100500-10	Killer whale - SW100743-10	Killer whale - SW100830-10	Mean ± SE	% Error	Total % Error
Capelin	41.43	55.69	38.62	21.75	39.37 ± 6.46	23.04	
Herring	57.26	42.59	50.42	66.65	54.23 ± 5.53	9.61	
Mackerel	1.31	0.00	6.56	0.00	1.97 ± 1.41	50.85	23.55
Salmon	0.00	1.72	4.40	11.60	4.43 ± 2.64	10.72	

Layer 10 blubber - Outer layer CCs - Dietary FA (21) - Aitchison distance

	Killer whale - SW080429-10	Killer whale - SW100500-10	Killer whale - SW100743-10	Killer whale - SW100830-10	Mean ± SE	% Error	Total % Error
Capelin	46.57	63.34	43.71	29.49	45.78 ± 6.39	43.06	
Herring	52.83	34.14	45.13	57.45	47.39 ± 5.89	21.02	
Mackerel	0.00	0.00	3.94	0.00	0.98 ± 0.84	75.40	46.43
Salmon	0.59	2.52	7.22	13.06	5.85 ± 3.03	46.25	

Table S4-9:QFASA diet estimates for managed care killer whales (n=4) layer-specific CC derived from the same managed care killer whales (average of the four individual CCs) with the KL distance. Diet estimates were more consistent and accurate with the Aitchison distance compared to the KL distance. The managed care killer whales' actual diet consisted of 32% capelin, 60% herring, 4% mackerel and 4% salmon. The mean and SE were bootstrapped 100 times. Layer 1 is closest to muscle, while layer 10 is closest to skin.

Full blubber - Full blubber CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	26.23	40.01	45.18	21.56	33.24 ± 6.52	3.89	21.72
Herring	71.59	59.99	43.48	72.72	61.95 ± 7.15	3.24	
Mackerel	2.18	0.00	0.00	5.72	1.97 ± 2.43	50.65	
Salmon	0.00	0.00	11.34	0.00	2.84 ± 3.07	29.09	
Layer 1 blubber - Layer 1 blubber CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	38.46	42.17	28.24	44.91	38.45 ± 5.63	20.14	18.11
Herring	61.54	54.31	56.67	48.82	55.33 ± 5.34	7.78	
Mackerel	0.00	3.52	12.08	0.00	3.90 ± 3.01	2.48	
Salmon	0.00	0.00	3.01	6.27	2.32 ± 2.80	42.02	
Layer 1 blubber - Inner blubber CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	52.93	58.90	44.51	60.93	54.32 ± 5.34	69.74	50.67
Herring	47.07	37.84	41.48	28.15	38.63 ± 6.43	35.61	
Mackerel	0.00	0.00	6.31	0.00	1.58 ± 1.80	60.54	
Salmon	0.00	3.26	7.70	10.93	5.47 ± 3.73	36.80	
Inner blubber - Inner blubber CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	28.68	40.05	34.19	40.52	35.86 ± 5.41	12.06	16.78
Herring	66.47	58.92	50.09	57.42	58.23 ± 5.51	2.96	
Mackerel	4.85	1.03	6.94	0.00	3.21 ± 2.97	19.86	
Salmon	0.00	0.00	8.78	2.06	2.71 ± 3.31	32.26	
Inner blubber - layer 1 CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	15.71	25.11	19.03	26.63	21.62 ± 5.36	32.44	48.40
Herring	75.95	68.93	64.59	73.37	70.71 ± 5.12	17.85	

Mackerel	8.34	5.96	12.50	0.00	6.70 ± 4.35	67.53	
Salmon	0.00	0.00	3.88	0.00	0.97 ± 2.53	75.77	
Outer layer blubber - Full CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	8.60	0.00	0.66	0.00	2.32 ± 3.43	92.76	
Herring	85.86	94.78	93.13	84.66	89.61 ± 5.40	49.35	86.00
Mackerel	5.54	5.22	6.21	15.34	8.08 ± 4.56	101.88	
Salmon	0.00	0.00	0.00	0.00	<0.01 ± 0.87	100.00	
Outer layer blubber - Outer layer CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	52.91	36.20	39.45	25.56	38.53 ± 6.78	20.40	
Herring	47.09	63.80	53.31	61.96	56.54 ± 6.58	5.77	25.73
Mackerel	0.00	0.00	0.00	12.48	3.12 ± 3.22	22.02	
Salmon	0.00	0.00	7.25	0.00	1.81 ± 2.59	54.72	
Outer layer blubber - layer 10 CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429	Killer whale - SW100500	Killer whale - SW100743	Killer whale - SW100830	Mean ± SE	% Error	Total % Error
Capelin	46.65	30.89	33.42	22.61	33.39 ± 6.71	4.35	
Herring	46.45	61.69	54.68	55.03	54.46 ± 6.51	9.23	67.79
Mackerel	6.90	7.42	8.21	22.36	11.22 ± 4.74	180.61	
Salmon	0.00	0.00	3.68	0.00	0.92 ± 2.19	76.97	
Layer 10 blubber - layer 10 CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429-10	Killer whale - SW100500-10	Killer whale - SW100743-10	Killer whale - SW100830-10	Mean ± SE	% Error	Total % Error
Capelin	51.45	26.58	41.82	0.89	30.18 ± 9.98	5.68	
Herring	47.02	73.42	45.06	92.55	64.52 ± 10.48	7.53	20.16
Mackerel	1.53	0.00	1.41	6.56	2.37 ± 2.74	40.67	
Salmon	0.00	0.00	11.72	0.00	2.93 ± 3.23	26.77	
Layer 10 blubber - Outer layer CCs - Dietary FA (21) - KL distance							
	Killer whale - SW080429-10	Killer whale - SW100500-10	Killer whale - SW100743-10	Killer whale - SW100830-10	Mean ± SE	% Error	Total % Error
Capelin	58.34	32.33	48.02	3.14	35.46 ± 10.45	10.81	
Herring	41.66	67.67	39.34	96.86	61.38 ± 11.78	2.30	33.53
Mackerel	0.00	0.00	0.00	0.00	<0.01 ± 0.50	100.00	
Salmon	0.00	0.00	12.64	0.00	3.16 ± 3.07	21.00	

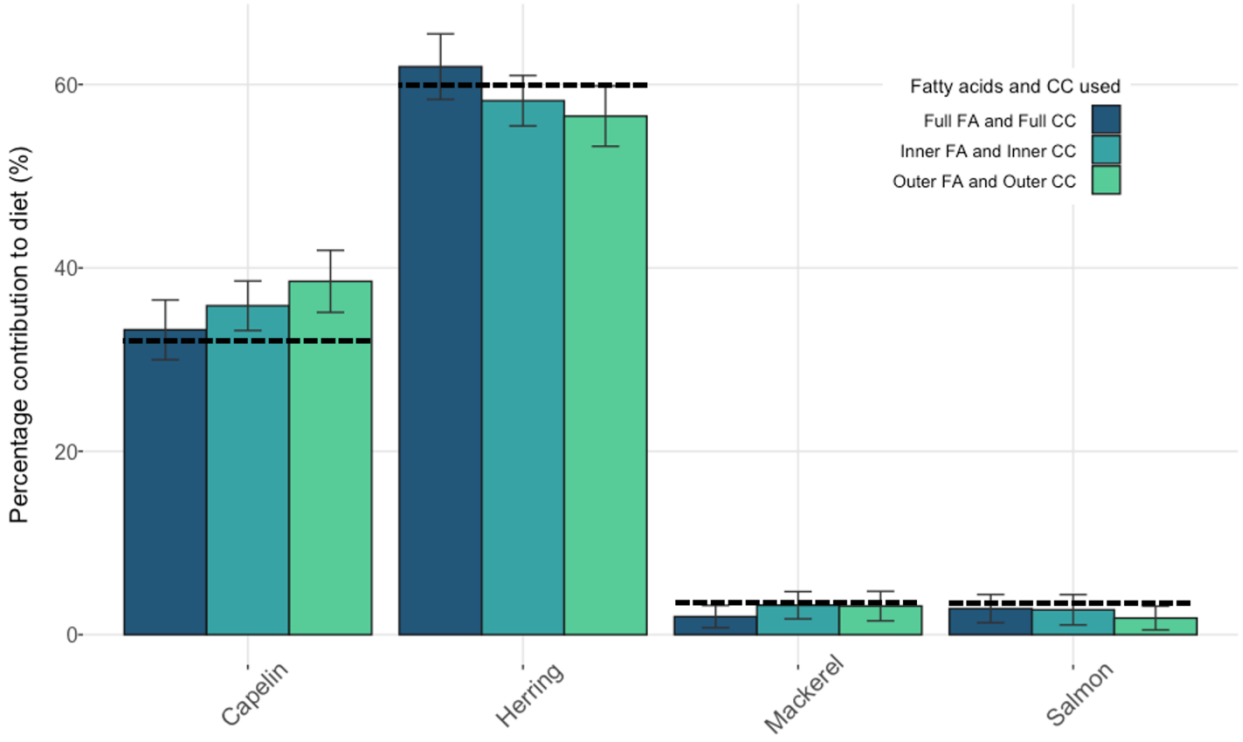

Figure S4-2: Mean diet estimates (in %) for the four managed-care killer whales when using the average CCs for the four individuals, based on the prey library consisting of capelin (n = 10), herring (n = 10), mackerel (n = 10) and salmon (n = 4). The Kullback-Leibler distance was used with the dietary FA set. The true diet (dash line) fed to the managed-care killer whales consisted of 32% capelin, 60% herring, 4% mackerel and 4% salmon.

QFASA validation on the free-ranging whales:

Very early on, we tried to include FA signatures from humpback whales (*Megaptera novaeangliae*) as they were identified as potential prey for East Greenlandic killer whales (Rosing-Asvid, pers. Comm.). However, the FA signature from the humpback whale samples we had in our lab varied so much that the humpback whale ellipse overlapped with all the other species in the PCA, and the species could not be identified during the LOPO analysis. It was thus dropped from the prey library. We found that CCs were essential to calculate QFASA estimates for the wild whales. Indeed, we ran a principal component analysis on the Greenlandic prey library and the Greenlandic killer whales before and after applying the killer whale-derived CCs;

and found that applying CCs to the free-ranging whales' FA brought the killer whales closer to their potential prey FA ranges (Fig S2). When we ran the *pred_beyond_prey* function on the full depth FA and full blubber CCs, we found that 27% of the killer whales' FA were outside the range of the potential prey FA, while 45% were outside the range of the potential prey FA without CCs, thus reinforcing the need for CCs in the QFASA modeling approach. We nonetheless ran QFASA on the free-ranging killer whales without CCs as a check and found that the main prey estimated was narwhal (94.28% of the diet), which seemed quite unrealistic, based on the limited stomach content data we obtained for these killer whales. We also decided to select the dietary FA set over the extended dietary FA set after running the *pred_beyond_prey* function. We found that only 27% of the predator FA were outside the range of the prey FA using the dietary FA set while 35% of the predator FA was outside the range of the prey FA with the extended dietary FA set, which indicates a poorer fit. Additionally, the LOPO estimates were not improved when using the extended FA set. We ran the LOPO with both distances on the dietary FA set and found that the KL distance performed better at separating the prey (Table S4-3), especially for the "harp and hooded seal" group. Indeed, the KL distance's percentage of correct attribution to the harp and hooded seal groups was 58% while it was only 37% with the Aitchison distance. Thus, we used the KL distance for diet modeling on the free-ranging killer whales.

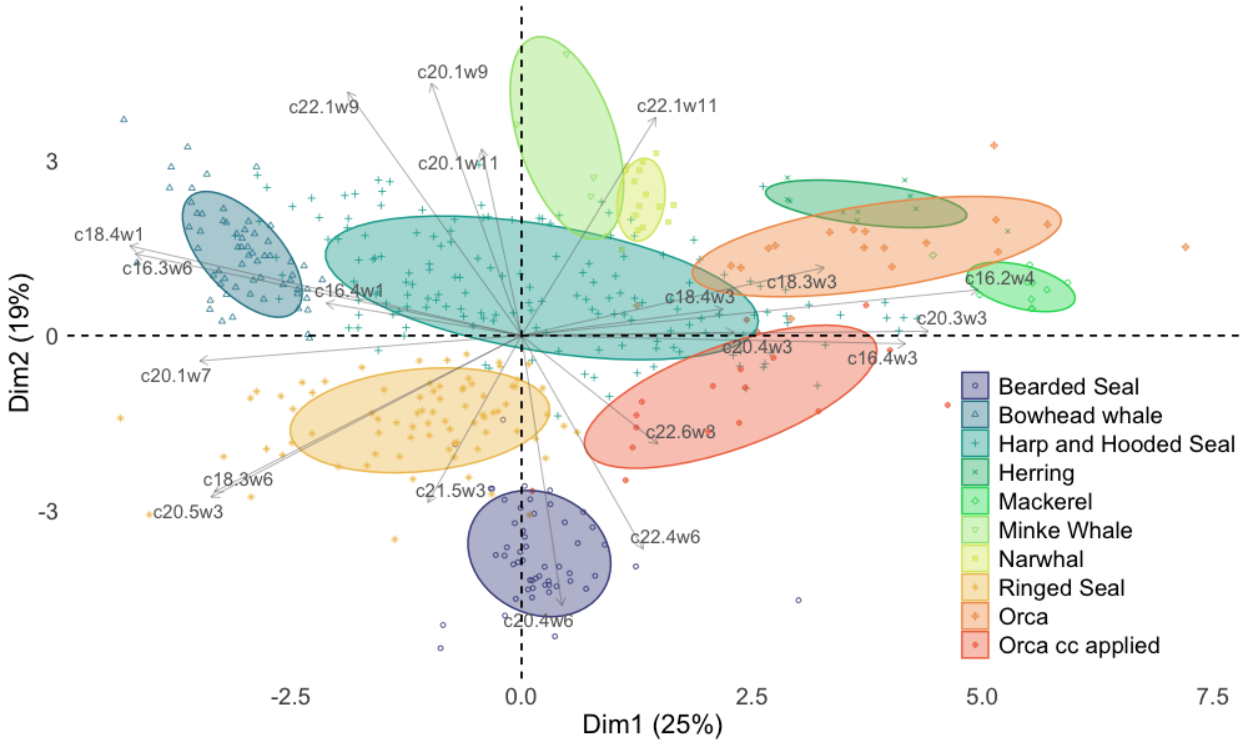

Figure S4-3:Principal component analysis of dietary FA signatures in prey and Greenlandic killer whales (full blubber) without calibration coefficients (in orange) and with calibration coefficients (full blubber) generated for this study (in red). Applying calibration coefficients to the predator puts them closer to the range of prey FA.

Table S4-10:Leave-one-prey-out analysis of the Greenlandic prey library using the Aitchison distance and the KL distance on the dietary FA set.

Dietary FA - Aitchison distance

	Bearded Seal	Bowhead whale	Harp and Hooded Seal	Herring	Mackerel	Minke Whale	Narwhal	Ringed Seal
Bearded Seal	**92.15**	1.30	0.80	0.02	0.25	0.45	1.55	3.47
Bowhead whale	0.98	**96.15**	<0.01	0.17	0.08	0.34	0.64	1.64
Harp and Hooded Seal	3.75	15.50	**37.08**	1.62	9.40	8.08	7.39	17.19
Herring	0.24	0.43	<0.01	**75.91**	17.63	1.40	2.94	1.46
Mackerel	0.34	0.05	<0.01	6.31	**92.58**	0.04	0.02	0.66
Minke Whale	1.45	1.20	1.45	0.19	3.40	**72.99**	11.28	8.04
Narwhal	0.21	5.24	<0.01	0.23	2.32	0.67	**91.20**	0.13

Ringed Seal	5.72	3.51	0.80	0.12	0.31	0.28	1.81	**87.44**

Dietary FA - KL distance

	Bearded Seal	Bowhead whale	Harp and Hooded Seal	Herring	Mackerel	Minke Whale	Narwhal	Ringed Seal
Bearded Seal	**90.41**	0.99	1.39	<0.01	0.64	0.10	3.01	3.46
Bowhead whale	6.96	**78.71**	0.45	0.49	0.32	5.94	4.48	2.65
Harp and Hooded Seal	3.85	6.03	**58.26**	0.83	5.99	8.51	3.61	12.92
Herring	<0.01	0.40	0.31	**81.84**	11.01	2.93	1.76	1.74
Mackerel	0.38	<0.01	<0.01	3.63	**94.28**	0.19	0.23	1.30
Minke Whale	2.45	0.44	6.56	7.20	2.05	**76.76**	3.47	1.07
Narwhal	1.65	0.42	1.94	<0.01	0.15	1.66	**92.98**	1.20
Ringed Seal	8.70	1.19	2.46	0.40	0.70	<0.01	2.48	**84.06**

Table S4-11:Intra-population variation in QFASA estimates for East Greenland killer whales, and relevance of estimates regarding existing stomach content data.

ID	Geography	Stomach Contents	Season	Date	Sex and Age	Herring	Mackerel	Bearded Seal	Harp and Hooded Seal	Ringed Seal	Narwhal	Minke Whale	Bowhead whale	Total Fish	Total Seal	Total Toothed whale	Total Baleen whale
35143	Greenland	Harp Seal, Minke whale	Summer	2013	Adult Female	15.25	13.18	30.37	21.33	0.00	19.87	0.00	0.00	28.43	51.70	19.87	0.00
38340	Greenland	Harp Seal	Summer	2012	Sub-adult	0.00	0.00	87.37	0.00	12.63	0.00	0.00	0.00	0.00	100.00	0.00	0.00
48335	Greenland	Harp, Hooded seal	Summer	2012	Adult Female	0.00	16.39	35.98	47.63	0.00	0.00	0.00	0.00	16.39	83.61	0.00	0.00
48336	Greenland	Harp Seal	Summer	2012	Adult Female	0.00	12.19	40.41	29.69	17.71	0.00	0.00	0.00	12.19	87.81	0.00	0.00
48337	Greenland	NA	Summer	2012	Sub-adult	0.00	16.26	48.68	25.73	9.34	0.00	0.00	0.00	16.26	83.74	0.00	0.00
48338	Greenland	Harp Seal	Summer	2012	Adult Female	0.00	17.95	32.81	49.24	0.00	0.00	0.00	0.00	17.95	82.05	0.00	0.00
48339	Greenland	NA	Summer	2012	Sub-adult	0.00	7.91	34.97	0.00	57.12	0.00	0.00	0.00	7.91	92.09	0.00	0.00
48732	Greenland	NA	Summer	2013	Adult Male	0.00	21.50	24.12	50.58	3.80	0.00	0.00	0.00	21.50	78.50	0.00	0.00
48733	Greenland	Harp Seal	Summer	2013	Adult Female	0.00	9.76	49.41	0.00	40.84	0.00	0.00	0.00	9.76	90.24	0.00	0.00
48735	Greenland	Harp Seal	Summer	2013	Sub-adult	0.00	19.83	28.54	51.63	0.00	0.00	0.00	0.00	19.83	80.17	0.00	0.00
48736	Greenland	NA	Summer	2013	Adult Female	0.00	20.32	37.26	20.05	22.37	0.00	0.00	0.00	20.32	79.68	0.00	0.00
51601	Greenland	NA	Summer	2014	Sub-adult	0.00	23.03	27.29	15.66	34.03	0.00	0.00	0.00	23.03	76.97	0.00	0.00
51606	Greenland	NA	Summer	2014	Sub-adult	0.00	7.14	53.41	0.00	39.45	0.00	0.00	0.00	7.14	92.86	0.00	0.00

51607	Greenland	NA	Summer	2014	Sub-adult	0.00	13.46	31.73	0.00	54.81	0.00	0.00	0.00	13.46	86.54	0.00	0.00
51610	Greenland	NA	Summer	2014	Sub-adult	0.00	7.66	41.84	0.00	40.91	9.59	0.00	0.00	7.66	82.75	9.59	0.00
51613	Greenland	NA	Summer	2014	Sub-adult	0.00	23.83	24.32	38.84	13.01	0.00	0.00	0.00	23.83	76.17	0.00	0.00
Bootstraped (n=100) mean % prey in Greenlandic killer whales' diet (±SE)						1.02 ± 1.10	14.51 ± 2.04	39.02 ± 5.71	22.15 ± 5.44	21.38 ± 6.65	1.91 ± 1.44	<0.01 ± <0.01	<0.01 ± <0.01	15.53 ± 1.57	82.56 ± 5.93	1.91 ± 1.44	<0.01 ± <0.01
40888	Faroe Islands	NA	Winter	2008	Adult Female	0.00	27.92	30.28	0.00	41.80	0.00	0.00	0.00	27.92	72.08	0.00	0.00
40889	Faroe Islands	NA	Winter	2008	Sub-adult	72.57	7.76	7.98	0.00	0.00	11.69	0.00	0.00	80.33	7.98	11.69	0.00
Bootstraped (n=100) % prey in Faroese0 killer whales' diet (±SE)						42.89 ± 24.91	16.01 ± 8.21	17.10 ± 7.54	<0.01 ± <0.01	17.10 ± 14.15	6.91 ± 3.98	<0.01 ± <0.01	<0.01 ± <0.01	58.89 ± 16.56	34.20 ± 7.23	6.91 ± 3.98	<0.01 ± <0.01

CONNECTING PARAGRAPH

After demonstrating that we can successfully apply Quantitative Fatty Acid Signature Analysis to wild killer whales, by collecting the outer part of their blubber, typically contained in a skin biopsy, I want to not only prove that this method can be applied to all North Atlantic killer whales, but also elucidate the diets of these individuals across the ocean. Driven by a sense of adventure and curiosity, I embarked on a remarkable mission: to initiate a ground-breaking transatlantic study on killer whales. This study, the largest and unprecedented in its scope, aims to gather extensive data and foster collaborations with diverse researchers, paving the way for new discoveries and insights into these remarkable marine creatures. The completion of this chapter demanded a substantial time investment of approximately two and a half years. Throughout this period, I dedicated considerable effort to conducting additional prey fatty acid extractions in the laboratory, consistently supplying the model with fresh prey data, and arranging multiple meetings with diverse coauthors to ensure a unified understanding of the findings. This collaborative effort culminated in the publication of a research paper in the esteemed *Journal of Animal Ecology*, which stands as one of the most influential journals in the field of ecology. Our study has sixteen authors (myself included).

5 CHAPTER FIVE: QUANTITATIVE FATTY ACID SIGNATURE REVEALS A HIGH LEVEL OF DIETARY SPECIALIZATION IN KILLER WHALES ACROSS THE NORTH ATLANTIC

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Keywords: blubber, cetacean, diets, feeding ecology, individual specialization, QFASA, predation, *Orcinus orca*

5.1 ABSTRACT:

1- Quantifying the diet composition of apex marine predators such as killer whales (*Orcinus orca*) is critical to assessing their food web impacts. Yet, with few exceptions, the feeding ecology of these apex predators remains poorly understood.

2- Here, we use our newly validated quantitative fatty acid signature analysis (QFASA) approach on nearly 200 killer whales and 900 potential prey to model their diets across the 5,000 km span of the North Atlantic.

3- Diet estimates show that killer whales mainly consume other whales in the western North Atlantic (Canadian Arctic, Eastern Canada), seals in the mid- North Atlantic (Greenland), and fish in the eastern North Atlantic (Iceland, Faroe Islands, Norway). Nonetheless, diet estimates also varied widely among individuals within most regions. This level of inter-individual feeding variation should be considered for future ecological studies focusing on North Atlantic killer whales.

4- These estimates reveal remarkable population- and individual-level variation in trophic ecology of these killer whales, which can help to assess how their predation impacts community and ecosystem dynamics in changing North Atlantic marine ecosystems.

5- This new approach provides researchers with an invaluable tool to study the feeding ecology of oceanic top predators.

5.2 ABSTRACT IN FRENCH:

1- Connaître en détails la composition du régime alimentaire des grands prédateurs marins tels que les orques (*Orcinus orca*) est primordial afin d'évaluer leurs impacts sur les écosystèmes. Pourtant, à quelques exceptions près, l'écologie alimentaire de ces super-prédateurs reste mal comprise.

2- Ici, nous utilisons notre nouvelle approche d'analyse quantitative des signatures d'acides gras (QFASA) sur près de 200 orques et 900 proies potentielles pour modéliser leur régime alimentaire à travers l'Atlantique Nord.

3- Les estimations de leurs régimes alimentaires montrent que les orques consomment principalement d'autres baleines dans l'ouest de l'Atlantique Nord (Arctique canadien, Est du Canada), des phoques dans le milieu de l'Atlantique Nord (Groenland) et des poissons dans l'est de l'Atlantique Nord (Islande, îles Féroé, Norvège). Néanmoins, ces estimations variaient considérablement d'un individu à l'autre dans la plupart des régions. Cette variation alimentaire importante entre les individus doit être prise en compte dans les futures études écologiques qui s'intéressent aux orques de l'Atlantique Nord et d'ailleurs.

4- Ces estimations révèlent des variations remarquables dans l'écologie trophique des orques tant au niveau des population que de l'individu, ce qui peut aider à évaluer l'impact de leur prédation sur la dynamique des communautés et des écosystèmes dans un contexte de changements climatiques en l'Atlantique Nord.

5- Cette nouvelle approche fournit aux chercheurs un outil inestimable pour étudier l'écologie alimentaire des super-prédateurs océaniques.

5.3 GRAPHICAL ABSTRACT AND CAPTION:

Figure 5-1: Quantifying the diets of killer whales and other top predators is crucial in a context of changing environments, because it can provide insights into how these animals adapt to shifts in their prey populations and habitat conditions. In our study, we found that killer whales have different diets across the North Atlantic, ranging from fish in the East, to marine mammals in the West. However, diets were not homogenous: individuals exhibited different prey preferences in all locations, encouraging further research on the ecology of individuals. These results were obtained by measuring the fatty acid compositions in ~200 killer whales' blubber and more than 900 potential prey items. (Photo: Anaïs Remili / The Icelandic Orca Project)

5.4 INTRODUCTION:

Elucidating the trophic interactions of marine predators is critical for understanding their ecological impacts on communities (Estes et al., 2016). It is also important to monitor the impacts of environmental changes like climate change on community dynamics (Grose et al., 2020; Sadykova et al., 2020). As the oceans warm, community dynamics are impacted,

especially in the higher latitudes (Kortsch et al., 2015; Pecuchet et al., 2020; Post et al., 2019). Indeed, climate change has already led to increases in the presence of predators like killer whales (*Orcinus orca*) in the Arctic and is expected to modify their feeding habits (Ferguson et al., 2010). Yet, the feeding ecology of killer whales across many ocean regions remains uncertain, despite decades of research on different populations.

Multiple recent studies have called for an ocean-wide comparison of the diets of North Atlantic (NA) killer whales (Dietz et al., 2020; Foote, 2022; E. Jourdain et al., 2019). Initial studies provided some insight into the trophic interactions of NA killer whales, although they were primarily based on behavioural observations. From these, Norwegian and Icelandic killer whales are thought to mostly forage on fish like Atlantic herring (*Clupea harengus*) and occasionally on marine mammals (Samarra et al., 2015; Sigurjónsson, 1988; Simila et al., 1993; Vongraven et al., 2014). Conversely, killer whales possibly target marine mammals off Greenland and along the east coast of Canada (Ferguson et al., 2012a; Ferguson et al., 2010; Higdon et al., 2012). Foote et al. (2009) suggested the existence of two NA killer whale ecotypes based on morphological and genetic data: Type 1 being a generalist that relies mostly on Atlantic herring, but also on some pinnipeds and cetaceans and Type 2 being a specialist that feeds predominantly on marine mammals (Foote et al., 2009). However, Foote recently published a letter calling to drop the type 1/type 2 classification for NA killer whales and focus on collecting more samples, specifically in remote areas, to understand the feeding ecology of these predators across the NA ocean (Foote, 2022). Understanding the feeding ecology of elusive and wide-ranging marine predators such as killer whales is challenging and requires the use of time-integrated dietary tracers such as stable isotopes or fatty acid signature analysis that represent the long-term diet of individuals, particularly when observational evidence is limited, or when stomach contents are unavailable (Kiszka et al., 2021; Trites et al., 2018).

To date, few studies have used chemical tracers to investigate the feeding ecology of NA killer whales. Studies of stable carbon and nitrogen isotope analysis and organic contaminants were consistent with observations in suggesting that Icelandic and Norwegian killer whales seem to rely mostly on fish, but also reported some degree of individual specialization on marine mammals like seals or porpoises (Andvik et al., 2020; Foote, 2012; Remili et al., 2021; Samarra et al., 2017c; Wolkers et al., 2007). Greenlandic and Canadian whales seem to rely to some extent on marine mammals based on chemical tracers (Bourque et al., 2018; Matthews et al., 2014; Matthews et al., 2021; Pedro et al., 2017). Although stable isotopes provide information on the carbon source and relative trophic position, stable isotope mixing models result in large confidence intervals for prey proportions, whereas fatty acid signatures may provide more precise estimates. Fatty acids are the main constituent of most lipids, and are released from ingested lipid molecules (e.g., triacylglycerols) during digestion (Budge et al., 2006). Fatty acids of carbon chain-length 14 or greater pass into an animal's circulation and are deposited into their lipid storage tissues, such as blubber, with little modification or in a predictable pattern, thus providing a time-integrated record of dietary intake (Iverson et al., 2004). In eastern North Pacific killer whales, fatty acid profiles were sufficiently distinct among the three reported ecotypes (resident, transient and offshore) to enable individual animals to be classified according to ecotype based on their fatty acid signature alone (Herman et al., 2005). Therefore, comparing fatty acid profiles, i.e., qualitative fatty acid analysis, among killer whales' populations and individuals may allow to identify foraging specialization across the NA (Budge et al., 2006). However, qualitative fatty acid analysis provides no information on the relative contribution of each prey species to a predator's diet.

A greater understanding of diets may be generated using quantitative fatty acid analysis (QFASA). QFASA was developed to estimate the combination of prey FA signatures that comes

closest to matching that observed in the predator, after accounting for predator metabolism and *de novo* synthesis (Iverson et al., 2004). The method requires information on the fatty acid composition (from a subset of fatty acids that is known to reflect dietary sources) of all major potential prey species and of the predator. The method also requires species-specific calibration coefficients (CCs) that account for predator metabolism, and a statistical model to minimize the statistical distance between the predator and the weighted mixture of prey species representing the diet (Iverson et al., 2004). The analysis results in diet estimates that represent the relative contribution of multiple prey sources for each sampled individual predator. We recently developed and validated QFASA for killer whales, including the determination of killer whale-specific CCs, allowing us to use this technique to explore inter- and intra-population variation in QFASA diet estimates for the first time in this species (Remili et al., 2022).

There is a need to use higher-resolution chemical tracers, like fatty acids, in samples collected within similar time frames and across regions to improve our understanding of killer whale feeding in the NA Ocean (Foote, 2022; E. Jourdain et al., 2019; Remili et al., 2022). Inter-population and inter-individual differences in feeding ecology may result in, e.g., differential risks related to changes in prey availability due to climate change and related to exposures to environmental contaminants for the killer whales (Andvik et al., 2020; Pedro et al., 2017; Remili et al., 2021). In addition, understanding the ecological impacts of killer whales on prey populations entails renewed efforts to resolve the question of the feeding ecology of NA killer whales. In this study, we present a new approach to estimate the diets of killer whales which may, in turn, inform on their predation pressure in a changing environment. We assess for the first time both inter- and intra-population variation in the diets of NA killer whales, using both qualitative and newly developed QFASA estimation approaches based on nearly 200 killer

whales sampled from west to east across the entire NA Ocean, as well as 900 specimens of their potential prey species.

5.5 MATERIAL & METHODS:

For killer whales, we collected 191 blubber samples from biopsied, stranded, or subsistence harvested individuals, including 58 individuals from the Eastern Canadian Arctic (Pond Inlet and Pangnirtung, Nunavut from 2009 to 2020), five individuals from Eastern Canada (Saint-Pierre & Miquelon, from 2019 to 2021), one individual from West Greenland (Nuuk, 2021), 18 individuals from East Greenland (Tasiilaq and Scoresby Sund, from 2012 to 2021), 48 individuals from Iceland (Grundarfjörður and Vestmannaeyjar, from 2014 to 2016), two individuals from the Faroe Islands (2008), and 59 individuals from Norway (Skjervøy area, from 2017 to 2019). Details of the samples collected from 2008-2021 are available in Table S1. For Greenlandic killer whales, full blubber samples (and attached skin for proper orientation) were opportunistically collected from subsistence harvest events and cut into ten equal layers, with layer 1 being closest to the muscle and layer 10 being closer to the skin of the animal. Faroese samples were collected from stranding events. The blubber was not oxidized, and the samples' surfaces were shaved to access the freshest tissue. Samples were then processed in a similar way to the Greenlandic samples as described in an earlier study (Bourque et al., 2018). Only the outer blubber from these samples, representing the length of a biopsy, was used in this study (Remili et al., 2022). The remaining samples consisted of skin and blubber biopsies were collected from live free-ranging killer whales using an ARTS pneumatic darting system (LKARTS-Norway, Norway) or a crossbow and stainless-steel biopsy tips (CetaDart, Denmark) ranging from 25 × 7 mm to 40 x 5 mm, depending on the location. Biopsy tips were sterilized before use and stored in clean plastic bags. All samples were generally collected from the body's midlateral region, below the dorsal fin, and stored frozen in the field at −20 °C in aluminum foil. Once back at the

lab, samples were stored at −80 °C until analysis. The full list of more than 900 prey samples collected (as well as their locations and the tissue type) includes Atlantic herring, Atlantic mackerel (*Scomber scombrus*), bearded seal (*Erignathus barbatus*), beluga whale (*Delphinapterus leucas*), bowhead whale (*Balaena mysticetus*), fin whale (*Balaenoptera physalus*), Greenland shark (*Somniosus microcephalus*), harbor porpoise (*Phocoena phocoena*), harbor seal (*Phoca vitulina*), harp seal (*Pagophilus groenlandicus*), hooded seal (*Cystophora cristata*), humpback whale (*Megaptera novaeangliae*), lumpfish (*Cyclopterus lumpus*), minke whale (*Balaenoptera acutorostrata*), narwhal (*Monodon monoceros*), and ringed seal (*Pusa hispida*) (Table S5-1). All details on fatty acid extractions and fatty acid QA/QC can be found in the supplemental text in the SI.

5.5.1 Statistical analyses

All fatty acid datasets containing the same number of fatty acids (n = 68) were renormalized to sum to 100% prior to subsequent data analysis. Only the fatty acids identified as mainly originating from diet were included (Iverson et al., 2004; Remili et al., 2022). Of those, only dietary fatty acids above 0.1% of the total FA signature (n = 15) were included to minimize analytical variation associated with small peaks on the GC-FID (Table S5-2). First, we performed a principal component analysis (PCA) on arcsine-transformed FA signatures across the NA to visually assess the FA niche widths and overlaps across the ocean basin (using the FactomineR package).

Following the PCA, we applied the newly validated QFASA model (Remili et al., 2022) to the 191 killer whales using the QFASAR package in R (version 3.6.1). QFASA produces diet estimates representing the estimated percentage of each prey species from the prey library in the diet of each predator (Remili et al., 2022). The means and standard error (SEs) of the diet estimates were obtained using bootstrap sampling (n = 100). The estimates were then corrected

to account for differences among prey species in lipid content (Table S5-1). QFASA is very sensitive to the choice of prey species included in the prey library, which prompted us to select different prey in different geographical regions, based on available literature regarding the known diet items of each killer whale regional group. For instance, we did not include beluga and narwhal in the Icelandic prey library because these prey species are not encountered in Iceland and were never reported to belong to Icelandic killer whales' diets. The list of prey species included in each prey library can be found in Table S5-1 and the justifications for the choice of prey can be found in the supplementary text.

QFASA relies on the principle that predator FA signatures can be modeled as a linear mixture of the prey FA signatures (Iverson et al., 2004). Thus, we expect the predator FA signature to be within the prey FA range. Not meeting this criterion indicates poor CCs or an incomplete prey library (Bromaghin, 2017). We tested our data using the function *pred_beyond_prey* in QFASAR to find the proportion of predator FA values outside the range of the prey values. A second QFASA assumption is limited overlap in the FA signatures among prey species (i.e., that the FA signature of each prey species is distinct) (Iverson et al., 2004). To test this assumption, we used the *leave_one_prey_out* (LOPO) function, which removes one prey signature from the library at a time and recomputes the mean prey-type and then estimates the diet of the removed prey signature. The analysis performs this computation on each prey signature, one at a time. The final output indicates the proportion of samples attributed to the correct species.

Following the QFASA analyses, we extracted the individual diet proportions and calculated the population-wide individual specialization (IS), which is the average individual proportional similarity (PS_i), with PS_i defined as the diet overlap between an individual *i* and the population:

$$PS_i = 1 - 0.5 \sum_j |p_{ij} - q_j|$$

where p_{ij} is the proportion of species *j* in the diet of individual *i*, and q_j is the average proportion of species *j* in the population's diet (Bolnick et al., 2002). The closer IS is to 100%, the more an individual's diet aligns with that of the whole population. Conversely, a lower IS percentage shows that an individual's diet differs from the population-wide diet.

Finally, as a check of the robustness of the QFASA approach, we tested for correlations between the percentage of marine mammal estimated in the diets (Arcsine-transformed) and nitrogen isotope ($\delta^{15}N$) values, and between marine mammal consumption and the sum concentrations of a diet-derived contaminant group, polychlorinated biphenyls (∑PCBs, log-transformed to achieve normality). These correlations were run for Icelandic male killer whales for which we had previously published both isotope and PCB data (Remili et al., 2021; Samarra et al., 2017c). We chose males because, unlike females, they do not transfer some of their contaminant load to their offspring, and thus their PCB concentrations are not impacted by pregnancies and lactation (Borrell et al., 1995; Wells et al., 2005).

5.6 RESULTS:

The QFASA modeling approach provided the first detailed species-specific diet estimates for NA killer whales, revealing a remarkable range of diet compositions among and within populations. Diet estimates ranged from cetacean-dominated in the western NA (Canadian Arctic, Eastern Canada) to pinniped-dominated in the mid-NA (Greenland) to fish-dominated in the eastern NA (Iceland, Faroe Islands, Norway) (Figure 5-2, Table S5-2 – S5-3).

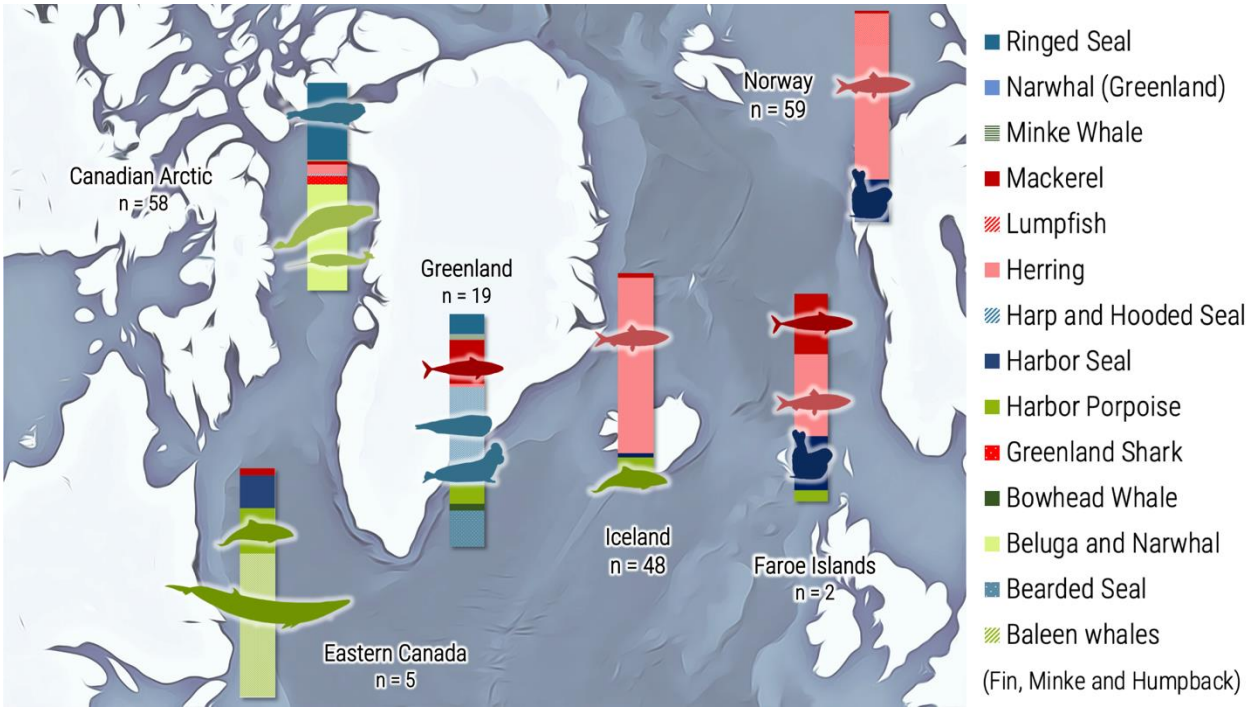

Figure 5-2:Mean proportional contributions of prey species to the diets of North Atlantic killer whales by region sampled from 2008 to 2021. Additional information can be found in Table S5-2.

QFASA estimates showed that killer whales from the western and mid-NA regions had high contributions of marine mammals in their diets, but with important differences among locations. Canadian Arctic and Eastern Canada killer whales mostly consumed cetaceans (53% ±2 and 82% ±14, respectively). Belugas and narwhals were the primary prey for Canadian Arctic killer whales, while baleen whales (fin, humpback, and minke whales) and harbor porpoises were the main prey identified for Eastern Canada killer whales. Additionally, in Canada, sampled killer whales exhibited significant spatial variation in their dietary preferences. In the Eastern Canadian Arctic, more than half of the killer whales (n=33) had beluga and narwhal diet contributions above 50%, while a quarter of the whales (n=14) had ringed seal diet contributions above 50%, and seven whales had herring diet contributions above 20% (Table S5-3). In Eastern Canada, four of the five killer whales mainly fed on baleen whales (above 60%), while one

individual mostly consumed harbor porpoise. In the mid-NA, Greenland killer whale diets included mainly seals (total seal: 66% ±5), and a lower contribution of cetaceans (total cetacean: 13% ±2) and fish (20 % ±3). For Greenland killer whales, mackerel was the most significant source of fish, and half of the sampled individuals had a contribution of mackerel above ~20% (Table S5-3).

The eastern NA killer whales showed high proportions of herring in their diets: 62% ±4 for Norway, 39% ±39 for the Faroe Islands, and 82% ±4 for Iceland, with minor contributions of lumpfish, mackerel, and marine mammals. One third of the individual (n=18) Norwegian killer whales had lumpfish contributing more than 20% to their diet. In Iceland, ten individuals had marine mammal estimates above 30% and in Norway, twelve individuals had harbor seal estimates over 30%.

The individual specialization (IS) index calculated for each regional group or subgroup revealed specialization differences across the NA (Figure 5-3A). The closer the IS index is to 1, the more the individuals' diets overlap with the population mean diet. Thus, a lower IS estimate indicates a stronger degree of individual specialization. In the western NA, Eastern Canadian Arctic killer whales showed a moderate degree of individual specialization (IS index: 0.72 ±0.02), with some individuals specializing on ringed seals and others on belugas and narwhals (Figure 5-3B). In Eastern Canada, individual specialization was also present (IS: 0.64 ±0.08), with individuals consuming varied combinations of marine mammal species (Figure 5-3C, Table S5-3). In the mid-NA, Greenland killer whales showed a higher degree of individual specialization (IS index: 0.58 ±0.04), with whales displaying varying feeding patterns ranging from seal-dominated diets to mixed diets with fish and marine mammals like seals or cetaceans (Figure 5-3D). In the Eastern NA, however, individual killer whales in Norway and Iceland showed substantial overlap with the population mean diet (IS index: 0.80 ±0.01 for Norway; 0.80

±0.03 for Iceland), indicating that most of the killer whale diets are similar and in accordance with the population use of resources (Figure 5-3F-G). For a handful of individuals in Norway (n=1) and Iceland (n=7) previously reported to feed on marine mammals based on visual observation, there was less overlap with the population mean diet (IS index: 0.58 for the Norwegian individual; 0.51 ±0.11 for the Icelandic individuals), indicating that these killer whales rely on different resources compared to most other individuals in the populations (Figure 5-3, Table S5-3). The IS index was low for Faroese whales (0.60) but based on only two individuals with different diets (Figure 5-3E).

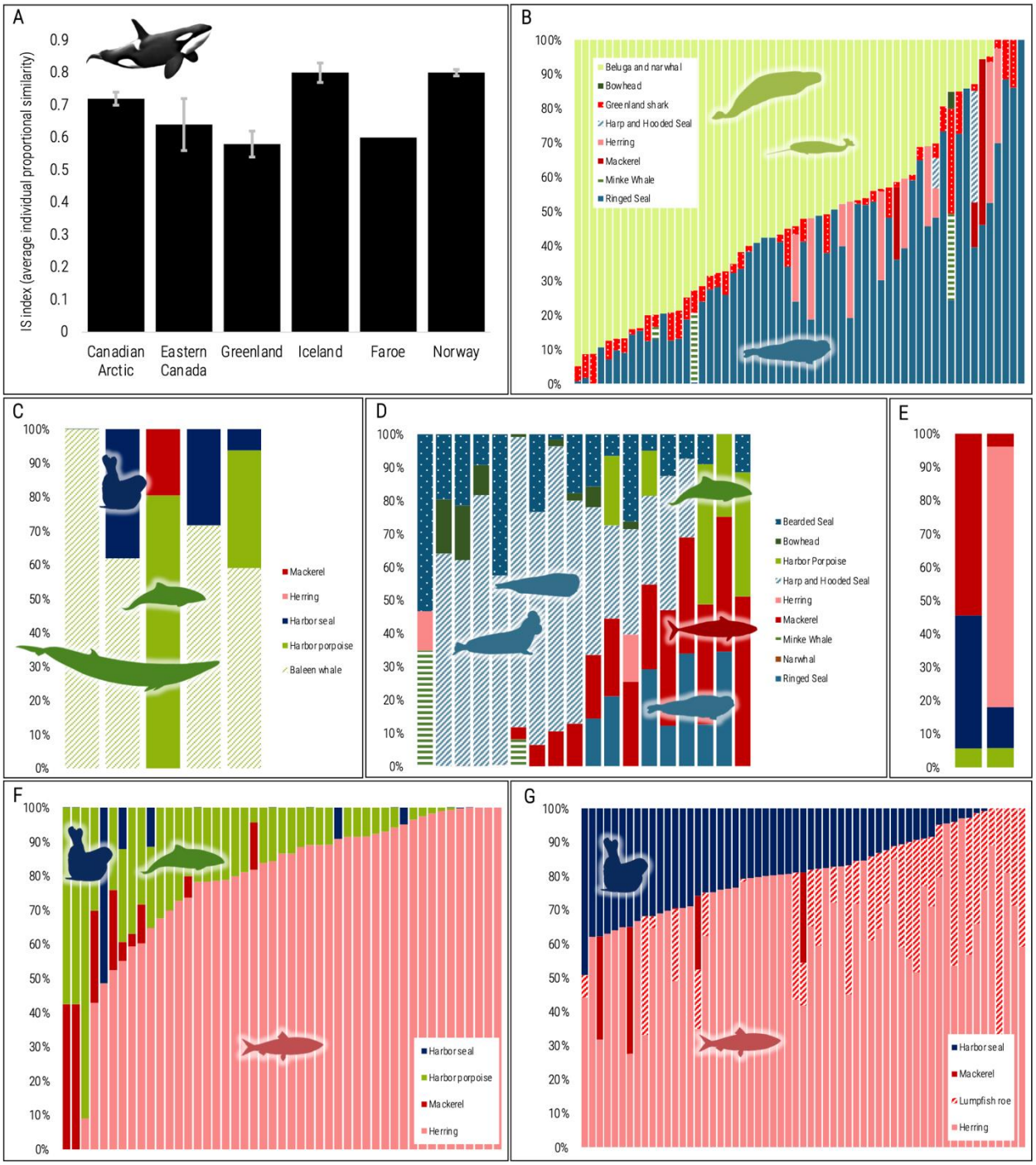

Figure 5-3: Individual dietary specialization among North Atlantic killer whales sampled from 2008-2021. A) The Individual specialization (IS) index across different geographical locations represents the average individual proportional similarity (PS_i), defined as the diet overlap between an individual i and the population mean diet; B) Individual dietary composition of Canadian Arctic killer whales; C) Individual feeding patterns of Eastern Canada killer whales; D) Individual feeding patterns of Greenlandic killer whales; E) Individual feeding patterns of Faroe Islands killer whales; F) Individual feeding patterns of Icelandic killer whales and G)

Individual feeding patterns of Norwegian killer whales. Each bar on the x-axis for figures 5-3B to 5-3G represents one individual from the location. The detailed estimates for each individual can be found in Table S5-3.

For Icelandic killer whales specifically, diets estimated by QFASA were also compared to other available indicators of their position in the food web, based on measurements realized on the same skin biopsies (Figure 5-4). Contaminant concentrations, *i.e.*, polychlorinated biphenyls (∑PCBs), and $\delta^{15}N$ values were both moderately correlated with the estimated percentage of marine mammals in the whales' diets (Figure 5-4). The Pearson correlation coefficient between the total percentage of marine mammals (Arcsine-transformed) and log ∑PCB concentrations was R=0.53 (p <0.01) in Icelandic male killer whales, while it was R=0.43 (p = 0.02) for the correlation with $\delta^{15}N$ values in the same whales. It should be noted that two killer whales that had previously been observed feeding on seals had a rather low estimated proportion of seal prey in their diet, even though their contaminant values were high (Figure S5-1).

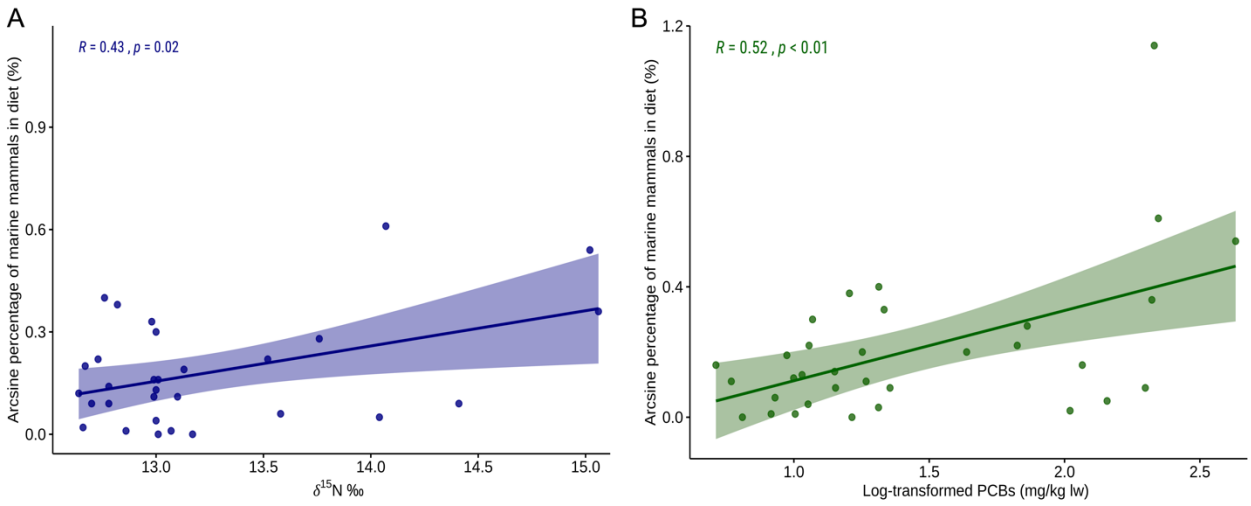

Figure 5-4:Relationship for Icelandic male killer whales (n=33) between quantitative fatty acid signature analysis (QFASA) based estimates of marine mammal consumption and A) polychlorinated biphenyl (∑PCBs) concentrations (Remili et al., 2021) and B) $\delta^{15}N$ values

(Samarra et al., 2017c). PCB concentrations, $\delta^{15}N$ values, and fatty acids signatures were determined on the same skin and blubber biopsies, allowing a meaningful comparison of the three measurements. The Pearson correlation was calculated for the total marine mammal estimate (harbor seal + harbor porpoise) vs. log ∑PCBs or $\delta^{15}N$.

The differences in QFASA estimates across the NA killer whales were further reinforced by qualitative differences, with killer whale fatty acid profiles themselves being distinctive of each region (Figure 5-5). Killer whales from Eastern Canada showed somewhat similar fatty acid profiles to Greenland and Eastern Canadian Arctic killer whales, but fatty acid signatures from these three regions were well separated from those of the killer whales from the Eastern NA. The Norwegian and Icelandic killer whales had highly overlapping fatty acid signatures. Nonetheless, for Norway and Iceland, several individuals identified on Figure 4 with an asterisk showed fatty acid profiles outside that of the eastern NA groups and closer in the PCA to the western and mid-NA groups. These individuals are known to have consumed marine mammals as inferred from previous observations and/or feeding tracers (Remili et al., 2021; Samarra et al., 2017c).

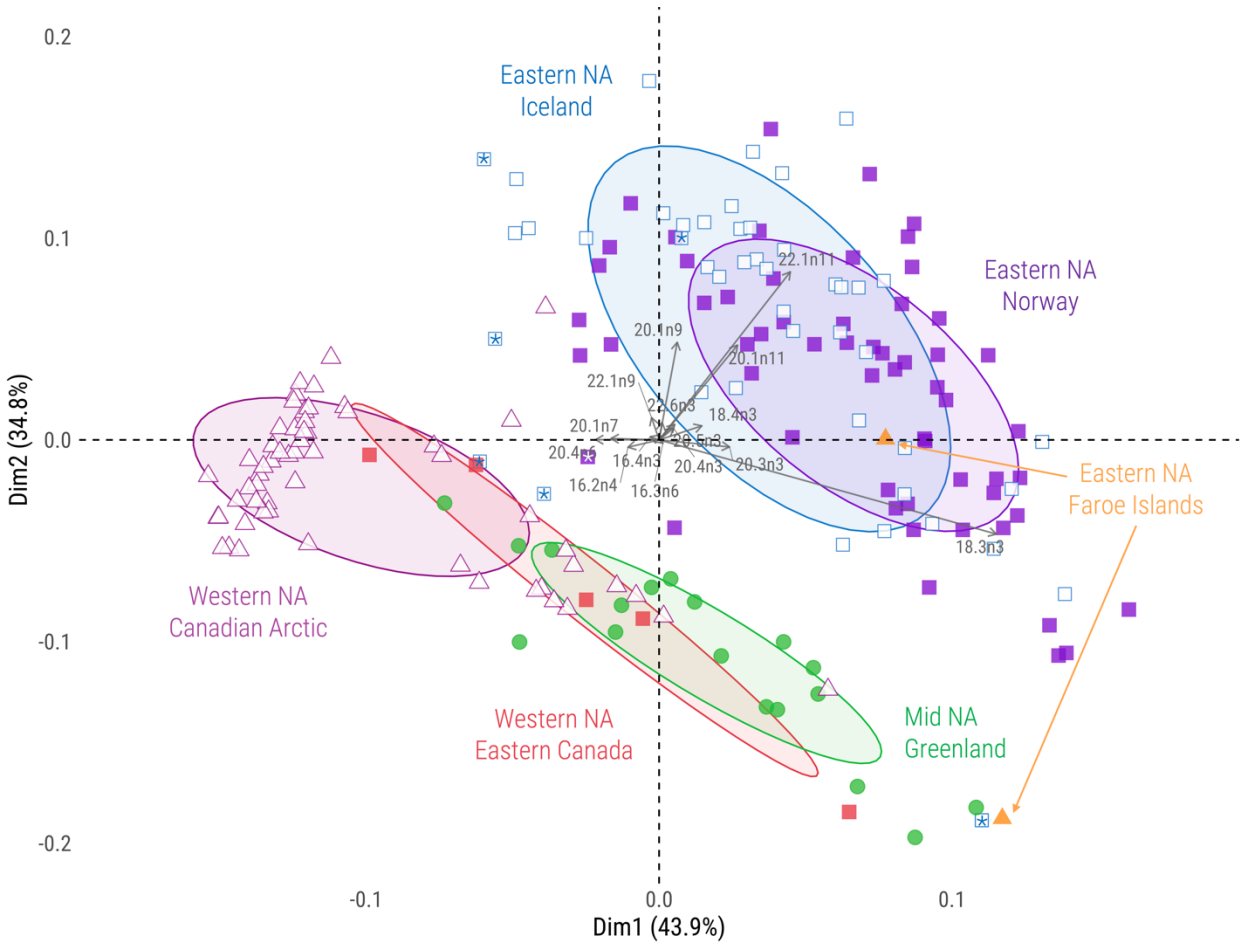

Figure 5-5:Principal component analysis of blubber fatty acid signatures of North Atlantic killer whales (n = 191) sampled from 2008-2021, grouped by region. The presence of an asterisk identifies individuals in Iceland and Norway known to have marine mammals in their diets, based on field observations and/or published studies featuring other chemical tracers.

All prey species included in the QFASA prey libraries separated relatively well on the prey fatty acid PCA (Figure 5-6). There was some noticeable overlap of certain cetacean species. Beluga whales had the largest ellipse, which caused their FA signatures to overlap slightly with the FA signatures of narwhals, bowhead whales, and harbor porpoises. The QFASA *leave_one_prey_out* (LOPO) diagnostic revealed that beluga and narwhal FA signatures were close enough that the model was unable to distinguish between the two species (Table S5-4). As a result, when included in the same library, we merged the two species. Harp and hooded seals

were also merged, based on the QFASA diagnostics of our previous study (Remili et al., 2022). Species sampled in different regions, like herring, mackerel, and narwhals, grouped close together in the prey PCA, which suggests a limited degree of geographical dietary variation within the species; thus, the ellipses for the same species but different regions still grouped closely enough that models could accurately identify them from other species (Table S5-4). Nonetheless, we decided to use region-specific prey libraries to be most representative of the potential diet of killer whales in each region. For example, Greenland herring was used to estimate the diets of Greenland killer whales, while Iceland herring was used to estimate the diets of Icelandic killer whales (more details in Table S5-1 and supplemental text). Fatty acid percentages for all NA killer whales, Icelandic prey (harbor seal, herring, mackerel) and Norwegian prey (herring, mackerel, and lumpfish) can be found in Tables S5-5 and S5-6.

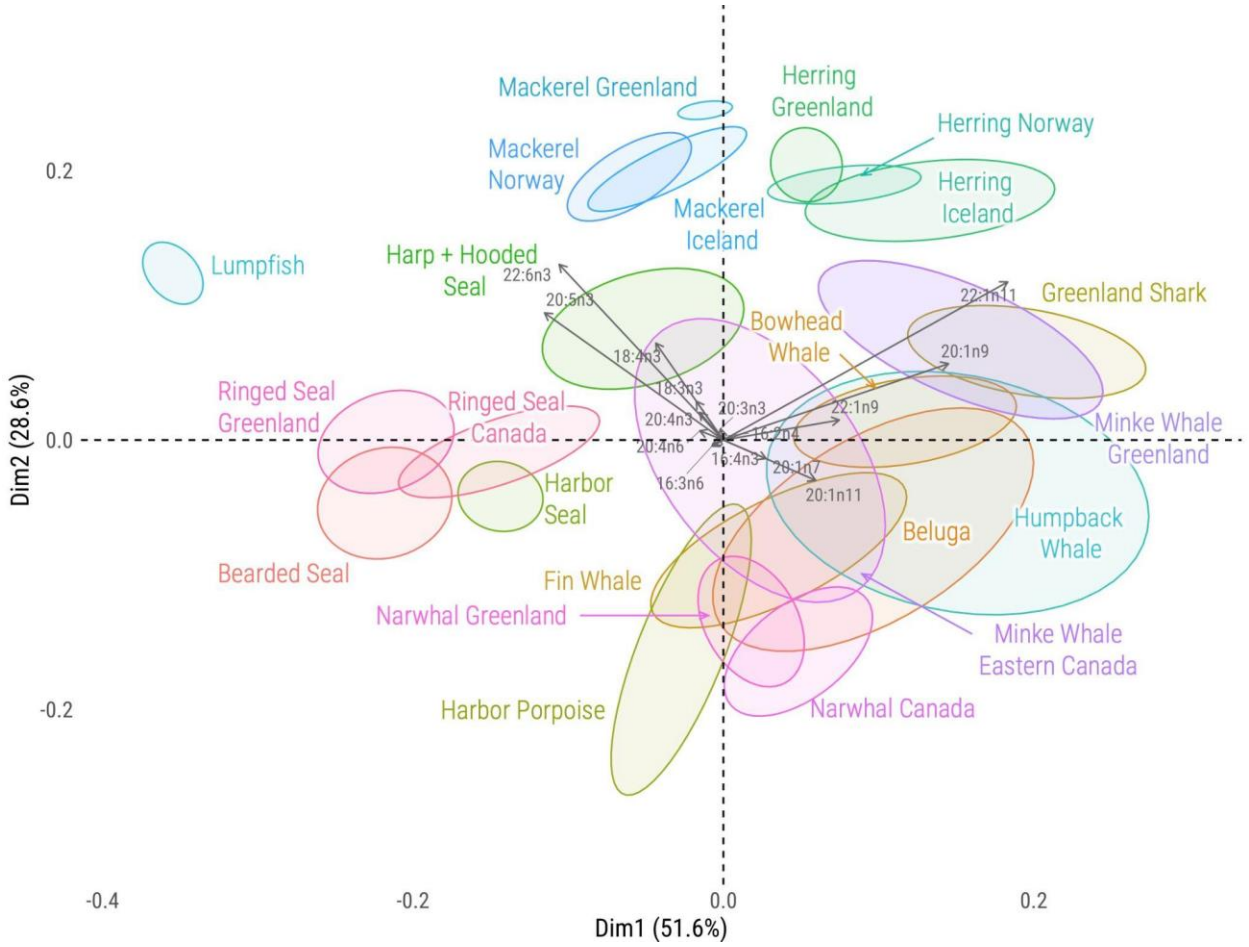

Figure 5-6:Principal component analysis of (n = 967) the fatty acid signatures of potential prey species of killer whales in the North Atlantic Ocean, representing the total prey input in all the quantitative fatty acid signature analysis (QFASA) models. The QFASA model for each killer whale group was run with subset of region-specific prey.

Various checks of the QFASA models supported its utility for modeling the diets of NA killer whales. Overall, the QFASA diagnostics indicated that the choices of prey species and calibration coefficients were adequate. The *Leave_one_prey_out* analyses ranged from 77.3% (Canada) to 89.1% (Norway) mean correct species attribution rates. The *Pred_beyond_prey* diagnostic, which represents the proportion of the predator fatty acid profiles outside the range of

the prey FA profiles, ranged from 27.1% (in Greenland) to 53.5% (in the Faroe Islands) (Table S5-4).

5.7 DISCUSSION:

The QFASA diet estimates obtained for each region identify new prey species and provide new species-level diet estimates for killer whales across the NA Ocean for the first time. Killer whales diet estimates showed that populations seem to feed on a mix of cetaceans and pinnipeds in the western NA, a mix of pinnipeds and fish in the mid-NA, and a majority of fish with some marine mammals in the eastern NA. Yet, within most locations, individual feeding preferences were also observed. These estimates are considered robust for these killer whales based on model diagnostics and consistency with other, more limited evidence from observation and measurements of other chemical tracers.

Estimates of predation on beluga and narwhal in the Canadian Arctic are consistent with local observations and coincide with a recent Arctic invasion by killer whales (Ferguson et al., 2010). In this region, the reduction of sea ice and northward range-shifting prey has led to an increasing occurrence of killer whales, and increasing predation pressure on Arctic cetaceans, particularly narwhal and beluga whales (Ferguson et al., 2012a; Ferguson et al., 2012b; Ferguson et al., 2010; Higdon et al., 2012; Matthews et al., 2019; Westdal et al., 2013). These reports have also suggested possible killer whale predation on ringed seals, the most abundant marine mammal in the Arctic (Ferguson et al., 2012a). Our QFASA estimates quantify this predation, with ringed seals estimated as the dominant prey in a quarter of the killer whales sampled in the Canadian Arctic. These findings are important in the context of changing predator-prey dynamics in the Arctic and support the need to further investigate the top-down impacts of increasing predation pressure of killer whales on Arctic marine mammals.

In Greenland, the high relative importance of harp and hooded seals was consistent with stomach contents recovered for some of the same individuals (Remili et al., 2022). A moderate contribution of mackerel was identified by QFASA, and could be explained by the possible northward distribution shifts of mackerel stocks in the NA (Berge et al., 2015; Jansen et al., 2016), and possibly by killer whales following such fish prey (Nøttestad et al., 2014; Remili et al., 2022). Predation on bearded seals has not been reported to the best of our knowledge, but this abundant prey species was consistently estimated in the diet of killer whales, particularly off Tasiilaq, Greenland, where the whales were harvested (Mattmüller et al., 2022).

Of all NA killer whales included in this study, Iceland and Norway individuals showed the highest contribution of herring in their diets, consistent with previous reports suggesting that herring is the main prey for both populations (E. Jourdain et al., 2019; Samarra et al., 2017a; Samarra et al., 2017c; Simila et al., 1996; Simila et al., 1993; Vogel et al., 2021). QFASA estimates also indicated harbor porpoise and pinnipeds in the diets of some individuals from Iceland and Norway. These specific individuals diverged from the most common, herring-dominated diet by having one-third to more than half of their diets comprised of marine mammal species. Feeding specialization among individuals in these populations is in line with distinct behavioral observations, stable carbon and nitrogen isotope values, and pollutant concentrations within individuals of the two populations (Andvik et al., 2020; Jourdain et al., 2020; Jourdain et al., 2017; Remili et al., 2021; Samarra et al., 2018; Samarra et al., 2017a; Samarra et al., 2017c).

We measured a substantial amount of dietary variation in each regional group, reflecting the complex feeding ecology of killer whales in the NA, supporting the recent suggestion to retire the terms “Ecotypes 1 and 2” from further use (Foote, 2022). Indeed, while Arctic and Eastern Canadian killer whales seem to predominantly prey on marine mammals according to their diet estimates, relative proportions for the different prey species consumed varied

substantially among individuals. In the Arctic, about a quarter of the killer whales showed diet estimates above 50% for ringed seals, while the remaining individuals showed high diet estimates for belugas and narwhals. Only three individuals in the Canadian Arctic had baleen whales in their diet estimates, which suggests minimal predation on baleen whales in this area or for these individuals. This finding is surprising, as previous research has suggested the possibility of the importance of bowhead whale predation in the Hudson Bay region of the Canadian Arctic (Galicia et al., 2016), a region not sampled in our study. Baleen whale predation may be lower than previously suggested, or Arctic individuals targeting bowhead were not captured in our study despite a reasonably large sample size. Nearly all killer whales in Eastern Canada, however, fed on baleen whales. In Greenland, we also measured strong individual dietary variation with half of the individuals showing a preference for seals, and the other half consuming both mackerel and seals. Distinct feeding preferences among individuals was also observed in Norway and Iceland, this time with most of the killer whales feeding predominantly on herring, while a small number of individuals showed a mixed diet of fish and more than half marine mammals, including either harbor seals, harbor porpoises or both. Previous research suggested that killer whales in Norway may have to supplement their herring-dominated diet with seals because they provide better nutritional value (Bories et al., 2021). These findings thus deserve further attention in the context of rapidly changing ecosystems and geographical shifts in prey availability as a result of climate change (Fossheim et al., 2015), as well as the threats posed by bioaccumulating organic contaminants (Andvik et al., 2020; Remili et al., 2021).

Qualitative comparisons of killer whale fatty acids revealed a gradient of FA profiles for killer whales across the NA. The FAs included in the analysis consisted of those fatty acids known to arise largely from dietary intake, thus minimizing possible confounding influences from physiological variation (e.g., *de novo* synthesis, metabolism) (Iverson et al., 2004).

According to our prey PCA, the FA signatures of individual species across regions did not differ substantially relative to the FA signature differences among species. It implies that the west to east FA gradient observed in the NA killer whales' profiles seems to be driven largely by differences in the diet composition and not geographic variation in prey FA profiles, in accordance with spatial FA variation shown in other studies (Thiemann et al., 2008b). The results of the PCA are thus consistent with previous knowledge of the feeding ecology of killer whales. Interestingly, some of the Iceland and Norway individuals known to prey on marine mammals to a certain extent grouped closer to the Canadian and Greenlandic whales, suggesting that qualitative FA profile analyses can at least differentiate between individuals feeding predominantly on fish vs. those feeding on marine mammals. The wide spread of these previously identified "mixed-diet" individuals in Iceland suggests a strong dietary plasticity in Iceland and Norway. Despite the relatively large dataset of the present study for killer whales in the North Atlantic compared to previous studies we were unable to assess temporal and seasonal variation within or among regions in the current study due to data limitations. This would be an important avenue for future research.

QFASA for killer whales offers an invaluable new ecological tool to quantify feeding preferences of marine predators such as cetaceans; however, some limitations should also be highlighted, specifically regarding prey library selection. Ideally, one should include all relevant prey species based on previous research using other methods, including stomach contents, stable isotopes, or behavioral observations. Conversely, researchers should select the species to include in their prey library very carefully to avoid different types of bias. Too few prey species in the library will generate false diet estimates, as the QFASA model will simply match the most probable prey based on the shortest statistical distance to the predator. If an important prey species is missing, the model will still gravitate towards the closest prey, which may not be

present or substantial in the true diet. For example, we did not include grey seal (*Halichoerus grypus*) in our Iceland and Norway prey libraries due to a lack of samples, despite reports indicating some whales feed on this species (Jourdain et al., 2017; Samarra et al., 2018). To ensure enough prey species are included in the library, researchers should pay attention that their *pred_beyond_prey* model diagnostics are not too high (Bromaghin, 2017). Anecdotally, our previous paper developing the QFASA approach for killer whales only contained Arctic seals in the prey library (Remili et al., 2022). When used on the Faroe Islands killer whales, the QFASA method estimated a high proportion (40%) of ringed seals in one of the whales' diets, which seemed unlikely based on the high-latitude habitats of ringed seals. When replacing ringed seals with our new FA data for harbor seals (a species sampled from Iceland, closer to the Faroe Islands) in the Faroe Islands prey library for the current study, the diet estimate was instead 40% harbor seal in the same killer whale. This result illustrates the need for a carefully curated prey library, with geographically relevant species. One potential caveat of this study is temporal variation in the geographic range of the predators or prey included in the models. Prey species with a large geographical range like baleen whales may show different FA profiles based on season. Future research efforts should be directed towards quantifying blubber FA turnover rates in marine mammal species to better constrain the period of feeding represented by the QFASA estimates. Another potential issue with prey libraries can arise when species with very similar FA signatures are included. In this case, the model may not be able to differentiate between species, which can cause a serious bias in the diet estimates. Researchers should thus check their values for the *leave_one_prey_out* model diagnostics and merge overlapping species when necessary (e.g., here, we merged species of baleen whales, or monodontidae in some of our libraries).

The reliability of our QFASA estimates was corroborated by the moderate correlations between the total proportion of marine mammal estimates and other feeding tracers (PCBs and $\delta^{15}N$ values). However, we observed some exceptions to the relationship between the QFASA-based diet estimates and the PCBs and $\delta^{15}N$. For instance, two of the previously identified "mixed-diet" Icelandic killer whales showed almost no marine mammal consumption based on QFASA but did show elevated blubber ∑PCB concentrations and skin $\delta^{15}N$ values. This discrepancy could be attributed to a different time-integrated diet signals from the blubber fatty acid signatures compared to the blubber PCB concentrations. PCBs and other persistent organic pollutants are extremely stable chemically, and not easily metabolized by cetaceans (Meyer et al., 2018). The only substantial way for cetaceans to reduce their blubber concentrations of most PCBs is via gestation, lactation or starvation (Tanabe et al., 1982). Values of $\delta^{15}N$ reflect the trophic position of an organism and in cetacean skin, may represent a feeding window from ~2.5 to 6 months, depending on the skin turnover rate (Wild et al., 2018). The fatty acid turnover rate in blubber is not certain but may be around a few weeks in the inner blubber, closest to the muscle for small odontocetes (Choy et al., 2019). To the best of our knowledge, estimates of the turnover rate of FAs in the outer blubber are not available but may represent the diet between several weeks and several months prior to sampling (Budge et al., 2006). As a result, blubber PCB concentrations and, possibly (although not certainly) skin $\delta^{15}N$ values may reflect dietary patterns over a longer period than outer blubber fatty acid signatures. This is an important consideration when applying QFASA to cetaceans and supports the use of multiple tracers to elucidate the feeding ecology across multiples temporal scales within a population or individual. The two Icelandic whales photographed targeting seals in Scotland were sampled in Icelandic waters, among herring-feeding killer whales. The PCBs, $\delta^{15}N$ values, and FA profiles used in this study were all derived from the same biopsy, and thus the difference in feeding patterns

suggested among the tracers supports seasonal variation in the dietary preferences of these two individuals (Remili et al., 2021; Samarra et al., 2017c). Therefore, combining multiple dietary tracers may allow for the identification of seasonal feeding patterns in future research.

Early studies suggested a possible classification of NA killer whales into Type 1/Type 2 based on evidence for different feeding ecologies (Foote et al., 2009). However, a decade of further research that combined field observations of photo-identified killer whales and dietary tracers across the NA indicates more complex patterns of variations within and among killer whale groups/populations leading to the recommendation of withdrawing the simplistic dichotomy Type 1/Type 2 (Foote, 2022). Our results of QFASA modeling based on ~200 killer whales spanning from the west to the east NA Ocean provide a panoramic view of the complex feeding strategies across the NA, as well as within-population individual feeding specialization. Further research could investigate this dietary plasticity from a genetic approach to understand how population structure may arise from this dietary variation (de Bruyn et al., 2013; Tavares et al., 2018). Regardless, our findings provide new identification of prey species and species-level diet estimates that can inform the predatory impacts of killer whales, perhaps as distinct ecological units, across the NA and other ocean basins worldwide inhabited by this ultimate apex predator.

5.8 ACKNOWLEDGMENTS:

This work was funded by the Canada Research Chairs Program (to M.A.M., 950−232183), the Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grants Program (to M.A.M., RGPIN-2019−05330), and a Canada Foundation for Innovation Grant (to M.A.M., #37873). Additional funds for sample collection in Greenland came from The Danish Cooperation for Environment in the Arctic (DANCEA) Programme (MST-112-00171 and MST-112-00199 to R.D., C.S.). Thanks for the support in the form of a

scholarship from the Fonds de Recherche du Québec Nature et Technologies (to A.Re.). The sampling of prey items in Norway was funded by the Norwegian Orca Survey and the Research Council of Norway through the MULTIWHALE project (315392) and was coordinated by Richard Karoliussen (Norwegian Orca Survey). Killer whale sampling in Norway was supported by UiT The Arctic University of Norway and the Regional Research Council of Norway. Samples collected in Saint-Pierre et Miquelon were collected under Arrêté n°431 (July 17th, 2019) of Préfecture de Saint-Pierre et Miquelon, with the collaboration of Cécile Vincent and Joël Detcheverry, and supported by Office Français de la Biodiversité (OFB). Sample collection in Iceland was possible with funding provided by an Icelandic Research Fund START Postdoctoral Fellowship (grant number 120248042 to F.I.P.S.) and Project Grant (grant number 163060 to F.I.P.S.) Thanks to Adam Pedersen, Haley Land-Miller, and Ambar Maldonado-Rodriguez for their help with FA extractions for some of the prey species and three of the Greenlandic killer whales. Thanks to Rob Letcher for overseeing the previously published PCB data for the Icelandic killer whales. Thanks to Tera Edkins from DFO, Canada, for her assistance with the sample shipping. Thank you to all the people involved in the fieldwork in Iceland for their help collecting data. Thank you to Ricky Kilabuk for the collection of killer whale samples from Pangnirtung, Nunavut. Funding for Canadian samples was provided by DFO's Nunavut Implementation Fund (NIF) and Species at Risk Act (SARA), the Nunavut General Monitoring Plan (NGMP), and the Nunavut Wildlife Management Board (NWMB).

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5.10 SUPLEMENTARY INFORMATION

Fatty acid analysis

FAs were extracted and quantified as described in previous studies (Bourque et al., 2018). In brief, for marine mammals, total lipids were extracted from blubber using the Folch method (Folch et al., 1957). Whole fish were first homogenized in a food processor, extracted via a modified Folch method and filtered (Budge et al., 2006). All marine mammal and fish FA extracts were *trans*-esterified using the Hilditch reagent to produce fatty acid methyl esters (FAMEs). The FAMEs were separated, identified, and the mass percentage of each of 68 FAs was quantified by gas chromatography with flame ionization detection (GC-FID) on an Agilent 8860 system (Santa Clara, CA, USA). Each FA is named using the nomenclature x:y*n*z, where x is the length of the carbon chain, y is the number of double bonds, and z is the position of the first double bond from the methyl ('n') end of the chain. Fatty acids for all NA killer whales can be found in Tables S5-5 and fatty acids for Icelandic and Norwegian prey can be found in Table S6.

Quality Assessment and Control (QA/QC)

QA/QC included the extraction and analysis of standard reference material, SRM1945 pilot whale blubber, from the US National Institute of Standards and Technology (NIST), with each batch of 11 samples for the Greenlandic, Norwegian, and Icelandic killer whales. The SRM was run 13 times, and the median relative standard deviation of the FA values was 9% compared to the published 27 individual FA values (Kucklick et al., 2010). A duplicate was run in every batch for the Icelandic prey samples and the Canadian killer whales. The percent difference of

the duplicates averaged 3% for the Icelandic prey and 9% for the Canadian killer whales. We thus decided to randomly select one of the duplicates for each individual in further analyses.

Prey selection per location

Table S5-1:List of sample and collection details for the killer whales (Orcinus orca) and their prey: Atlantic herring (Clupea harengus), Atlantic mackerel (Scomber scombrus), bearded seal (Erignathus barbatus), beluga (Delphinapterus leucas), bowhead whales (Balaena mysticetus), fin whale (Balenoptera physalus), Greenland shark liver (Somniosus microcephalus), harbor porpoise (Phocoena phocoena), harbor seal (Phoca vitulina), harp seal (Pagophilus groenlandicus), hooded seal (Cystophora cristata), humpback whale (Megaptera novaeangliae), lumpfish roe (Cyclopterus lumpus), minke whale (Balaenoptera acutorostrata), narwhal (Monodon monoceros), and ringed seal (Pusa hispida).

Species	N	ID	Geography	Date	Prey Library	% lipid used	Paper originally published (FA)
Predator - Free-ranging killer whales (n=193)							
Killer whale	48	IS003-IS423	Iceland	2014-2016	B	–	This paper
Killer whale	59	17001-19023	Northern Norway	2017-2019	A	–	This paper
Killer whale	2	40888-40889	Faroe Islands	2008	B	–	Bourque et al. 2018
Killer whale	19	35143-51613	Greenland	2012-2014, 2021	C	–	Bourque et al. 2018 & This paper
Killer whale	5	KW1-KW5	Eastern Canada	2019-2021	E		This paper
Killer whale	58	KW01-KW18 KW4001-KW4008 ARPI_00_02_2019-ARPI_00_11_2019 KWPG1-2020-KWPI15-2020	Canadian Arctic	2009-2020	D	–	This paper
Prey for the QFASA library on the free-ranging killer whales							
Fish (Whole fish homogenized, unless otherwise mentioned)							
Atlantic Herring	10	GLHerring-31-40	East Greenland	NA	C – D - E	16	Remili et al. 2022
Atlantic Herring	10	H1-H10	Iceland	2014-2020	B	14	This paper
Atlantic Herring	19	He2021H1-He2021H19	Norway	2021	A	16	This paper
Atlantic Mackerel	10	GLMackerel-41-50	East Greenland	NA	C – D - E	22	Remili et al. 2022

Atlantic Mackerel	10	M1-M10	Iceland	2014-2020	B	26	This paper
Atlantic Mackerel	3	Ma20211-Ma20213	Norway	2021	A	19	This paper
Lumpfish (Roe)	6	Lu201804-Lu202104	Norway	2018-2021	A	7	This paper
Greenland Shark (Liver)	17	BMGS41L - BMGS58L	Greenland	2009	D	72	Unpublished
			Seals (Blubber)				
Bearded Seal	55	SGBS1-SGBS8	South Greenland	NA	C	30	McKinney et al. 2013
		SIP107-SIP317	Davis Strait	NA			Thiemann et al. 2008
		UMP197-UMP480	Davis Strait	NA			Thiemann et al. 2008
Harbor Seal	15	HS1-HS17	Iceland	NA	A - B - E	30	This paper
Harp Seal	239	SIP001-SIP283	Davis Strait	NA	C - D	30	Thiemann et al. 2008
		Tucker001-Tucker 114	Davis Strait	NA			Thiemann et al. 2008
Hooded Seal	32	SIP098-SIP272	Davis Strait	NA	C - D	30	Thiemann et al. 2008
		Tucker115-Tucker133	Davis Strait	NA			Thiemann et al. 2008
Ringed Seal	132	EGRS24931-EGRS34946	East Greenland	NA	C	30	Mckinney et al. 2013
		SIP227-SIP319	Davis Strait	NA			Thiemann et al. 2008
		UMP332-333	Davis Strait	NA			Thiemann et al. 2008
Ringed Seal	9	RB2018-02 - RB2018-20	Canadian Arctic	2018	D	30	Facciola et al. 2022
			Cetaceans (Blubber)				
Beluga	269	ARGF-0-1036 - PGDL-92-11	Canadian Arctic	NA	D	30	DFO
Bowhead Whale	54	BM-01-2008-BMIG-09-014	Canadian Arctic	2008-2009	C - D	30	DFO
Fin Whale	7	FIN01-FIN13	Newfoundland	NA	E	30	DFO
Harbor Porpoise	10	PP1-PP13	East Greenland	2018	B - E	30	Unpublished
Humpback Whale	7	HUM02-HUM11	Newfoundland	NA	E	30	DFO
Minke Whale	5	Ba_001-2017/0001	East Greenland	NA-2017	C - D	30	Unpublished
Minke Whale	19	BA19-1-BA19-22	Saint Pierre & Miquelon	2019	E	30	Unpublished
Narwhal	16	53801-53846	East Greenland	NA	C	30	Unpublished
Narwhal	13	39270-57590	Canadian Arctic	2011	D	30	DFO

Supplementary text: Prey selection per location

To select the different prey species to be added to the prey libraries for each location, we relied on the literature and the prey we had available in each location.

For Iceland, killer whales were previously separated into two diet-types (fish-feeding and mixed-diet) based on behavioural observations, stable isotopes, and persistent organic pollutants (Samarra et al., 2017b; Remili et al., 2021). These previous studies mentioned predation events on seals and porpoises, as well as herring. Mackerel was also added because of sample availability, its relative abundance around Iceland, and the reported depredation event around fishing vessels in the area (Luque et al., 2006).

For the Norwegian prey library, we included herring since the whales were sampled on herring wintering grounds. We also included mackerel as local fishermen reported killer whale depredation events around mackerel fishing boats (A. Rikardsen., pers. comm.). One of our Norwegian individuals may have been seal-feeding, based on nearshore foraging behavior observed at time of sampling (A. Rikardsen., pers. comm.). Finally we added Norwegian lumpfish roe to the prey library, as previous research reported lumpfish consumption by Norwegian killer whales (Eve Jourdain et al., 2019). This lumpfish consumption was observed for individuals also present at herring wintering grounds.

Greenlandic and Faroese whales were harvested, and stomach contents reported harp and hooded seals in the diet of the whales (Pedro et al., 2017; Remili et al., 2022). Since not much literature exists on Greenlandic killer whales, we included as many species as we could from the geographical area, based on publicly available FA databases, and private, unpublished FA data from the area.

Canadian Arctic killer whales were previously reported feeding on marine mammals, including seals, bowhead whales, belugas, and narwhals (Ferguson et al., 2012a; Ferguson et al., 2012b; Ferguson et al., 2010; Lefort et al., 2020; Matthews et al., 2020). Other reports mentioned predation on fish (Laidre et al., 2006). As herring and mackerel were the only fish we had for the general area, we included them in the model. We also relied on unpublished data reporting deep dives and potential Greenland shark predation for some Canadian Arctic killer whales and included Greenland shark in the library (S. Ferguson, pers. com).

Finally, Eastern Canadian whales off Saint Pierre and Miquelon are largely understudied. Thus, we included prey from a range of large range of geographically relevant potential species including baleen whales, toothed whales (harbor porpoise), seal (harbor seal) and fish (herring and mackerel).

Additional information on the killer whale PCA (Figure 5-5):

PC1 and PC2 contributed to 43.9% and 34.8% of the total variance respectively, thus both contributing to ~ 80% of the fatty acid variation between individuals. PC1 was mainly driven by differences in 18:3n3 (74.8% contribution) and 22:1n11 (11.3% contribution) while PC2 was driven mostly by 22:1n11 (49.6% contribution), 20:1n9 (16.7% contribution), and 20:1n11 (15.7% contribution). Norwegian and Icelandic killer whales show higher percentages of 22:1n11, 20:1n11, 20:1n9 and 18:3n3 than killer whales from the rest of the Atlantic. When considering the prey PCA (Figure 5-6), we can notice that herring and mackerel, two pelagic fish species, seem to have higher 22:1n11, 20:1n11, 20:1n9 and 18:3n3, compared to cetaceans and pinnipeds. While it is difficult to infer prey preferences based on qualitative data, the clear difference between the eastern NA killer whale niches and those of the mid- and western NA seems realistic, based on previous observations of herring predation in the Eastern NA.

Table S5-2:Mean ± SE QFASA diet estimates (in %) for all North Atlantic killer whales (n=150), and individual specialization (mean percentage similarity) based on the whole population average. Please note: The IS measure of individual specialisation corresponds to the average similarity between individuals' diet and the population diet. When all individuals consume the full set of population resources, IS equals 1.0. As individuals use smaller subsets of the population diet, IS declines towards zero.

	Canadian Arctic	Eastern Canada	Greenland	Iceland	Faroe	Norway
Baleen whales (Fin, Minke and Humpback)	–	62.6 ± 16.1	–	–	–	–
Bearded Seal	–	–	15.6 ± 3.6	–	–	–
Beluga and Narwhal	51.8 ± 6.0	–	–	–	–	–
Bowhead Whale	0.1 ± < 0.1	–	2.9 ± 1.3	–	–	–
Greenland Shark	4.0 ± 1.5	–	–	–	–	–
Harbor Porpoise	–	19.8 ± 12.3	8.1 ± 4.2	13.7 ± 3.3	5.7 ± <0.1	–
Harbor Seal	–	14.3 ± 8.3	–	2.1 ± 1.6	26.1 ± 13.8	20.6 ± 3.2
Harp and Hooded Seal	0.7 ± 0.9	–	41.7 ± 7.4	–	–	–
Herring	4.9 ± 1.4	<0.1 ± <0.1	1.5 ± 1.3	81.7 ± 4.3	39.1 ± 39.1	62.43 ± 4.2
Lumpfish	–	–	–	–	–	15.2 ± 6.3
Mackerel	1.6 ± 0.9	3.4 ± 3.0	18.8 ± 4.4	2.7 ± 1.0	29.2 ± 25.3	1.8 ± 0.8
Minke Whale	0.7 ± 0.6	–	2.4 ± 2.3	–	–	–
Narwhal (Greenland)	–	–	< 0.1 ± < 0.1	–	–	–
Ringed Seal	36.2 ± 5.2	–	9.0 ± 4.0	–	–	–
Individual Specialization Index (IS)	0.72 ± 0.02	0.64 ± 0.08	0.58 ± 0.04	0.80 ± 0.03	0.6 ± 0.00	0.80 ± 0.01

Table S5-3:Individual estimates for all killer whales in the NA (in %).

Population	ID	Year	Baleen Whale	Bearded Seal	Beluga and Narwhal	Bowhead Whale	Greenland Shark	Harbor Porpoise	Harbor Seal	Harp and Hooded Seal	Herring	Lumpfish	Mackerel	Minke Whale	Narwhal (Greenland)	Ringed Seal
Norway	no17001	2017	–	–	–	–	–	–	33.21	–	66.79	0	0	–	–	–
Norway	no17002	2017	–	–	–	–	–	–	28.92	–	71.08	0	0	–	–	–
Norway	no17003	2017	–	–	–	–	–	–	9.3	–	51.68	39.03	0	–	–	–
Norway	no17004	2017	–	–	–	–	–	–	30.18	–	69.82	0	0	–	–	–
Norway	no17005	2017	–	–	–	–	–	–	24.82	–	62.31	12.86	0	–	–	–
Norway	no17006	2017	–	–	–	–	–	–	19.39	–	80.61	0	0	–	–	–
Norway	no17008	2017	–	–	–	–	–	–	31.86	–	64.67	3.47	0	–	–	–
Norway	no17009	2017	–	–	–	–	–	–	15.47	–	71.73	12.8	0	–	–	–
Norway	no17010	2017	–	–	–	–	–	–	35.06	–	64.94	0	0	–	–	–
Norway	no17011	2017	–	–	–	–	–	–	29.42	–	48.98	21.31	0.28	–	–	–
Norway	no17012	2017	–	–	–	–	–	–	37.79	–	31.74	0	30.47	–	–	–
Norway	no17013	2017	–	–	–	–	–	–	9.94	–	55.59	34.47	0	–	–	–
Norway	no17014	2017	–	–	–	–	–	–	19.91	–	80.09	0	0	–	–	–
Norway	no17015	2017	–	–	–	–	–	–	13.17	–	64.38	22.45	0	–	–	–
Norway	no17016	2017	–	–	–	–	–	–	8.38	–	71.04	20.59	0	–	–	–
Norway	no17017	2017	–	–	–	–	–	–	31.87	–	33.09	35.04	0	–	–	–
Norway	no17018	2017	–	–	–	–	–	–	16.86	–	45.05	38.09	0	–	–	–
Norway	no17019	2017	–	–	–	–	–	–	17.64	–	82.36	0	0	–	–	–
Norway	no17021	2017	–	–	–	–	–	–	11.1	–	84.89	4.01	0	–	–	–
Norway	no18001	2018	–	–	–	–	–	–	19.77	–	80.23	0	0	–	–	–
Norway	no18005	2018	–	–	–	–	–	–	18.93	–	43.51	37.56	0	–	–	–
Norway	no18006	2018	–	–	–	–	–	–	14.13	–	60.98	24.89	0	–	–	–
Norway	no18007	2018	–	–	–	–	–	–	4.45	–	95.55	0	0	–	–	–
Norway	no18008	2018	–	–	–	–	–	–	20.26	–	79.74	0	0	–	–	–

Norway	no18009	2018	–	–	–	–	–	–	31.04	–	68.96	0	0	–	–	–
Norway	no18010	2018	–	–	–	–	–	–	8.87	–	77	14.13	0	–	–	–
Norway	no18011	2018	–	–	–	–	–	–	23.73	–	76.27	0	0	–	–	–
Norway	no18012	2018	–	–	–	–	–	–	20.75	–	78.31	0.94	0	–	–	–
Norway	no18014	2018	–	–	–	–	–	–	20.56	–	79.44	0	0	–	–	–
Norway	no18015	2018	–	–	–	–	–	–	4.02	–	53.53	42.45	0	–	–	–
Norway	no18016	2018	–	–	–	–	–	–	18.91	–	41.77	12.69	26.63	–	–	–
Norway	no18017	2018	–	–	–	–	–	–	12.35	–	71.93	15.72	0	–	–	–
Norway	no18020	2018	–	–	–	–	–	–	19.58	–	80.42	0	0	–	–	–
Norway	no18021	2018	–	–	–	–	–	–	34.97	–	27.6	0	37.42	–	–	–
Norway	no18022	2018	–	–	–	–	–	–	2.78	–	56.77	40.45	0	–	–	–
Norway	no18023	2018	–	–	–	–	–	–	0	–	59.12	40.88	0	–	–	–
Norway	no18024	2018	–	–	–	–	–	–	24.05	–	75.95	0	0	–	–	–
Norway	no18025	2018	–	–	–	–	–	–	37.01	–	62.99	0	0	–	–	–
Norway	no18026	2018	–	–	–	–	–	–	23.42	–	76.58	0	0	–	–	–
Norway	no18027	2018	–	–	–	–	–	–	24.76	–	75.24	0	0	–	–	–
Norway	no18029	2018	–	–	–	–	–	–	29.41	–	70.59	0	0	–	–	–
Norway	no19002	2019	–	–	–	–	–	–	2.89	–	97.11	0	0	–	–	–
Norway	no19005	2019	–	–	–	–	–	–	35.96	–	64.04	0	0	–	–	–
Norway	no19007	2019	–	–	–	–	–	–	18.16	–	65.11	16.72	0	–	–	–
Norway	no19008	2019	–	–	–	–	–	–	17.82	–	59.44	22.74	0	–	–	–
Norway	no19009	2019	–	–	–	–	–	–	37.95	–	62.05	0	0	–	–	–
Norway	no19010	2019	–	–	–	–	–	–	10.61	–	58.88	30.51	0	–	–	–
Norway	no19011	2019	–	–	–	–	–	–	0	–	21.08	78.92	0	–	–	–
Norway	no19013	2019	–	–	–	–	–	–	15.38	–	84.62	0	0	–	–	–
Norway	no19014	2019	–	–	–	–	–	–	0	–	88.97	11.03	0	–	–	–
Norway	no19015	2019	–	–	–	–	–	–	17.08	–	82.92	0	0	–	–	–

Norway	no19016	2019	–	–	–	–	–	–	0.76	–	75.92	23.32	0	–	–	–
Norway	no19017	2019	–	–	–	–	–	–	4.83	–	79.65	15.52	0	–	–	–
Norway	no19018	2019	–	–	–	–	–	–	1.38	–	65.8	32.82	0	–	–	–
Norway	no19019	2019	–	–	–	–	–	–	0	–	80.98	19.02	0	–	–	–
Norway	no19020	2019	–	–	–	–	–	–	0	–	71.22	28.78	0	–	–	–
Norway	no19021	2019	–	–	–	–	–	–	17.21	–	72.14	10.65	0	–	–	–
Norway	no19023	2019	–	–	–	–	–	–	25.83	–	34.86	17.58	21.73	–	–	–
Norway	no1900X	2019	–	–	–	–	–	–	49.2	–	44.14	6.66	0	–	–	–
Iceland	Mixed - IS015	2014-2016	–	–	–	–	–	0	51.35	–	48.65	–	0	–	–	–
Iceland	Mixed - IS256	2014-2016	–	–	–	–	–	57.56	0	–	0	–	42.44	–	–	–
Iceland	Mixed - IS003	2014-2016	–	–	–	–	–	90.89	0	–	9.11	–	0	–	–	–
Iceland	Mixed - IS241	2014-2016	–	–	–	–	–	23.79	11.44	–	64.77	–	0	–	–	–
Iceland	Mixed - IS243	2014-2016	–	–	–	–	–	0	9.13	–	90.87	–	0	–	–	–
Iceland	Mixed - IS172	2014-2016	–	–	–	–	–	0	4.97	–	95.03	–	0	–	–	–
Iceland	Mixed - IS253	2014-2016	–	–	–	–	–	57.56	0	–	0	–	42.44	–	–	–
Iceland	Fish - IS251	2014-2016	–	–	–	–	–	15.73	0	–	84.27	–	0	–	–	–
Iceland	Fish - IS011	2014-2016	–	–	–	–	–	1.76	0	–	98.24	–	0	–	–	–
Iceland	Fish - IS136	2014-2016	–	–	–	–	–	27.26	0	–	72.74	–	0	–	–	–
Iceland	Fish - IS151	2014-2016	–	–	–	–	–	21.66	0	–	78.34	–	0	–	–	–
Iceland	Fish - IS211	2014-2016	–	–	–	–	–	20.09	0	–	73.61	–	6.3	–	–	–

Iceland	Fish - IS152	2014-2016	–	–	–	–	–	4.3	0	–	81.83	–	13.86	–	–	–
Iceland	Fish - IS122	2014-2016	–	–	–	–	–	8.49	0	–	91.51	–	0	–	–	–
Iceland	Fish - IS257	2014-2016	–	–	–	–	–	0	0.2	–	99.8	–	0	–	–	–
Iceland	Fish - IS018	2014-2016	–	–	–	–	–	32.43	0	–	67.57	–	0	–	–	–
Iceland	Fish - IS028	2014-2016	–	–	–	–	–	27.11	12.21	–	55.11	–	5.57	–	–	–
Iceland	Fish - IS071	2014-2016	–	–	–	–	–	2.52	0	–	97.48	–	0	–	–	–
Iceland	Fish - IS280	2014-2016	–	–	–	–	–	10.89	0	–	89.11	–	0	–	–	–
Iceland	Fish - IS046	2014-2016	–	–	–	–	–	20.08	0	–	79.92	–	0	–	–	–
Iceland	Fish - IS068	2014-2016	–	–	–	–	–	0	0	–	100	–	0	–	–	–
Iceland	Fish - IS159	2014-2016	–	–	–	–	–	36.93	0	–	59.4	–	3.67	–	–	–
Iceland	Fish - IS008	2014-2016	–	–	–	–	–	8.51	0	–	91.49	–	0	–	–	–
Iceland	Fish - IS155	2014-2016	–	–	–	–	–	13.48	0	–	86.52	–	0	–	–	–
Iceland	Fish - IS139	2014-2016	–	–	–	–	–	30.03	0	–	69.97	–	0	–	–	–
Iceland	Fish - IS169	2014-2016	–	–	–	–	–	21.44	0	–	78.56	–	0	–	–	–
Iceland	Fish - IS183	2014-2016	–	–	–	–	–	3.53	0	–	96.47	–	0	–	–	–
Iceland	Fish - IS117	2014-2016	–	–	–	–	–	13.43	0	–	86.57	–	0	–	–	–
Iceland	Fish - IS062	2014-2016	–	–	–	–	–	21.18	0	–	78.82	–	0	–	–	–

Iceland	Fish - IS306	2014-2016	–	–	–	–	–	0.72	0	–	99.28	–	0	–	–	–
Iceland	Fish - IS067	2014-2016	–	–	–	–	–	11.54	0	–	88.46	–	0	–	–	–
Iceland	Fish - IS267	2014-2016	–	–	–	–	–	18.94	0	–	81.06	–	0	–	–	–
Iceland	Fish - IS010	2014-2016	–	–	–	–	–	5.79	0	–	94.21	–	0	–	–	–
Iceland	Fish - IS288	2014-2016	–	–	–	–	–	24.18	0	–	52.39	–	23.42	–	–	–
Iceland	Fish - IS143	2014-2016	–	–	–	–	–	1.02	0	–	98.98	–	0	–	–	–
Iceland	Fish - IS423	2014-2016	–	–	–	–	–	0	0	–	100	–	0	–	–	–
Iceland	Fish - IS047	2014-2016	–	–	–	–	–	0	0	–	100	–	0	–	–	–
Iceland	Fish - IS045	2014-2016	–	–	–	–	–	7.06	0	–	92.94	–	0	–	–	–
Iceland	Fish - IS254	2014-2016	–	–	–	–	–	10.93	0	–	89.07	–	0	–	–	–
Iceland	Fish - IS078	2014-2016	–	–	–	–	–	16.21	0	–	83.79	–	0	–	–	–
Iceland	Fish - IS262	2014-2016	–	–	–	–	–	0	0	–	100	–	0	–	–	–
Iceland	Fish - IS174	2014-2016	–	–	–	–	–	10.95	0	–	89.05	–	0	–	–	–
Iceland	Fish - IS274	2014-2016	–	–	–	–	–	21.74	0	–	78.26	–	0	–	–	–
Iceland	Fish - IS279	2014-2016	–	–	–	–	–	30.17	0	–	42.9	–	26.93	–	–	–
Iceland	Fish - IS154	2014-2016	–	–	–	–	–	7.66	0	–	92.34	–	0	–	–	–
Iceland	Fish - IS271	2014-2016	–	–	–	–	–	28.31	0	–	60.24	–	11.45	–	–	–

Iceland	Fish - IS104	2014-2016	–	–	–	–	–	8.58	0	–	91.42	–	0	–	–	–
Faroe Islands	40888	2008	–	–	–	–	–	5.72	39.86	–	0	–	54.42	–	–	–
Faroe Islands	40889	2008	–	–	–	–	–	5.75	12.25	–	78.12	–	3.87	–	–	–
East Greenland	35143	2012-2014	–	9	–	0	–	42.28	–	0	0	–	36.14	0	0	12.58
East Greenland	38340	2012-2014	–	42.5	–	0	–	0	–	57.08	0	–	0	0.42	0	0
East Greenland	48335	2012-2014	–	4.95	–	0	–	13.55	–	26.76	0	–	25.57	0	0	29.17
East Greenland	48336	2012-2014	–	6.51	–	0	–	20.91	–	28.11	0	–	23.38	0	0	21.09
East Greenland	48337	2012-2014	–	9.29	–	9.02	–	0	–	81.7	0	–	0	0	0	0
East Greenland	48338	2012-2014	–	0	–	0	–	24.81	–	0	0	–	40.63	0	0	34.56
East Greenland	48339	2012-2014	–	0	–	0.82	–	0	–	87.36	0	–	3.68	8.15	0	0
East Greenland	48733	2012-2014	–	7.34	–	0	–	0	–	23.72	0	–	35.02	0	0	33.92
East Greenland	48735	2012-2014	–	19.71	–	16.21	–	0	–	64.08	0	–	0	0	0	0
East Greenland	48736	2012-2014	–	12.53	–	0	–	0	–	40.49	0	–	34.81	0	0	12.17
East Greenland	51601	2012-2014	–	15.69	–	6.28	–	0	–	44.55	0	–	19.14	0	0	14.35
East Greenland	51606	2012-2014	–	21.57	–	16.35	–	0	–	62.09	0	–	0	0	0	0
East Greenland	51607	2012-2014	–	26.32	–	2.24	–	0	–	31.84	14.14	–	25.45	0	0	0
East Greenland	51610	2012-2014	–	53.35	–	0	–	0	–	0	11.8	–	0	34.85	0	0

East Greenland	51613	2012-2014	–	1.55	–	2.08	–	0	–	85.83	0	–	10.55	0	0	0
East Greenland	GL1-EGR	2021	–	79.12	–	20.88	–	0	–	0	0	–	0	0	0	0
West Greenland	GL2-WGR	2021	–	23.4	–	0	–	0	–	70.19	0	–	6.4	0	0	0
East Greenland	GL3-EGR	2021	–	17.69	–	2.29	–	0	–	67.18	0	–	12.85	0	0	0
Canadian Arctic	ARPI_00_02_2019	2019	–	–	0	0	2.51	–	–	0	27.55	–	0	0	–	69.95
Canadian Arctic	ARPI_00_03_2019	2019	–	–	30.08	0	4.34	–	–	8.79	8.53	–	0	0	–	48.25
Canadian Arctic	ARPI_00_04_2019	2019	–	–	54.15	0	2.45	–	–	0	19.44	–	0	0	–	23.96
Canadian Arctic	ARPI_00_06_2019	2019	–	–	4.8	0	1.73	–	–	0	40.86	–	0	0	–	52.61
Canadian Arctic	ARPI_00_08_2019	2019	–	–	12.78	0	2.01	–	–	32.52	0	–	12.96	0	–	39.73
Canadian Arctic	ARPI_00_09_2019	2019	–	–	41.27	0	1.51	–	–	0	0	–	21.17	0	–	36.06
Canadian Arctic	ARPI_00_10_2019	2019	–	–	54.85	0	11.09	–	–	0	0	–	0	0	–	34.05
Canadian Arctic	ARPI_00_11_2019	2019	–	–	5.59	0	0	–	–	0	0	–	48.2	0	–	46.21
Canadian Arctic	ARRB-09-4001	2009	–	–	14.12	0	0	–	–	0	0	–	0	0	–	85.88
Canadian Arctic	KW 01-2018	2018	–	–	79.26	0	8.13	–	–	0	0	–	0	0	–	12.61
Canadian Arctic	KW 02-2018	2018	–	–	78.63	0	8.25	–	–	0	0	–	0	0	–	13.11
Canadian Arctic	KW 03-2018	2018	–	–	72.87	0	6.52	–	–	0	0	–	0	20.21	–	0.4
Canadian Arctic	KW 05-2018	2018	–	–	51.82	0	0	–	–	0	29.42	–	0	0	–	18.76

Canadian Arctic	KW 06-2018	2018	–	–	47.05	0	0	–	–	0	33.84	–	0	0	–	19.11
Canadian Arctic	KW 10-2018	2018	–	–	91.28	0	8.72	–	–	0	0	–	0	0	–	0
Canadian Arctic	KW 13-2018	2018	–	–	30.95	0	0	–	–	0	23.2	–	0	0	–	45.85
Canadian Arctic	KW 15-2018	2018	–	–	79.92	0	3.59	–	–	0	0	–	0	3.45	–	13.04
Canadian Arctic	KW 18-2018	2018	–	–	40.3	0	0	–	–	0	20.31	–	0	0	–	39.39
Canadian Arctic	KW4001-2013	2013	–	–	47.74	0	0	–	–	0	12.25	–	0	0	–	40.02
Canadian Arctic	KW4002-2013	2013	–	–	15	0	12.31	–	–	0	0	–	0	0	–	72.7
Canadian Arctic	KW4003-2013	2013	–	–	43.26	0	0.86	–	–	0	25.79	–	0	0	–	30.09
Canadian Arctic	KW4005-2013	2013	–	–	87.5	0	5.41	–	–	0	0	–	0	0	–	7.08
Canadian Arctic	KW4006-2013	2013	–	–	15.08	4.97	30.6	–	–	0	0	–	0	25	–	24.35
Canadian Arctic	KW4007-2013	2013	–	–	59.9	0	1.7	–	–	0	0	–	0	0	–	38.41
Canadian Arctic	KW4008-PG-2013	2013	–	–	39.16	0	1.69	–	–	0	0	–	0	0	–	59.15
Canadian Arctic	KWPG1-2020	2020	–	–	61.69	0	4.92	–	–	0	0	–	0	0	–	33.39
Canadian Arctic	KWPG10-2020	2020	–	–	19.29	0	7.3	–	–	0	0	–	0	0	–	73.41
Canadian Arctic	KWPG11-2020	2020	–	–	86.88	0	3.43	–	–	0	0	–	0	0	–	9.69
Canadian Arctic	KWPG12-2020	2020	–	–	71.56	0	4.56	–	–	0	0	–	0	0	–	23.88
Canadian Arctic	KWPG13-2020	2020	–	–	57.51	0	0	–	–	0	0	–	0	0	–	42.49

Canadian Arctic	KWPG15-2020	2020	–	–	83.82	0	0.74	–	–	0	0	–	0	0	–	15.44
Canadian Arctic	KWPG16-2020	2020	–	–	74.9	0	6.43	–	–	0	0	–	0	0	–	18.67
Canadian Arctic	KWPG17-2020	2020	–	–	46.01	0	1.96	–	–	0	0	–	0	0	–	52.03
Canadian Arctic	KWPG18-2020	2020	–	–	84.06	0	1.63	–	–	0	0	–	0	0	–	14.31
Canadian Arctic	KWPG19-2020	2020	–	–	57.46	0	0	–	–	0	0	–	0	0	–	42.54
Canadian Arctic	KWPG2-2020	2020	–	–	67.3	0	6.9	–	–	0	0	–	0	0	–	25.8
Canadian Arctic	KWPG20-2020	2020	–	–	68.64	0	3.9	–	–	0	0	–	0	0	–	27.46
Canadian Arctic	KWPG21-2020	2020	–	–	65.05	0	2.73	–	–	0	0	–	0	0	–	32.22
Canadian Arctic	KWPG22-2020	2020	–	–	51.11	0	0	–	–	0	0	–	0	0	–	48.89
Canadian Arctic	KWPG23-2020	2020	–	–	0	0	11.49	–	–	0	0	–	0	0	–	88.51
Canadian Arctic	KWPG24-2020	2020	–	–	56.63	0	2.18	–	–	0	0	–	0	0	–	41.19
Canadian Arctic	KWPG3-2020	2020	–	–	91.37	0	6.94	–	–	0	0	–	0	0	–	1.68
Canadian Arctic	KWPG4-2020	2020	–	–	67.79	0	4.05	–	–	0	0	–	0	0	–	28.15
Canadian Arctic	KWPG5-2020	2020	–	–	0	0	13.92	–	–	0	0	–	0	0	–	86.08
Canadian Arctic	KWPG6-2020	2020	–	–	52.04	0	6.67	–	–	0	0	–	0	0	–	41.29
Canadian Arctic	KWPG7-2020	2020	–	–	94.86	0	4.39	–	–	0	0	–	0	0	–	0.75
Canadian Arctic	KWPG8-2020	2020	–	–	50.7	0	11.31	–	–	0	0	–	0	0	–	37.99

Canadian Arctic	KWPG9-2020	2020	–	–	49.27	0	0	–	–	0	0	–	0	0	–	50.73
Canadian Arctic	KWPI10-2020	2020	–	–	42.92	0	8.81	–	–	0	0	–	0	0	–	48.27
Canadian Arctic	KWPI12-2020	2020	–	–	80.12	0	7.42	–	–	0	0	–	0	0	–	12.46
Canadian Arctic	KWPI13-2020	2020	–	–	43.88	0	3.23	–	–	0	0	–	0	0	–	52.89
Canadian Arctic	KWPI15-2020	2020	–	–	58.98	0	0	–	–	0	0	–	0	0	–	41.02
Canadian Arctic	KWPI2-2020	2020	–	–	89.43	0	0	–	–	0	0	–	0	0	–	10.57
Canadian Arctic	KWPI3-2020	2020	–	–	86.79	0	4.19	–	–	0	0	–	0	0	–	9.03
Canadian Arctic	KWPI5-2020	2020	–	–	31.11	0	3.92	–	–	0	0	–	0	0	–	64.97
Canadian Arctic	KWPI6-2020	2020	–	–	46.59	0	1.08	–	–	0	0	–	0	0	–	52.33
Canadian Arctic	KWPI7-2020	2020	–	–	0	0	0	–	–	0	0	–	0	0	–	100
Canadian Arctic	KWPI8-2020	2020	–	–	79.6	0	0	–	–	0	0	–	0	0	–	20.4
Saint-Pierre & Miquelon	SPMKW1	2019-2021	100	–	–	–	–	0	0	–	0	–	0	–	–	–
Saint-Pierre & Miquelon	SPMKW2	2019-2021	61.93	–	–	–	–	0	38.07	–	0	–	0	–	–	–
Saint-Pierre & Miquelon	SPMKW3	2019-2021	0	–	–	–	–	80.5	0	–	0	–	19.5	–	–	–
Saint-Pierre & Miquelon	SPMKW4	2019-2021	71.63	–	–	–	–	0	28.37	–	0	–	0	–	–	–
Saint-Pierre & Miquelon	SPMKW5	2019-2021	59.03	–	–	–	–	34.74	6.23	–	0	–	0	–	–	–

Table S5-4:QFASA modeling diagnostics: Leave_one_prey_out (LOPO) as the mean of correct species attribution and Pred_beyond_prey estimates for all regional groups of killer whales in our study.

Population	LOPO (%)	Pred_beyond_prey (%)
Norway	89.1	48.3
Iceland	88.9	53.1
Faroe	88.9	53.5
Greenland	79	27.1
Canada	77.3	37.2
Saint Pierre & Miquelon	82	49.6

Table S5-5:FA percentages (mean ± SE) in North Atlantic killer whales (n=150). Only FA above 0.1% are shown. Bold fatty acids are the dietary set used in the QFASA models (16:2n4,16:3n6, 16:4n3, 18:3n3, 18:4n3, 20:1n11, 20:1n9, 20:1n7, 20:4n6, 20:3n3, 20:4n3, 20:5n3, 22:1n11, 22:1n9, 22:6n3).

	Arctic Canada (n = 58)	Eastern Canada (n = 5)	Greenland (n = 19)	Iceland (n = 48)	Faroe Islands (n = 2)	Norway (n = 59)
Saturated FA						
12:0	1.10 ± 0.05	0.05 ± 1.34	1.34 ± 0.36	0.36 ± 0.63	0.63 ± 0.06	0.06 ± 0.87
13:0	0.16 ± 0.02	0.02 ± 0.09	0.09 ± 0.01	0.01 ± 0.05	0.05 ± 0.00	0.00 ± 0.11
Iso14	0.27 ± 0.05	0.05 ± 0.28	0.28 ± 0.04	0.04 ± 0.39	0.39 ± 0.14	0.14 ± 0.33
14:0	5.66 ± 0.12	0.12 ± 6.21	6.21 ± 0.69	0.69 ± 5.44	5.44 ± 0.15	0.15 ± 6.94
Iso15	0.65 ± 0.06	0.06 ± 0.46	0.46 ± 0.04	0.04 ± 0.91	0.91 ± 0.19	0.19 ± 0.64
Anti15	0.23 ± 0.02	0.02 ± 0.25	0.25 ± 0.02	0.02 ± 0.20	0.20 ± 0.03	0.03 ± 0.33
15:0	0.49 ± 0.01	0.01 ± 0.81	0.81 ± 0.37	0.37 ± 0.89	0.89 ± 0.08	0.08 ± 0.90
Iso16	0.23 ± 0.03	0.03 ± 0.20	0.20 ± 0.05	0.05 ± 0.64	0.64 ± 0.21	0.21 ± 0.34
16:0	5.57 ± 0.13	0.13 ± 5.39	5.39 ± 0.45	0.45 ± 5.17	5.17 ± 0.12	0.12 ± 5.81
7Me16:0	0.38 ± 0.01	0.01 ± 0.26	0.26 ± 0.02	0.02 ± 0.32	0.32 ± 0.01	0.01 ± 0.30
Iso17	0.25 ± 0.02	0.02 ± 0.06	0.06 ± 0.00	0.00 ± 0.09	0.09 ± 0.01	0.01 ± 0.14
17:0	0.29 ± 0.04	0.04 ± 1.25	1.25 ± 0.51	0.51 ± 0.96	0.96 ± 0.25	0.25 ± 0.98
18:0	1.22 ± 0.09	0.09 ± 1.09	1.09 ± 0.19	0.19 ± 0.84	0.84 ± 0.04	0.04 ± 1.48
∑SFA	16.51 ± 0.66	17.68 ± 2.80	16.53 ± 1.30	19.17 ± 0.90	19.35 ± 4.40	19.32 ± 0.70
Mono-Unsaturated FA						

14:1n9	0.89 ± 0.04	0.04 ± 1.23	1.23 ± 0.36	0.36 ± 0.73	0.73 ± 0.09	0.09 ± 0.91
14:1n7	0.85 ± 0.05	0.05 ± 1.26	1.26 ± 0.37	0.37 ± 0.65	0.65 ± 0.09	0.09 ± 0.56
14:1n5	3.57 ± 0.11	0.11 ± 3.67	3.67 ± 0.37	0.37 ± 3.34	3.34 ± 0.11	0.11 ± 5.71
15:1n6	0.18 ± 0.01	0.01 ± 0.16	0.16 ± 0.02	0.02 ± 0.18	0.18 ± 0.00	0.00 ± 0.26
16:1n11	0.60 ± 0.06	0.06 ± 1.29	1.29 ± 0.24	0.24 ± 1.06	1.06 ± 0.07	0.07 ± 1.67
16:1n9	3.07 ± 0.10	0.10 ± 3.42	3.42 ± 0.48	0.48 ± 2.15	2.15 ± 0.23	0.23 ± 1.75
16:1n7	22.72 ± 0.36	0.36 ± 20.63	20.63 ± 1.02	1.02 ± 22.76	22.76 ± 0.41	0.41 ± 17.33
17:1	0.64 ± 0.02	0.02 ± 1.22	1.22 ± 0.51	0.51 ± 0.49	0.49 ± 0.01	0.01 ± 0.47
18:1n11	4.61 ± 0.35	0.35 ± 5.31	5.31 ± 0.34	0.34 ± 4.78	4.78 ± 0.18	0.18 ± 6.69
18:1n9	26.90 ± 0.48	0.48 ± 24.80	24.80 ± 2.60	2.60 ± 23.79	23.79 ± 0.44	0.44 ± 17.87
18:1n7	3.17 ± 0.08	0.08 ± 2.86	2.86 ± 0.47	0.47 ± 3.29	3.29 ± 0.18	0.18 ± 2.09
18:1n5	0.25 ± 0.01	0.01 ± 0.22	0.22 ± 0.05	0.05 ± 0.26	0.26 ± 0.02	0.02 ± 0.21
20:1n11	2.88 ± 0.08	0.08 ± 2.80	2.80 ± 0.29	0.29 ± 2.04	2.04 ± 0.06	0.06 ± 5.19
20:1n9	3.39 ± 0.11	0.11 ± 3.16	3.16 ± 0.68	0.68 ± 3.39	3.39 ± 0.26	0.26 ± 4.52
20:1n7	0.29 ± 0.02	0.02 ± 0.15	0.15 ± 0.03	0.03 ± 0.12	0.12 ± 0.01	0.01 ± 0.12
22:1n11	1.98 ± 0.08	0.08 ± 1.66	1.66 ± 0.20	0.20 ± 1.26	1.26 ± 0.08	0.08 ± 6.15
22:1n9	0.28 ± 0.01	0.01 ± 0.19	0.19 ± 0.03	0.03 ± 0.17	0.17 ± 0.01	0.01 ± 0.37
24:1n9	0.20 ± 0.01	0.01 ± 0.09	0.09 ± 0.02	0.02 ± 0.08	0.08 ± 0.02	0.02 ± 0.19
∑MUFA	76.48 ± 1.99	74.14 ± 8.07	70.54 ± 2.28	72.06 ± 2.51	63.86 ± 8.45	68.02 ± 2.03
Poly-Unsaturated FA						
16:2n4	0.65 ± 0.03	0.03 ± 0.54	0.54 ± 0.03	0.03 ± 0.31	0.31 ± 0.01	0.01 ± 0.33
16:3n6	0.47 ± 0.01	0.01 ± 0.49	0.49 ± 0.08	0.08 ± 0.60	0.60 ± 0.02	0.02 ± 0.52
16:3n4	0.19 ± 0.03	0.03 ± 0.10	0.10 ± 0.06	0.06 ± 0.10	0.10 ± 0.00	0.00 ± 0.04
16:3n1	0.20 ± 0.02	0.02 ± 0.23	0.23 ± 0.05	0.05 ± 0.39	0.39 ± 0.02	0.02 ± 0.69
16:4n3	0.17 ± 0.01	0.01 ± 0.12	0.12 ± 0.02	0.02 ± 0.15	0.15 ± 0.01	0.01 ± 0.16
18:2n6	1.25 ± 0.03	0.03 ± 1.23	1.23 ± 0.18	0.18 ± 1.66	1.66 ± 0.05	0.05 ± 1.12
18:3n4	0.16 ± 0.01	0.01 ± 0.13	0.13 ± 0.01	0.01 ± 0.14	0.14 ± 0.01	0.01 ± 0.14
18:3n3	0.99 ± 0.18	0.18 ± 3.46	3.46 ± 1.31	1.31 ± 5.02	5.02 ± 0.62	0.62 ± 3.42
18:3n1	0.33 ± 0.04	0.04 ± 0.14	0.14 ± 0.09	0.09 ± 1.19	1.19 ± 0.13	0.13 ± 0.04
18:4n3	0.12 ± 0.01	0.01 ± 0.06	0.06 ± 0.02	0.02 ± 0.29	0.29 ± 0.03	0.03 ± 0.17
20:4n6	0.31 ± 0.03	0.03 ± 0.26	0.26 ± 0.09	0.09 ± 0.23	0.23 ± 0.03	0.03 ± 0.30
20:3n3	0.16 ± 0.02	0.02 ± 0.11	0.11 ± 0.06	0.06 ± 0.19	0.19 ± 0.03	0.03 ± 0.30
20:4n3	0.16 ± 0.01	0.01 ± 0.11	0.11 ± 0.03	0.03 ± 0.24	0.24 ± 0.03	0.03 ± 0.18
20:5n3	0.65 ± 0.04	0.04 ± 0.47	0.47 ± 0.17	0.17 ± 1.01	1.01 ± 0.17	0.17 ± 0.61
22:5n3	0.58 ± 0.04	0.04 ± 0.34	0.34 ± 0.14	0.14 ± 0.58	0.58 ± 0.10	0.10 ± 0.24

22:6n3	0.65 ± 0.04	0.04 ± 0.40	0.40 ± 0.17	0.17 ± 0.83	0.83 ± 0.12	0.12 ± 0.51
∑PUFA	7.10 ± 0.20	8.10 ± 2.60	12.90 ± 1.10	8.60 ± 0.90	17.10 ± 4.90	12.70 ± 0.70

Table S5-6: FA percentages (mean ± SE) in Icelandic prey including harbor seals (n=14), Atlantic herring (n=10) and Atlantic mackerel (n=10), and in Norwegian lumpfish roe (n=4). Only FA above 0.1% are shown. Bold fatty acids are the dietary set used in the QFASA models.

	Harbor seal (n = 14)	Herring Iceland (n = 10)	Mackerel Iceland (n = 19)	Herring Norway (n = 48)	Lumpfish Norway (n = 2)	Mackerel Norway (n = 59)
Saturated FA						
12:0	0.20 ± 0.01	0.01 ± 0.09	0.09 ± 0.00	0.00 ± 0.08	0.08 ± 0.00	0.00 ± 0.09
13:0	0.02 ± 0.00	0.00 ± 0.04	0.04 ± 0.00	0.00 ± 0.05	0.05 ± 0.00	0.00 ± 0.05
Iso14	0.04 ± 0.00	0.00 ± 0.03	0.03 ± 0.00	0.00 ± 0.03	0.03 ± 0.00	0.00 ± 0.03
14:0	4.96 ± 0.09	0.09 ± 7.32	7.32 ± 0.38	0.38 ± 7.68	7.68 ± 0.27	0.27 ± 7.71
Iso15	0.18 ± 0.01	0.01 ± 0.23	0.23 ± 0.02	0.02 ± 0.23	0.23 ± 0.01	0.01 ± 0.28
Anti15	0.09 ± 0.01	0.01 ± 0.08	0.08 ± 0.01	0.01 ± 0.08	0.08 ± 0.00	0.00 ± 0.09
15:0	0.27 ± 0.01	0.01 ± 0.39	0.39 ± 0.03	0.03 ± 0.53	0.53 ± 0.01	0.01 ± 0.47
Iso16	0.08 ± 0.00	0.00 ± 0.09	0.09 ± 0.01	0.01 ± 0.09	0.09 ± 0.00	0.00 ± 0.08
16:0	11.41 ± 0.25	0.25 ± 12.61	12.61 ± 0.73	0.73 ± 13.53	13.53 ± 0.43	0.43 ± 13.45
7Me16:0	0.30 ± 0.01	0.01 ± 0.22	0.22 ± 0.01	0.01 ± 0.31	0.31 ± 0.01	0.01 ± 0.32
Iso17	0.10 ± 0.01	0.01 ± 0.06	0.06 ± 0.01	0.01 ± 0.08	0.08 ± 0.00	0.00 ± 0.06
17:0	0.12 ± 0.00	0.00 ± 0.18	0.18 ± 0.01	0.01 ± 0.36	0.36 ± 0.02	0.02 ± 0.17
18:0	1.03 ± 0.03	0.03 ± 1.32	1.32 ± 0.08	0.08 ± 2.40	2.40 ± 0.16	0.16 ± 1.21
∑SFA	18.78 ± 0.42	22.67 ± 1.30	25.43 ± 0.92	24.00 ± 0.44	23.39 ± 1.31	25.15 ± 1.86
Mono-Unsaturated FA						
14:1n9	0.03 ± 0.00	0.00 ± 0.00	0.03 ± 0.00	0.04 ± 0.01	0.01 ± 0.00	0.06 ± 0.01
14:1n7	0.08 ± 0.00	0.27 ± 0.02	0.19 ± 0.01	0.02 ± 0.00	0.00 ± 0.00	0.03 ± 0.00
14:1n5	0.14 ± 0.01	0.03 ± 0.00	0.03 ± 0.00	0.12 ± 0.00	0.02 ± 0.00	0.08 ± 0.00
15:1n6	0.09 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.00 ± 0.00	0.01 ± 0.00
16:1n11	0.55 ± 0.02	0.35 ± 0.02	0.68 ± 0.06	0.38 ± 0.01	0.46 ± 0.07	0.64 ± 0.05
16:1n9	0.63 ± 0.02	0.17 ± 0.01	0.24 ± 0.01	0.19 ± 0.00	0.42 ± 0.01	0.26 ± 0.02
16:1n7	24.60 ± 1.15	4.14 ± 0.13	3.41 ± 0.17	4.52 ± 0.10	1.26 ± 0.15	3.32 ± 0.11
17:1	0.31 ± 0.01	0.25 ± 0.02	0.36 ± 0.01	0.32 ± 0.01	0.31 ± 0.04	0.40 ± 0.01
18:1n11	3.35 ± 0.17	0.97 ± 0.05	0.52 ± 0.04	0.63 ± 0.02	4.26 ± 0.21	0.47 ± 0.05
18:1n9	14.13 ± 0.38	9.39 ± 0.84	9.71 ± 0.96	10.14 ± 0.40	13.66 ± 0.38	11.00 ± 2.39

18:1n7	3.42 ± 0.12	2.26 ± 0.23	1.98 ± 0.17	1.45 ± 0.05	3.37 ± 0.34	2.31 ± 0.44
18:1n5	0.38 ± 0.00	0.29 ± 0.02	0.32 ± 0.01	0.44 ± 0.01	0.61 ± 0.04	0.36 ± 0.02
20:1n11	1.59 ± 0.09	2.05 ± 0.13	0.81 ± 0.04	1.31 ± 0.05	0.51 ± 0.02	0.69 ± 0.06
20:1n9	6.13 ± 0.28	11.39 ± 0.58	8.72 ± 0.49	11.90 ± 0.31	3.11 ± 0.16	8.26 ± 0.61
20:1n7	0.27 ± 0.02	0.21 ± 0.02	0.20 ± 0.01	0.19 ± 0.01	0.37 ± 0.04	0.18 ± 0.01
22:1n11	1.69 ± 0.07	23.18 ± 1.50	13.96 ± 0.82	18.62 ± 0.54	0.80 ± 0.06	11.62 ± 1.41
22:1n9	0.28 ± 0.01	0.88 ± 0.05	0.80 ± 0.03	0.94 ± 0.05	0.46 ± 0.02	0.83 ± 0.04
24:1n9	0.11 ± 0.02	0.81 ± 0.03	0.79 ± 0.04	0.88 ± 0.02	0.20 ± 0.02	0.75 ± 0.06
∑MUFA	57.78 ± 2.38	56.64 ± 3.65	42.75 ± 2.87	52.11 ± 1.60	29.80 ± 1.56	41.24 ± 5.28
Poly-Unsaturated FA						
16:2n4	0.65 ± 0.03	0.03 ± 0.54	0.54 ± 0.03	0.03 ± 0.31	0.31 ± 0.01	0.01 ± 0.33
16:3n6	0.47 ± 0.01	0.01 ± 0.49	0.49 ± 0.08	0.08 ± 0.60	0.60 ± 0.02	0.02 ± 0.52
16:3n4	0.19 ± 0.03	0.03 ± 0.10	0.10 ± 0.06	0.06 ± 0.10	0.10 ± 0.00	0.00 ± 0.04
16:3n1	0.20 ± 0.02	0.02 ± 0.23	0.23 ± 0.05	0.05 ± 0.39	0.39 ± 0.02	0.02 ± 0.69
16:4n3	0.17 ± 0.01	0.01 ± 0.12	0.12 ± 0.02	0.02 ± 0.15	0.15 ± 0.01	0.01 ± 0.16
18:2n6	1.25 ± 0.03	0.03 ± 1.23	1.23 ± 0.18	0.18 ± 1.66	1.66 ± 0.05	0.05 ± 1.12
18:3n4	0.16 ± 0.01	0.01 ± 0.13	0.13 ± 0.01	0.01 ± 0.14	0.14 ± 0.01	0.01 ± 0.14
18:3n3	0.99 ± 0.18	0.18 ± 3.46	3.46 ± 1.31	1.31 ± 5.02	5.02 ± 0.62	0.62 ± 3.42
18:3n1	0.33 ± 0.04	0.04 ± 0.14	0.14 ± 0.09	0.09 ± 1.19	1.19 ± 0.13	0.13 ± 0.04
18:4n3	0.12 ± 0.01	0.01 ± 0.06	0.06 ± 0.02	0.02 ± 0.29	0.29 ± 0.03	0.03 ± 0.17
20:4n6	0.31 ± 0.03	0.03 ± 0.26	0.26 ± 0.09	0.09 ± 0.23	0.23 ± 0.03	0.03 ± 0.30
20:3n3	0.16 ± 0.02	0.02 ± 0.11	0.11 ± 0.06	0.06 ± 0.19	0.19 ± 0.03	0.03 ± 0.30
20:4n3	0.16 ± 0.01	0.01 ± 0.11	0.11 ± 0.03	0.03 ± 0.24	0.24 ± 0.03	0.03 ± 0.18
20:5n3	0.65 ± 0.04	0.04 ± 0.47	0.47 ± 0.17	0.17 ± 1.01	1.01 ± 0.17	0.17 ± 0.61
22:5n3	0.58 ± 0.04	0.04 ± 0.34	0.34 ± 0.14	0.14 ± 0.58	0.58 ± 0.10	0.10 ± 0.24
22:6n3	0.65 ± 0.04	0.04 ± 0.40	0.40 ± 0.17	0.17 ± 0.83	0.83 ± 0.12	0.12 ± 0.51
∑PUFA	21.11 ± 0.75	18.63 ± 1.47	29.10 ± 1.07	21.86 ± 0.64	45.23 ± 1.61	31.04 ± 2.02

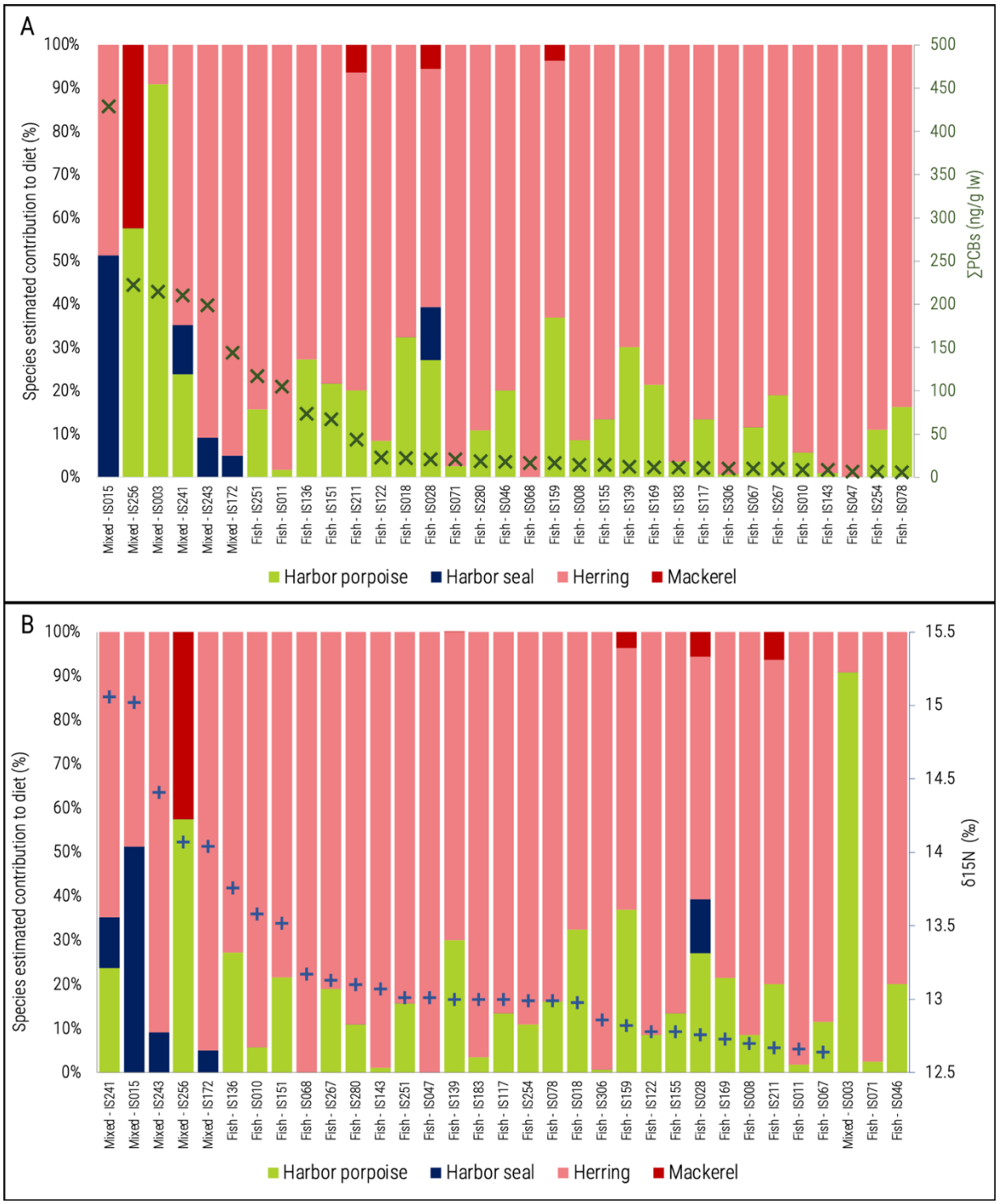

Figure S5-1:Relationship between A) contaminants (∑PCBs data from Remili et al., 2021) and diet estimates in Icelandic male killer whales (n=33) and B) δ15N values and diet estimates in the same whales (δ15N data from Samarra et al., 2017). PCBs, δ15N, and fatty acids measurements were performed on the same skin and blubber biopsies, thus allowing an accurate comparison of the three measurements. The colored bars represent the proportion of prey species in the diet, the green crosses represent the ∑PCBs in ng/g lipid weight (lw), and the blue plus signs represent the δ15N values (‰).

CONNECTING PARAGRAPH

Having successfully unraveled the mystery surrounding killer whales' diets across the entire ocean, my next objective is to model the implications of these dietary habits on the risks associated with persistent organic pollutants, specifically polychlorinated biphenyls. This chapter represents the culmination of four and a half years of dedicated and intensive research, standing as one of my most gratifying accomplishments. Right from the outset, this study sparked a strong sense of enthusiasm within me, even before I began my PhD journey. Our results provide clear and compelling evidence that dietary habits play a pivotal role in shaping the variations of contaminants observed in North Atlantic killer whales. Furthermore, killer whales' predation of marine mammals exposes them to contamination levels that far exceed established thresholds for reproductive failure in marine mammal populations. While these thresholds may vary among species and may not be universally precise, our calculated risk levels for killer whales relying on marine mammal prey, including those with mixed diets of marine mammals and fish, indicate significant health risks. These risks pose a potential threat to the future well-being and sustainability of some North Atlantic killer whale populations. This chapter is published in *Environmental Science and Technology* and has twelve coauthors, myself included.

6 CHAPTER SIX: VARYING DIET COMPOSITION CAUSES STRIKING DIFFERENCES IN LEGACY AND EMERGING CONTAMINANT CONCENTRATIONS IN KILLER WHALES ACROSS THE NORTH ATLANTIC

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6.1 ABSTRACT

Lipophilic persistent organic pollutants (POPs) tend to biomagnify in food chains, resulting in higher concentrations in species such as killer whales (*Orcinus orca*) feeding on marine mammals compared to those consuming fish. Advancements in dietary studies include the use of quantitative fatty acid signature analysis (QFASA) and the differentiation of feeding habits within and between populations of North Atlantic (NA) killer whales. This comprehensive study assessed the concentrations of legacy and emerging POPs in 162 killer whales from across the NA. We report significantly higher mean levels of polychlorinated biphenyls (PCBs), organochlorine pesticides, and flame retardants in Western NA killer whales compared to eastern NA conspecifics. PCBs ranged from ~100 mg/kg lipid weight (lw) in the Western NA (Canadian Arctic, Eastern Canada) to ~50 mg/kg lw in the mid-NA (Greenland, Iceland), to ~10 mg/kg lw in the Eastern NA (Norway, Faroe Islands). The observed variations in contaminant levels were strongly correlated with diet compositions across locations (inferred from QFASA), emphasizing the crucial role of feeding habits in assessing contaminant-associated health risks. These findings highlight the urgency for implementing enhanced measures to safely dispose of POP-contaminated waste, prevent further environmental contamination, and mitigate the release of newer and potentially harmful contaminants.

Keywords: Orcinus orca, biomagnification, PCBs, health risks, top predator, diet specialization, blubber, marine mammals

6.2 SYNOPSIS

Throughout the North Atlantic, killer whales feeding on dolphins and seals have higher contaminant concentrations than fish-feeding individuals, thereby increasing contaminant-associated health risks for these apex predators.

6.3 GRAPHICAL ABSTRACT

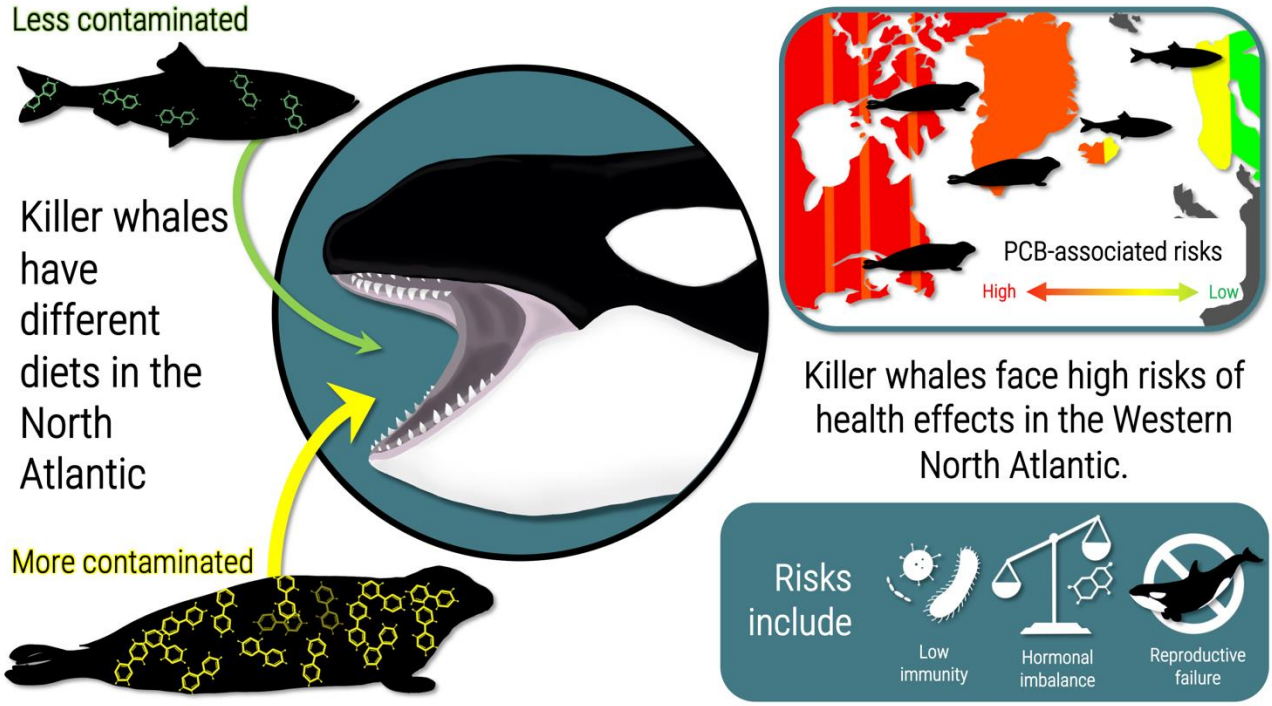

Figure 6-1:Graphical Abstract (Illustrations by A. Remili)

6.4 INTRODUCTION

Recent studies have highlighted the critical threat posed by consistently high concentrations of persistent organic pollutants (POPs) in at least some populations of killer

whales (Desforges et al., 2018; Hall et al., 2018) (*Orcinus orca*), due to their high trophic positions (R. Dietz et al., 2019) and limited biotransformation and elimination capacities (McKinney et al., 2011a; Meyer et al., 2018). As the ocean's ultimate apex predators, killer whales from certain populations exhibit among the highest POP concentrations in the animal kingdom (R. Dietz et al., 2019; Lawson et al., 2020; Lee et al., 2023). High levels of legacy contaminants like polychlorinated biphenyl (PCBs), organochlorines (OCs), polybrominated diphenyl ethers (PBDEs) and emerging flame retardants (FRs) have been associated with health issues that include increased risks of altered immune, endocrine, and reproductive functions in marine mammals, as well as carcinogenicity (Desforges et al., 2016; R. Dietz et al., 2019; Mos et al., 2006). Toxicity effects from PCB exposure are estimated to occur past the 9 mg/kg lipid weight (lw) threshold in marine mammals and reach a high risk of reproductive failure past 41 mg/kg lw (R. Dietz et al., 2019; Helle et al., 1976; Jepson et al., 2016; Kannan et al., 2000). While the thresholds for risks of health effects have not been established for other POP classes in marine mammals, previous *in vitro* research using killer whale and polar bear (*Ursus maritimus*) immune cells suggest that the immunotoxic effects of POP mixtures are greater than for a single compound (Desforges et al., 2017). Thus, there is a need to assess both the levels, toxicity, and risks caused by POPs for marine mammal populations around the globe.

Previous studies have demonstrated the importance of diet in the accumulation of POPs in killer whales (Krahn et al., 2007; Krahn et al., 2008; Ross et al., 2000a). Indeed, the lipophilic legacy POPs and legacy (e.g., PBDEs) and emerging FRs have tendencies to biomagnify, i.e., to increase in concentration with each trophic position. Thus, individuals feeding on high-trophic marine mammals such as pinnipeds and cetaceans may accumulate levels of contaminants putting them at higher risk compared to their conspecifics primarily feeding on fish (Lawson et al., 2020). Individual variation in diet can also occur within a population. Recent studies

measuring POPs (including legacy and emerging classes) in killer whales from Norway and Iceland reported high PCB levels for mixed-diet individuals, i.e., those feeding on marine mammals and fish, as opposed to those only known to feed on herring (*Clupea harengus*) (Andvik et al., 2020; Remili et al., 2021). In both populations, levels of PCBs in mixed-diet individuals were typically above the maximum threshold for risks of health effects. Conversely, fish-eating individuals had PCB concentrations associated with low risk for health effects.

Although tissue concentrations of legacy POPs and their relationship with diet habits are well documented for Northeast Pacific killer whales (Herman et al., 2005; Krahn, 2006; Lee et al., 2023), considerably less is known for North Atlantic (NA) conspecifics and on emerging POPs (E. Jourdain et al., 2019). POP concentrations were high in killer whales sampled in Greenland, while they remained lower in those sampled in the Eastern NA (Iceland and Norway) (Andvik et al., 2020; Pedro et al., 2017; Remili et al., 2021). Yet, POP concentrations remain unknown in killer whales from the Western NA, including in the Eastern Canadian Arctic and Eastern Canada. Few studies reported moderate to high levels of POPs (~2 mg/kg lw to ~10 mg/kg lw for ∑PCBs) in these regions for other marine mammals (Noël et al., 2018; Simond et al., 2020). As a result of biomagnification, marine mammal-eating killer whales could be exposed to high levels of contaminants in the Eastern Canadian Arctic and Eastern Canada, although this remains an understudied area requiring further investigation.

Although POP concentrations are influenced by feeding habits, quantitative estimates of killer whale diet composition, especially in remote areas of the NA Ocean, were not available until recently. The recent use of quantitative fatty acid signature analysis (QFASA) on ~200 NA killer whales spanning from Eastern Canada to Norway revealed important differences in their diet between and within populations (Remili et al., 2023). The diet estimates obtained in Remili et al. 2023 revealed that killer whales sampled in the Eastern NA feed on a high proportion of

herring, while mid-NA killer whales feed on a mixture of Arctic seals and mackerel (*Scomber scombrus*) and Western NA killer whales prey largely on marine mammals such as baleen whales and porpoises (*Phocoena phocoena*) in Eastern Canada, and belugas (*Delphinapterus leucas*), narwhals (*Monodon monoceros*), and ringed seals (*Pusa hispida*) in the Canadian Arctic (Remili et al., 2023). Nonetheless, these estimates calculated for each individual showed some marked differences among individual killer whales within populations. For example, all Arctic Canadian killer whales fed mainly on cetaceans or ringed seals, while Greenlandic killer whales showed more generalist feeding patterns that may suggest opportunistic foraging. Killer whales sampled in Norway and Iceland generally showed a strong preference for herring, but some individuals also consumed porpoises or seals around Iceland, and seals or lumpfish (*Cyclopterus lumpus*) in Norway. These inter and intra-population differences may result in different POP exposure and associated risks in killer whales across the NA.

Here, we first compare legacy and new POP concentrations (PCBs, OCs, PBDEs and non-BDE FRs) in 162 individuals across the NA, including, for the first time, Western NA killer whales. To our knowledge, this represents the largest NA killer whale contaminant dataset to date. We then assess the relationship between POP concentrations, sex and diet composition, using diet estimates previously inferred from QFASA for the same individuals (Remili et al., 2023). Finally, we assess the risks associated with PCBs for all individuals sampled across the NA Ocean, depending on their sex and diet types.

6.5 MATERIAL AND METHODS

6.5.1 Sampling

Killer whale blubber samples were collected from 162 individuals across the NA (details can be found in Table S6-1, S6-6). Thirty killer whales were sampled in the Eastern Canadian Arctic (Pond Inlet and Pangnirtung from 2013 to 2019), five in Eastern Canada (off the French

territory of Saint-Pierre & Miquelon and Newfoundland from 2019 to 2022), nineteen in Greenland (Tasiilaq, Scoresby Sund and Nuuk from 2012 to 2021), two in the Faroe Islands (2008), forty-eight in Iceland (Vestmannaeyjar and Grundarfjörður from 2014 to 2016) and fifty-eight from Norway (Skjervøy from 2017 to 2019). Sampling was performed via dart biopsies in the Canadian Arctic, Eastern Canada, Iceland and Norway. Briefly, skin and blubber biopsies were collected from free-ranging killer whales using an ARTS pneumatic darting system (LKARTS-Norway, Norway) or a crossbow and stainless-steel biopsy tips (CetaDart, Denmark) ranging from 25 × 7 mm to 40 x 5 mm, depending on the location. In Greenland, blubber samples were collected from individuals after subsistence-harvest, and in the Faroe Islands, samples were collected from two stranded individuals. All samples were stored at -80 °C until analysis. Upon arrival in their respective extraction laboratories, samples were cut in half longitudinally: one half was used for fatty acid analysis (Remili et al., 2023), while the other half was kept for contaminant analyses. Contaminant analyses could only be performed when the sample weight was sufficient for both analyses. Sexing and age class was assessed in the field thanks to photo-identification or detailed field observation (in Iceland and Norway), directly on the animal when harvested or stranded (in Greenland and the Faroe Islands) or genetically (for Western NA individuals, as part of the federal Canadian Department of Fisheries and Oceans's usual monitoring routine).

6.5.2 POPs analyses

POPs were extracted and quantified in four different laboratories (see Table S6-2), with each laboratory having slight variations in the suite of target compounds. We thus only reported concentrations for those compounds analyzed in all four different laboratories. This included thirty PCB congeners, and seventeen OCs, and for a subset of individuals, brominated and non-brominated flame retardants (twenty-four BDE and twenty-one non-BDE FRs) in the killer

whale biopsies (see supplementary text for the detailed list of targeted compounds and methods). Concentrations of POPs are reported in mg/kg (ppm) of lipid weight (lw) and sums for each contaminant class were calculated including only the compounds analyzed in all four labs. Given the small sample weights for the biopsies, we were unable to perfom interlab comparisons. However, Pedersen et al. re-extracted in our laboratory (McGill) PCB and OC compounds in the subsistence-harvested Greenlandic killer whales' blubber previously analyzed by Pedro et al., using the QuEChERS method and reported no significant differences between the two extraction methods, and the two laboratory analyses (Pedersen et al., 2023; Pedro et al., 2017). Additionally, while an interlaboratory difference might result in a small bias in the contaminant concentrations, the killer whale blubber showed orders of magnitude variation, which is well beyond what might be expected from interlaboratory differences (Pedersen et al., 2023). Therefore, any potential minor bias should not lead to a significant influence on the results or interpretation. Details on each procedure, and instrument analyses can be found in the SI.

6.5.3 QA/QC

The standard reference materials (NIST 1945 "pilot whale blubber" or 1946 "Great Lakes fish homogenate") were run with each batch of ten samples and checked for precision and accuracy. Accuracies for each laboratory can be found in Table S2. Method limits of detection (MLODs) and quantification (MLOQs) were defined as the minimum amount of analyte which produced a peak with a signal-to-noise ratio of 3 and 10, respectively. A procedural blank was run with each batch as well. Only a small contamination of heptachlor epoxide was reported for the Greenlandic samples (see SI for details) (Pedro et al., 2017). For these, the blank concentrations were subtracted from the sample concentrations. Recoveries for spiked internal standards (^{13}C-labelled compounds) are reported in Table S6-2.

6.5.4 Fatty acid analyses and QFASA

All fatty acid data was obtained from Remili et al. (2023) and can be found on the Polar Data Catalogue: https://doi.org/10.21963/13299. Fatty acid analyses were performed on the same individuals as previously described (Bourque et al., 2018). QFASA diet estimates representing the estimated percentage of each prey species from the prey library in the diet of each predator were obtained using the *QFASAR* package in R (Bromaghin, 2017). To calculate the diet estimates, killer whale calibration coefficients were used and developed by Remili et al. (2022) as well as 900+ prey in the prey library as described earlier (Remili et al., 2022; Remili et al., 2023). Model diagnostics were validated using the *leave_one_prey_out* and the *prey_beyond_pred* functions of the *QFASAR* package.

6.5.5 Statistical analyses

All statistical analyses were performed in R (version 4.2.3). The five main contaminant classes, i.e., ∑PCBs, dichlorodiphenyltrichloroethane (∑DDTs), chlordane (∑CHLs), and chlorobenzenes (∑ClBz) were quantified in all samples, while ∑HCHs were detected in > 90% of the samples. Contaminant concentrations were log-transformed (log $x+1$) to improve normality which was evaluated and confirmed with qqplots on residuals and/or Shapiro-Wilk tests. Any non-detects (N.D.) were assigned a random value between 0 and the MLOD of the compound before inferential statistical analysis. Compounds detected, but below the MLOQ, were assigned a random value between MLOD and MLOQ.

Before applying statistical tests and GLM models on our dataset, we had to remove certain individuals for the datasets prior to analyses. In Norway, eleven individuals had to be excluded from modelling because their sex could not be identified (they were identified in the

field as “females or juveniles”). We also had to remove the two Faroese females, since they were under the minimum number for statistical analyses. Two extra individuals in Norway, one in Eastern Canada, and one in the Eastern Canadian Arctic had to be removed because their sex was unknown. Finally, six individuals were removed from Eastern Canadian Arctic, and one from Iceland due to no diet estimates being available for these individuals, bringing the total number of individuals included in the statistical analyses to 138. Because the diet varied significantly across the NA (Remili et al., 2023), we first tested the impact of *location* on POP class concentrations through ANOVAs and *post-hoc* Tukey tests. This analysis could only be done in Norway and Iceland for ∑PBDEs and ∑non-BDE FRs because they were the only locations where we had a sufficient number of sampled individuals.

We employed generalized linear models to examine the influence of multiple factors contributing to the variability in PCB and OC classes among killer whales in the NA Ocean. The following variables were considered to determine the strongest influence on variations in the log-transformed concentrations of ∑PCBs, ∑DDTs, ∑CHLs, ∑ClBz, and ∑HCHs: *location, sex/age class* (adult males, adult females, and juveniles), and *diet-type*. To investigate how diet influenced POP concentrations within and among killer whale groups, we first separated the individuals into feeding types. This categorization was necessary because QFASA estimates for each prey species are interdependent and would violate model assumptions. Consequently, we separated our individuals into a “fish-dominant”, “mixed-diet” (i.e.: mix of marine mammals and fish), “pinniped-dominant”, “baleen whale-dominant” and “toothed whale-dominant” feeding types, based on their QFASA estimates. Mixed-diet individuals (i.e.: fish and marine mammals) were classified as such if their percentage of fish was < 65% and their marine mammal percentage was > 35%. In the Canadian Arctic, individuals with toothed whale percentages > 50 % were identified as toothed whale-dominant, while individuals with > 50 % of pinnipeds in

their diets were identified as “pinniped-dominant”. The exact diet composition of each individual included in this analysis can be found in the SI of Remili et al. (2023) and the diet-type for each individual can be found in Table S6-6 of this present study. To prevent overparameterization of the models, we did not test any interactions between variables. We utilized the Akaike information criterion corrected for small sample sizes (AICc) scores to select the most appropriate models. When multiple models had a difference in AICc (ΔAICc) of less than 4, we averaged all the models with a ΔAICc of 4 or lower, to obtain an average effect for each variable (Tables S6-4 and S6-5). For ∑PBDEs and ∑non-BDE FRs, we resorted to ANOVAs and *post hoc* Tukey tests to investigate the impact of sex/age and diet-type on killer whales sampled in Norway and Iceland.

We then visualized how PCB and OC profiles of compounds that were detected in > 70 % of the individuals varied by dietary habits, by computing a principal component analysis (PCA) on the scaled percentage contaminant concentrations, as described previously (Remili et al., 2021). For FRs, since the detection percentages were lower, we included the compounds detected in > 50 % of the individuals. The 27 legacy compounds included in this analysis were: Hexachlorobenzene, Oxychlordane, *cis*-Chlordane, *trans*-Nonachlor, *cis*-Nonachlor, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT, Heptachlor Epoxide, Dieldrin, CB-52, -74, -95, -99, -101, -105, -118, -138, -149, -151, -153, -156, -158, -170, -180, -183 and -187. The eight FR compounds included BDE-47, -85/-155, -99, -100, -153 and -154, BB-153, and α-HBCDD. PCAs were also computed on the log-transformed PCB, OC and FR concentrations (not the profiles) to test for visual intra-population variations (Fig. S2).

Finally, to estimate the risks associated with ∑PCBs, we calculated the risk quotient (RQ; RQ = Body Residue /Critical Body Residue) for each individual in this trans-Atlantic study, based on a conservative 10 mg/kg lw critical body residue, as previously described (Dietz et al.,

2015; R. Dietz et al., 2019). This threshold, established by Dietz et al. (2019) considers immunotoxic effects as well as endocrine disrupting effects, which also corresponds to the upper limit reported as the immune threshold modelled for cetaceans (Desforges et al., 2016). Hence, if future studies reveal lower critical daily doses, it is likely that the RQs observed in this study would be higher.

6.6 RESULTS AND DISCUSSION

6.6.1 Concentrations of persistent organic contaminants in NA killer whales

This study is the most comprehensive assessment of legacy and emerging contaminant concentrations in killer whale across the NA. Mean concentrations of PCBs ranged from a high of ~ 100 mg/kg lw in the Western NA (mean: 92.0 ± 9.8 mg/kg lw in the Canadian Arctic; 106.1 ± 31.1 mg/kg lw in Eastern Canada) to about 50 mg/kg in the mid NA (mean: 66.1 ± 10.6 mg/kg lw in Greenland; 42.1 ± 11.1 mg/kg lw in Iceland) to lower levels in the Eastern NA (mean: 2.9 ± 1.5 mg/kg lw in the Faroe Islands; 12.2 ± 10.8 mg/kg lw in Norway). Killer whales sampled in the Eastern Canadian Arctic had mean ∑PCB concentrations 7-fold higher than killer whales sampled in Norway, while the difference for ∑DDTs and ∑CHLs were 16-fold and 32-fold between the same two populations (Fig. 6-2, Table S6-3). A notable observation was the prevalence of higher DDT concentrations surpassing PCB concentrations in the Eastern Canadian Arctic (mean: 108.1 mg/kg lw for DDTs), whereas DDT levels were lower than PCBs in other regions and significantly lower in Iceland and Norway (Fig. 6-2).

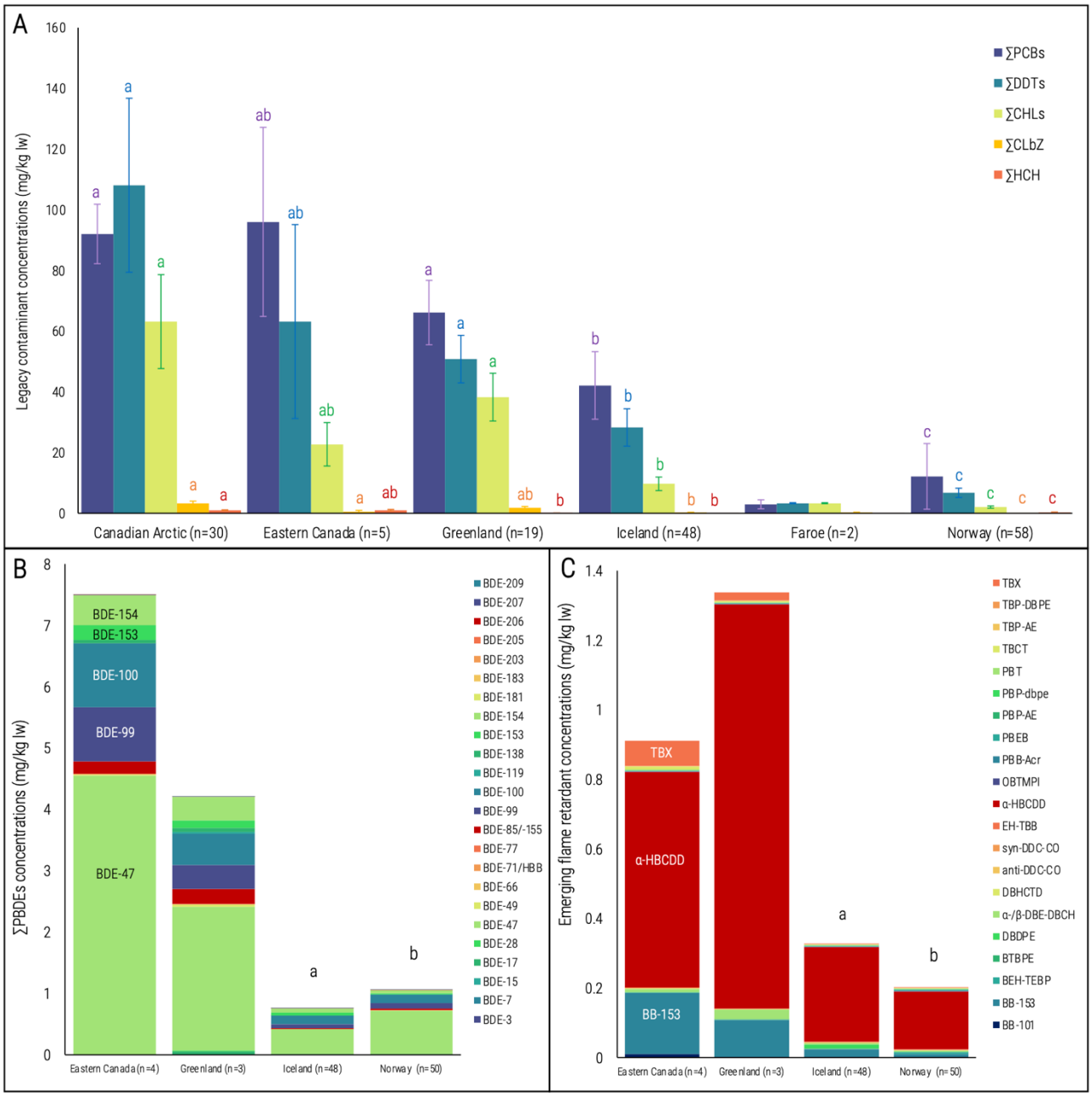

Figure 6-2: Total concentrations of mean (±SE) A) legacy polychlorinated biphenyls (PCB) and organochlorine contaminant classes (mg/kg lw), B) Polybrominated diphenyl ethers (PBDEs) (in mg/kg lw) and C) Emerging flame retardants (in mg/kg lw) in North Atlantic killer whales sampled from 2008 to 2022. Legacy contaminants were measured in 162 individuals while the ∑PBDEs and FRs analyses were conducted only on a subset of individuals (n = 105). (Note: the legend is in the same order as the bars). The letters indicate the results of the Tukey post-hoc tests on the various contaminant classes tested against location (p-value threshold set at 0.05).

Although much lower than for the legacy POPs, for ∑PBDEs and non-BDE FRs, killer

whales in the Western NA again showed higher concentrations than those in the Eastern NA. The predominant PBDE congeners were BDE-47 followed by BDE-99 and 100 at all locations and ∑PBDEs were significantly higher in Norway than in Iceland ($F = 8.1$; $p = 0.005$). For emerging BFRs, α-HBCDD dominated across locations with levels in Iceland being statistically higher than in Norway ($F = 10.1$; $p = 0.002$), reaching the highest concentration in Greenlandic killer whales at 1.2 ± 0.4 mg/kg lw. This compound had a similar concentration in mixed-diet individuals from Iceland, reported in our previous study, which found a mean concentration of 1.0 mg/kg lw in the mixed-diet individuals (Remili et al., 2021). In Eastern Canada, α-HBCDD had a mean concentration of 0.6 ± 0.3 mg/kg lw. These α-HBCDD concentrations in Iceland, Eastern Canada and Greenland are among the highest reported for any marine mammal (including killer whales) to date, far exceeding HBCDD concentrations reported in southern resident killer whale blubber (0.1 mg/kg lw) or transient killer whale liver (0.2 mg/kg lw) (Jayda, 2018; Lee et al., 2023). The inclusion of HBCDD (hexabromocyclododecane) in the Stockholm Convention only occurred in 2013, as the environmental concentrations of this compound were increasing (Covaci et al., 2006). Since the ban, α-HBCDD has become the main HBCDD congener in biota and has shown biomagnifying capabilities (Li et al., 2018). Due to its stable structure and widespread distribution, α-HBCDD has not significantly decreased in the environment (Su et al., 2018).

Prior to discussing the effects of sex and diet composition on contaminant concentrations in NA killer whales, it is worth considering how historic usage of legacy contaminants may influence contaminant variation in killer whales across the NA. Killer whales located in the Western part of North America, specifically the Eastern Canadian Arctic and Eastern Canada, exhibited the highest concentrations of legacy POPs and ∑PBDEs. This distribution pattern contrasts with the findings in other Arctic biotic and abiotic compartments, where POP levels are

typically higher in Greenland and Norway rather than in the Canadian Arctic (Muir et al., 2000; Su et al., 2008; Vorkamp et al., 2016). The traditional trend of higher concentrations in the Eastern NA can be attributed to the historical use of contaminants, followed by their transportation through the atmosphere and oceans towards the East (Brown et al., 2018). Consequently, when assessing contaminant exposures in killer whales, it is imperative to consider the influence of both sex, diet and age.

6.6.2 Effect of sex on POP concentrations

Male killer whales were significantly more contaminated than females, with the GLM effect for "adult males" being significant and positive in PCBs, DDTs, CHLs and ClBz, but not HCHs (Fig. 6-3, Table S6-4, and S6-5). For FRs, we found that *sex* in Iceland and Norway had a significant effect on PBDE concentrations (F: 14.06, $p < 0.01$). However, this test resulted in non-significant differences in non-BDE FR concentrations. Lower concentrations in females are most likely attributed to the maternal offloading of lipophilic contaminants from mammalian mothers to their offspring. During gestation and lactation, adult female cetaceans transfer approximately 10% and 60% of their body burdens to their offspring, respectively (Borrell et al., 1995; Ross et al., 2000a; Tanabe et al., 1982; Wells et al., 2005). Since a significant portion of these burdens is unloaded during the first pregnancy and nursing period, contaminant levels may also differ depending on the number of births and, consequently, the age of the individuals. Juveniles included in our analyses did not significantly differ from adult females (Fig. 6-3, Table S64 and 6-5), but future studies should include the actual age of the individuals to account for this likely source of variation. To do so, precise age estimates could be obtained from photo-identification for long-term monitored populations, or in future studies through the development of DNA-methylation methods to age killer whales (Peters et al., 2023).

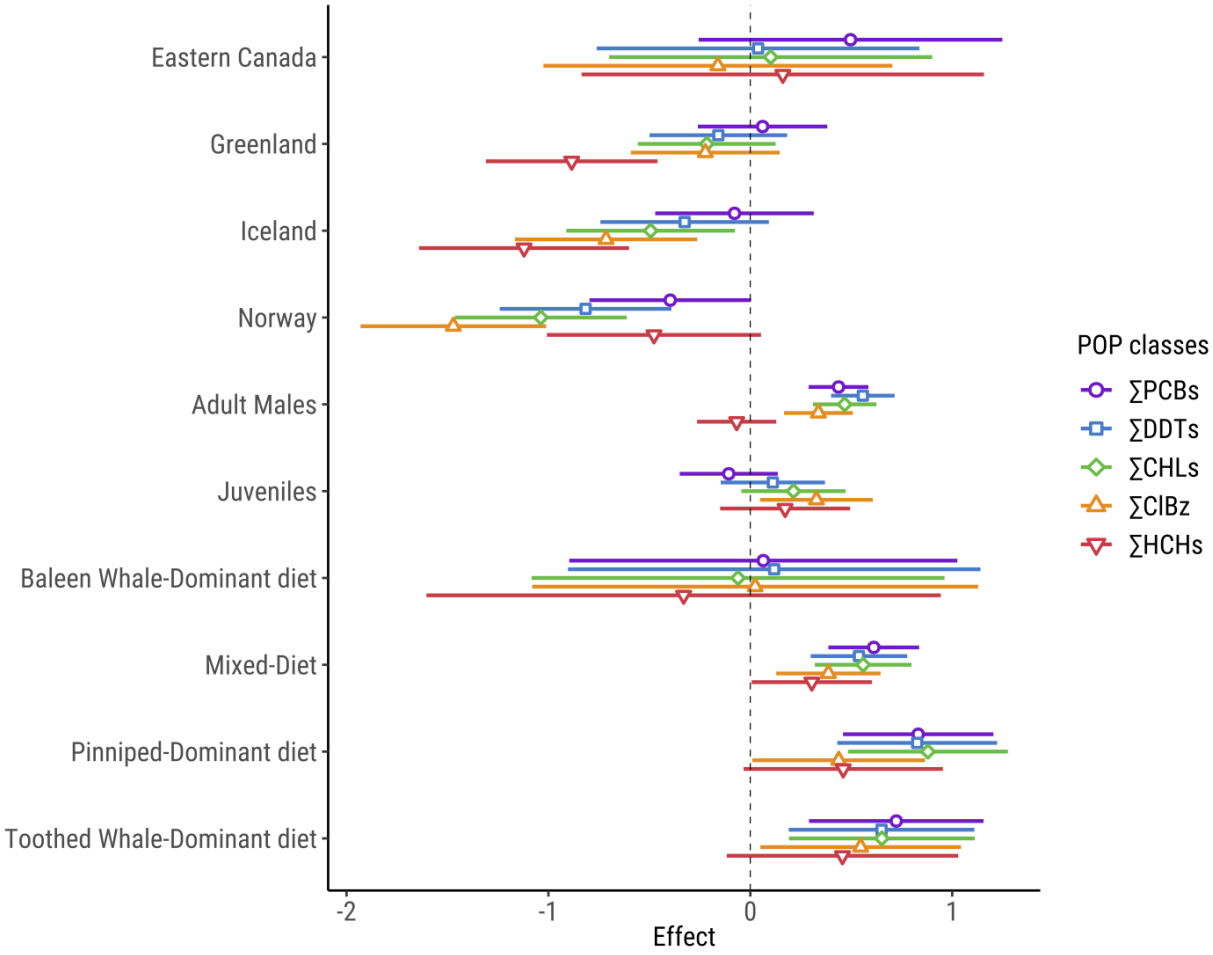

Figure 6-3:Summary results from the generalized linear modelling approach testing the effects [and 95% confidence intervals] of the following independent variables: location, sex / age, and diet-type (inferred from QFASA) on the log-transformed concentrations of ∑PCBs, ∑DDTs, ∑CHLs, ∑HCHs, and ∑ClBzs in the blubber biopsies of North Atlantic killer whales. The intercept represents adult females (for sex / age), Eastern Canadian Arctic (for location) and fish-dominant diets (for diet-type). Model selection table and precise effects, confidence intervals and significance are available in the SI (Table S6-4-S6-5).

6.6.3 Effect of the diet on contaminant concentrations

Across the NA, the *diet-type* predictor had a stronger effect than *sex* or *location* for PCBs, DDTs, CHLs and ClBz, but not HCHs (Fig. 6-3, Tables S6-4 and S6-5). Specifically, and compared to fish-dominant diets, diets including pinnipeds and toothed whales (mixed-diet included) resulted in significantly higher concentrations for these contaminant classes, with

pinniped-dominant diets having a stronger effect (mean effect across PCBs, DDTs, CHLs and ClBz: 0.75) compared to toothed whale-dominant diets (mean effect: 0.64) or mixed-diets including fish and pinnipeds or toothed whales (mean effect: 0.53). Baleen whale diets did not differ significantly from the fish diets, probably due to a low sample size and high variability in the concentrations across the individuals feeding on baleen whales. The stronger effect for *diet-type*, in relation to *sex* or even *location* is of particular interest, and shows that, at least for the most abundant POP classes, dietary habits impact contaminant accumulations more than sex differences or geographical variations in contaminant distribution for NA killer whales. For FRs, we found that *diet-type* in Iceland and Norway influenced PBDEs concentrations (F: 4.3, p = 0.04). However, this effect was lower than for sex. While statistical testing was not conducted to determine the impact of diet types on α-HBCDD, the most detected emerging FR, important variations in α-HBCDD concentrations were observed between killer whales that primarily feed on marine mammals and those that primarily feed on fish. Individuals feeding on fish had mean α-HBCDD concentrations of 0.2 ± 0.1 mg/kw lw, while individuals feeding on marine mammals had mean concentrations of 0.7 ± 0.1 mg/kg lw. PCAs focused on the log-transformed concentrations of PCB, OC and FR compounds showed a striking trend of compound concentrations across the NA, with fish feeding individuals having lower contaminant concentrations, followed by the mixed-diet individuals, and toothed whale and pinniped-feeding individuals having the highest concentrations (Fig. S6-2).

Within the marine mammal-dominant diets, we observed that PCB and OC profiles (expressed as % contribution of congeners to the ∑PCBs or ∑OCs) differed between diets including pinnipeds, and diets including toothed whales (Fig. 6-4). Specifically, for PCBs, pinniped-dominant diets were associated with higher concentrations of highly chlorinated compounds (e.g., CB-138, -170, -180, -183 and -187). Conversely, toothed whale-dominant diets

were associated with lower chlorinated compounds (e.g., CB-74, -95, -99, -101 and -149) (Fig. 3A). Interestingly, contaminant profiles were reported to show similar patterns between serum samples of harbor seals (a pinniped) and harbor porpoises (a toothed whale) (Weijs et al., 2009). Pinniped-dominant diets showed higher proportions of oxychlordane and heptachlor epoxide among the OC compounds, while toothed whale-dominant diets exhibited higher percentages of DDT. Previous reports indicated that pinnipeds in the Arctic generally have relatively higher proportions of CHL (within their overall OC levels) compared to toothed whales (Muir et al., 1999). This finding may explain why killer whales feeding on pinnipeds displayed higher CHL percentages than those feeding on toothed whales (Fig. 6-4B). The higher percentages of DDT in diets primarily consisting of odontocetes (toothed whales) is perhaps not surprising, considering the comparatively lower contaminant-eliminating capacities of cetaceans when compared to pinnipeds (DDE being a metabolite of DDT) (Meyer et al., 2018). Killer whales who had baleen whale-dominant diets (specifically in Eastern Canada) overlapped with toothed whale-dominant diets and fish-dominant diets for PCB and OC profiles (Fig. 6-4A-B), suggesting little distinction in POP accumulation between toothed and baleen whale feeding killer whales.

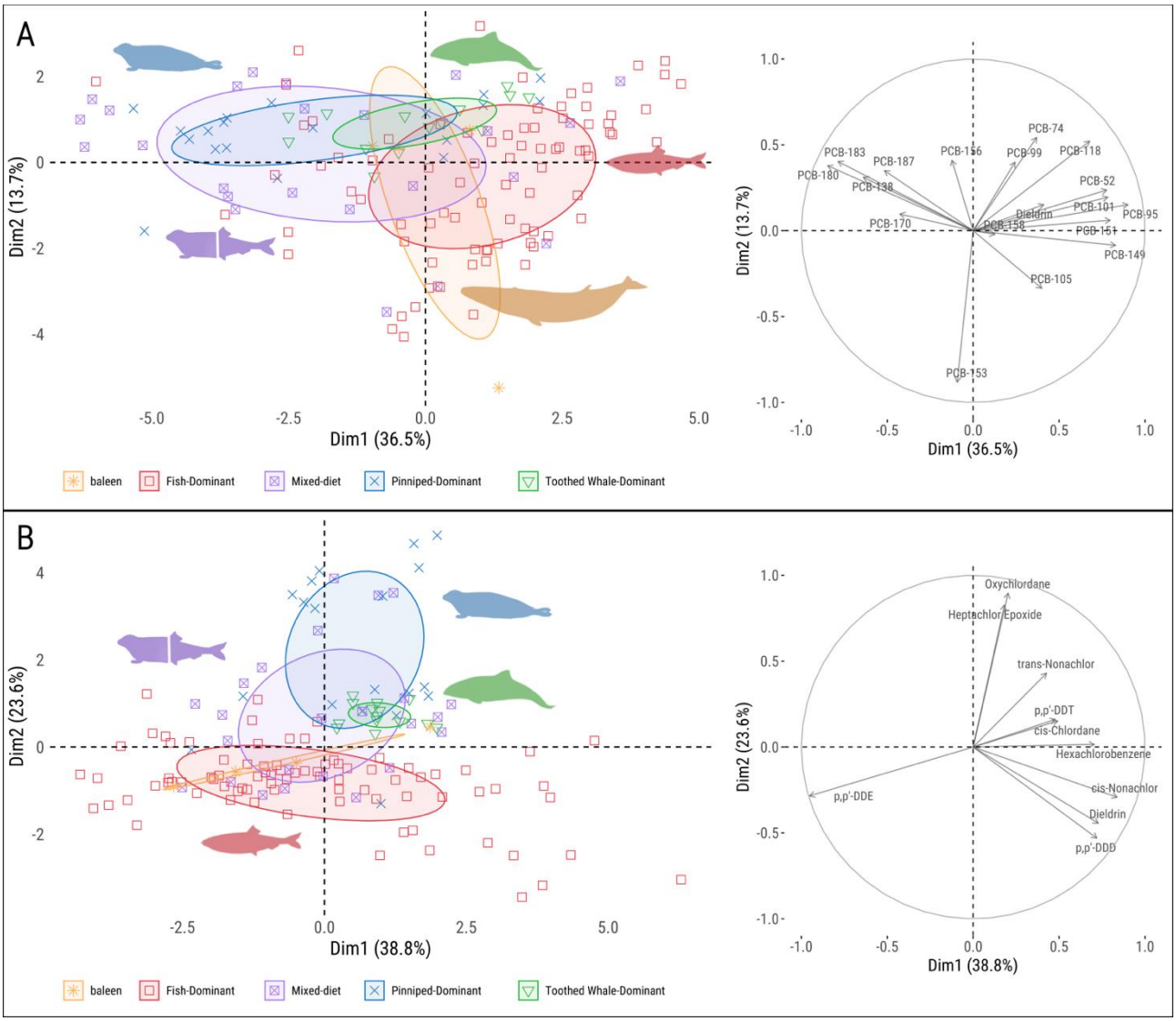

Figure 6-4:Principal component analysis on the individual compounds for A) polychlorinated biphenyls (PCBs) and B) organochlorine pesticides (OCs) in the blubber of North Atlantic killer whales (Panels A and B share a legend). Only the PCB and OC compounds detected in > 70 % of the individuals were included. The FR equivalent of this PCA can be found in the SI (Fig. S6-1) Each point represents an individual killer whale. The animal shapes represent the diet-types of killer whales, inferred from QFASA (fish for fish-dominant diets, mixed fish/seal for mixed diets, porpoise for toothed whale-dominant diets, etc.)

Intra-population diet variation estimated using QFASA (Remili et al. 2023) allowed for interpretation of the differences in accumulation of PCBs, OCs and FRs within several

populations. In Iceland, killer whales with mixed diets showed significant variation in contaminant concentrations. Their concentrations of POPs sometimes overlapped with both fish-eating killer whales in the same population and with killer whales in Greenland and Western NA (Fig. S6-2). In terms of PCBs and OCs specifically (Fig. S6-2A-B), the overlap between the two ellipses representing the two diet types in Iceland was negligible or non-existent, indicating pronounced diet-related differences in contaminant accumulation within this single group. These dietary variations among killer whales in Iceland, and the rest of the NA deserve further research, especially for the individuals known to consume marine mammals (Remili et al., 2023; Remili et al., 2021; Samarra et al., 2017c).

6.6.4 Risk Assessment for PCBs

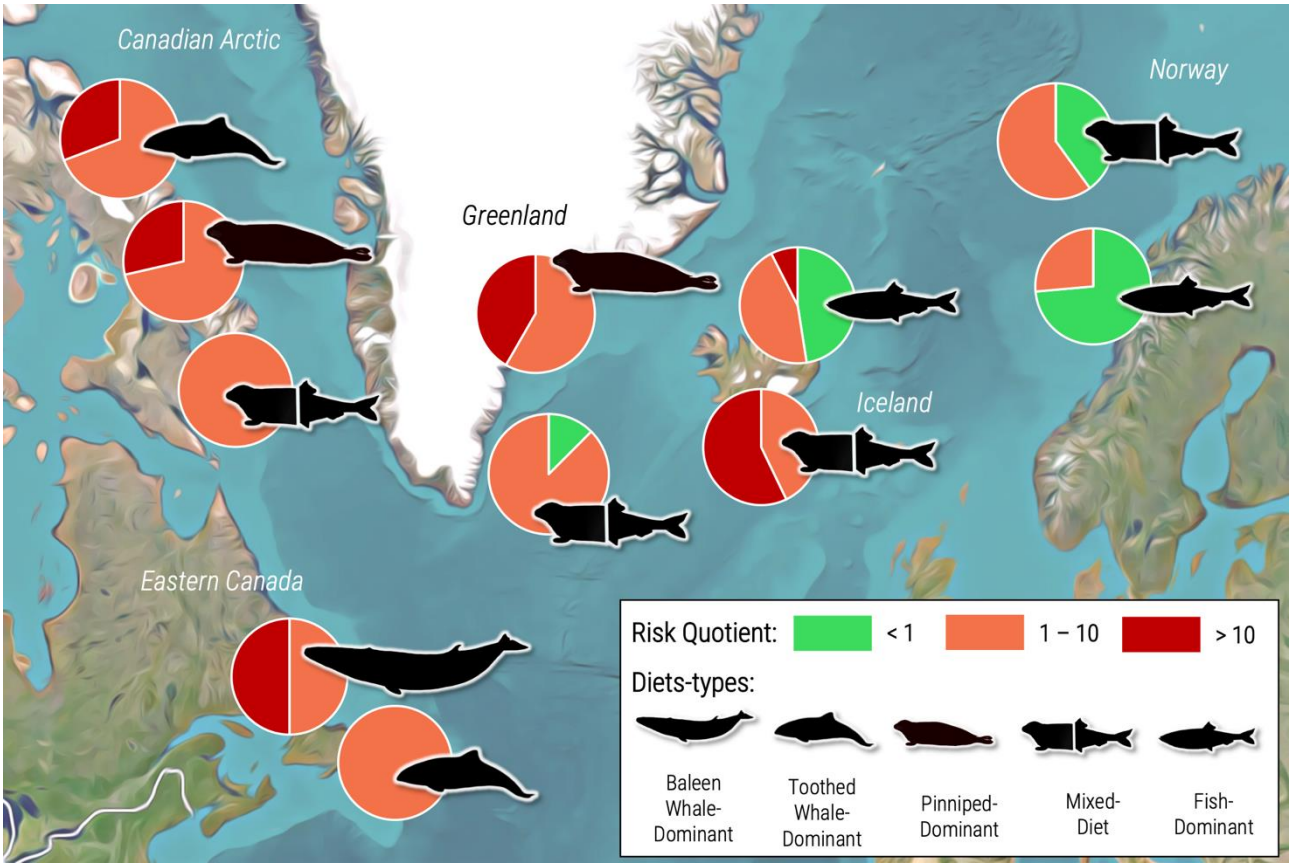

Figure 6-5:Risk Quotient estimated for ∑PCBs based on a conservative 10 mg/kg lw threshold for immunotoxic and hormonal imbalance effects (from Dietz et al. 2019) in North Atlantic killer whales divided by location and diet-types inferred from quantitative fatty acid signature analysis on the same individuals. See Table S6 for more information and see Fig. S3 for the same map with RQ differences based on sex/age.

Risk quotients for killer whales in the Western NA (Canadian Arctic, Eastern Canada) and mid-NA (Greenland) were consistently higher than those in the Eastern NA, regardless of their sex/age or diet-type. The risks were similar among killer whales feeding on toothed whales (mean RQ: 11.4 ± 4.4) and killer whales feeding on pinnipeds (mean RQ: 8.6 ± 2.3) in the Eastern Canadian Arctic. Killer whales sampled in Eastern Canada and feeding on baleen whales had a moderate to high risk of health effects (mean RQ: 11.3 ± 3.4). This risk, associated to high PCB concentrations may also be attributed to local sources of PCBs from the contaminated Great Lakes area into the Gulf of Saint Lawrence (Metcalfe et al., 2004; Westgate et al., 1997). Eastern Canadian killer whales thus deserve further attention in future ecotoxicological studies, since preying on pinnipeds or toothed whales may significantly incerase their PCB-associated risks. In Greenland, killer whales feeding on pinnipeds were more at risk (mean RQ: 8.1 ± 1.5) than those having a mixed diet (mean RQ: 4.0 ± 0.7). In the Eastern NA, i.e., Iceland and Norway, killer whales showed lower RQs for ∑PCBs, but these RQs were consistently greater for individuals with a mixed diet compared to those feeding on fish (Fig. 3, Table S6). Previous studies reported that individual killer whales in Iceland and Norway who do prey on marine mammals in addition to fish face significantly greater risks than individuals in the same regions that have fish-dominant diets (Andvik et al., 2020; Remili et al., 2021). Males across these two locations also faced the highest risks of health effects compared to females and/or juveniles (Fig. S3). For example, male Icelandic killer whales who seasonally travel to Scotland to prey on seals or who were photographed preying on porpoises in Iceland (i.e.: individuals IS015, 172, 241 & 256) all

had RQs higher than 10 (mean RQ: 22.8 ± 11.0), which represents a high risk of health effects including reproductive failure (Helle et al., 1976; Jepson et al., 2016). In Norway, the two males identified by QFASA as having a mixed diet (i.e.: individuals 17010 and 18025) had an RQ of 4.0 on average, which represents a significantly lower risk than in Iceland. However, it should be noted that an RQ of 4 represents a ∑PCBs concentration of ~ 40 mg/kg lw, similar to the 41 mg/kg lw threshold for risk of reproductive failure (Helle et al., 1976). The findings presented in this study are of concern, particularly when considering that mixtures of contaminants (not just PCBs, but DDTs, CHLs, and newer POPs) may have a greater immunotoxic effect on killer whales compared to individual contaminants alone (e.g., just PCBs) (Desforges et al., 2017). These results highlight the necessity for improved risk assessment methods specific to these ecologically significant top predator species (Desforges et al., 2018; Hall et al., 2018).

Our results highlight the need for further efforts in legacy and emerging pollutant management and waste disposal when it comes to reducing the risks faced by the oceans' top predator. The Stockholm Convention will likely fail to meet its 2025 and 2028 targets for the phase-out of hazardous substances and safe waste disposal (Melymuk et al., 2022). Addressing the more specific issue of pollution in marine mammals will necessitate a pragmatic and systematic approach to mitigate its adverse effects. First, enhancing monitoring programs in the NA and elsewhere is crucial to gather reliable data on pollutant levels in marine mammal populations. Second, interdisciplinary and international collaboration among ecotoxicologists, conservation biologists, policymakers, and other stakeholders is crucial in the near future. Other recommendations regarding emerging chemicals of concern include holding chemical producers responsible for data generation, protecting high-risk populations, avoiding assumptions of "safe" exposure levels, and addressing financial conflicts of interest in assessments of chemical risks (Woodruff et al., 2023). These collaborations and recommendations may facilitate knowledge

exchange and resource sharing, enabling the development of targeted strategies for pollution mitigation, and enhanced cetacean conservation.

In this first detailed analysis of POP concentrations in killer whales across the NA, an almost two orders-of magnitude difference in means for PCBs and OCs were found between the individuals of the Eastern NA and Western NA. Across locations, there were also large differences in diets and this diet variation explained the majority of the contaminant differences, indicating how critical this feeding variation among killer whale groups is for their resulting contaminant loads and risks for health effects. Nonetheless, wide intrapopulation differences in contaminant concentrations and associated health risks were also found. The findings of this study support the need for additional measures to be taken to ensure the safe disposal of POP-contaminated waste and to prevent the continued runoff and deposition of these contaminants into the environment and living organisms. It is crucial that these conservation efforts also focus on preventing the release of newer and potentially highly toxic contaminants into the environment.

6.7 ACKNOWLEDGEMENTS

This research received financial support from the Canada Research Chairs Program (grant 950-232183) and the Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grants Program (grant RGPIN-2019-05330) to M. A. McKinney. Support is acknowledged from a Canada Foundation for Innovation Grant (grant #37873 to M. A. McKinney). Additional funds for sample collection in Greenland were provided by The Danish Cooperation for Environment in the Arctic (DANCEA) Programme, specifically grants MST-112-00171 and MST-112-00199 to R. Dietz. And C. Sonne. A. Remili thanks the Fonds de Recherche du Québec Nature et Technologies Doctoral Scholarship, as well as ECOTOQ and the

Quebec Center for Biodiversity Science for the funding during her doctoral research. Samples collected in Saint-Pierre et Miquelon were obtained under the authorization of Arrêté n°431 (July 17th, 2019) from Préfecture de Saint-Pierre et Miquelon, with the collaboration of Cécile Vincent and Joël Detcheverry, and support from the Office Français de la Biodiversité (OFB). In Iceland, sample collection was made possible through funding from an Icelandic Research Fund START Postdoctoral Fellowship (grant number 120248042 to F.I.P. Samarra) and an Icelandic Research Fund Project Grant (grant number 163060 to F.I.P. Samarra). Funding for Canadian samples was provided by the Fisheries and Oceans Canada (DFO) Nunavut Implementation Fund (NIF) and Species at Risk Act (SARA), the Nunavut General Monitoring Plan (NGMP), and the Nunavut Wildlife Management Board (NWMB).

Thanks to A. Pedersen for assistance with the QuEChERS extractions and GC-MS training at McGill University and to D. Blair for the BFR analyses at NWRC, Ottawa. We also thank T. Edkins from DFO, Canada, for support with sample shipping and all individuals involved in the fieldwork in Iceland for data collection efforts, as well as R. Kilabuk for contributing to the collection of killer whale samples from Pangnirtung, Nunavut. Thanks to Lisa Kettemer for arranging the shipment of the Norwegian biopsies to Canada. Finally, thanks to Eve Jourdain, Katrine Borgå, Anders Ruus and Richard Karoliussen for their involvement in the prey sample collection in Norway for the fatty acid analyses.

6.8 AUTHORS' CONTRIBUTIONS

M.A. McKinney, A. Remili, R. Dietz, and C. Sonne designed the study with input from all co-authors. F.I.P. Samarra, A.H. Rikardsen, A. Rosing-Asvid, S.H. Ferguson, C.A. Watt, C.J. D. Matthews; R. Dietz, C. Sonne, and J.J. Kiszka provided the killer whale samples/data. M.A.

McKinney, R.J. Letcher, C.J.D. Matthews, C.A. Watt, S.H. Ferguson and A. Remili performed or supervised the contaminant analyses. A. Remili performed the data analysis and wrote the original draft of the manuscript with input from M.A. McKinney. All authors reviewed and edited subsequent versions of the manuscript.

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6.10 SUPLLEMENTARY INFORMATION

Table S6-1: Sample collection dates and locations for the 162 killer whales in our study.

Species	N	ID	Geography	Date	Paper originally published
Killer whale	48	IS003-IS423	Iceland	2014-2016	Remili et al. 2021
Killer whale	58	17001-19023	Northern Norway	2017-2019	This paper
Killer whale	2	40888-40889	Faroe Islands	2008	Pedro et al. 2017
Killer whale	19	35143-51613	Greenland	2012-2014, 2021	Pedro et al. 2018 & This paper
Killer whale	5	KW1-KW5	Eastern Canada	2019-2021	This paper
Killer whale	30	ARPI-2013-4001 - 4008 ARPI-OO.012018 - ARPI-OO.182018 ARPI_00_02_2019-ARPI_00_11_2019	Canadian Arctic	2013-2019	This paper

Details on each laboratory extraction:

Table S6-2: List of laboratories and analyses used to extract and quantify legacy and emerging POPs in this study. NIST 1945 refers to pilot whale blubber, while NIST 1946 refers to Great Lakes fish homogenates. Blanks were run with each extraction batch and only the Greenlandic samples from Pedro et al. 2017 had some compounds detected in the blanks and substracted from the concentrations. Additional information, including the instrument methods can be found in the SI. List of abbreviations: ASE= accelerated solvent extraction, GPC= gel permeation chromatography, SPE= solid phase extraction.

Sample locations	Number of samples	Laboratory	Extraction method (brief)	Classes of contaminants analyzed			NIST used for QC	NIST accuracies	Internal standards recoveries	Blanks ran with each batch	References
				PCBs	OCs	PBDEs/ FRs					
Canadian Arctic	30	ALS Global Laboratories (Burlington, Canada)	Soxhlet extraction - GPC – SPE cleanup	✓	✓	X	1946	113 ± 24% (PCBs) 116 ± 24% (OCs)	99 ± 13% (PCBs)	✓	**This paper**
Greenland	16	Center for Environmental Science and Engineering (University of Connecticut, USA)	ASE - GPC - SPE cleanup	✓	✓	X	1945	80 ± 17% (PCBs) 73 ± 11% (OCs)	90 ± 20% (PCBs) 77 ± 13% (OCs)	✓	Pedro et al. 2017
Greenland Eastern Canada Norway	3 5 58	Department of Natural Resources (McGill University, Canada)	QuEChERS (Pedersen et al., 2023)	✓	✓	✓ (subset)	1945	125 ± 17% (PCBs) 80 ± 8% (OCs) 110 ± 22% (FRs)	91 ± 21% (PCBs) 66 ± 15% (OCs) 63 ± 27% (FRs)	✓	**This paper**
Iceland	48	National Wildlife Research	ASE - GPC - SPE cleanup	✓	✓	✓	1945	105 ± 6% (PCBs) 102 ± 6% (OCs)	85 ± 12% (PCBs) 70 ± 13% (OCs)	✓	Remili et al. 2021

Center, ECCC (Ottawa, Canada)		115 ± 17% (FRs)	150 ± 67% (FRs)

List of POPs in common:

PCBs: CB-18, -28/31, -44,-49, -52, -70, -74, -82, -87, -95, -99, -101, -105, -110, -118, -128, -138, -149, -151, -153, -156, -158, -169, -170, -177, -180, -183, -187, -194, -195, -199, -206 and -209.

OCs: 1,2,4,5-Tetrachlorobenzene, 1,2,3,4-Tetrachlorobenzene, Pentachlorobenzene, Hexachlorobenzene, α-Hexachlorocyclohexane, β-Hexachlorocyclohexane, Oxychlordane, *trans*-Chlordane, *cis*-Chlordane, *trans*-Nonachlor, *cis*-Nonachlor, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT, Mirex, Heptachlor Epoxide, and Dieldrin.

PBDEs and FRs: BDE-3, -7, -15, -17, -28, -47, -49, -66, -71(/HBB), -77, -85/-155, -99, -100, -119, -138, -153, -154, -181, -183, -203, -205, -206, -207 and -209, BB-101, BB-153, BEH-TEBP, BTBPE, DBDPE, α-/β-DBE-DBCH, DBHCTD, anti-DDC-CO, syn-DDC-CO, EH-TBB, α-HBCDD, OBTMPI, PBB-Acr, PBEB, PBP-AE, PBP-dbpe, PBT, TBCT, TBP-AE, TBP-DBPE, and TBX.

ALS Global Laboratories (Burlington, Canada)

Blubber samples were analyzed for PCBs and OCPs following US EPA Method 1699 (US EPA 2007) by ALS Global Laboratories (Burlington ON). In brief, blubber was thoroughly homogenized with pre-cleaned anhydrous Na_2SO_4 and Soxhlet extracted with dichloromethane (DCM). Prior to extraction a suite of 25 $^{13}C_{12}$-PCBs and 15 ^{13}C-OCP-related compounds were added as recovery surrogates, and $^{13}C_{12}$-133 was added after extraction as a recovery standard for gel permeation chromatography (GPC) performance. Lipids were removed by GPC with n-

hexane:DCM (1:1) as the eluent. The GPC eluate was split into OCP and PCB fractions. The OCP fraction was cleaned up on 2 % deactivated silica gel then reduced to 0.05 mL for analysis by gas chromatography-high resolution mass spectrometry (GC-HRMS). The PCB fraction was cleaned up on an acid-silica gel column (45% w/w H_2SO_4) then reduced to 0.04 mL for GC-low resolution (LR) MS analysis. OCPs were analysed by GC-HRMS at 10000 mass resolution and PCBs by GC-LRMS using a 30 m HP5MS capillary column and quantified by isotope dilution using the ^{13}C-surrogates (USEPA 2007). A certified reference material (SRM 1946 fish tissue; National Institute of Standards and Technology [NIST]) and a laboratory control sample (PCB congeners in corn oil) were analysed with the samples. Average recoveries of 15 OCPs and 37 PCB congeners relative to the certified values for the NIST fish tissue were 113 ± 24% for OCPs and 116 ± 24% for PCBs, respectively. Recovery of 96 PCB congeners from the laboratory control samples averaged 98.5 ± 12.5%.

National Wildlife Research Center, ECCC (Ottawa, Canada)

Extraction and analysis of PCBs/OCs/PBDEs/non-PBDE FRs were based on methods previously described (McKinney et al., 2009). Briefly, blubber biopsies were cut lengthwise into two equal depth segments: one slice (excluding skin) for analysis of POP concentrations and the other preserved for future studies. The blubber subsample for POP analysis (mean weight: 0.04 g, range: 0.01 to 0.18 g) was then accurately weighed into a mortar and homogenized with precleaned diatomaceous earth (DE). An aliquot was used to determine lipid content gravimetrically. After spiking with a mixture of 13C-labeled and nonlabelled C/PCB/FR surrogates as internal standards, extraction was performed by accelerated solvent extraction; then, extracts were subjected to cleanup by gel permeation chromatography and solid phase extraction. The final extract was separately analyzed for PCBs and OCs, by gas

chromatography–mass spectrometry (GC-MS) with electron ionization (EI), and then for PBDE/non-PBDE FRs, by GC-MS with electron capture negative ionization (ECNI). Identification and quantification were performed using MassHunter Quantitative Analysis software (Version B.07.01, Agilent Technologies). Each batch included ten samples, a blank, and standard reference material, the National Institute of Standards and Technology pilot whale (*Globicephala melas*) blubber homogenate (NIST-1945). The full details can be found in Remili et al. 2021 (Remili et al., 2021).

The standard reference material SRM (NIST 1945 pilot whale blubber) was run eight times and checked for precision and accuracy. The overall POP recovery was 102% (96–109%) for ∑OCs (14 compounds), 105 % (99–111 %) for ∑PCBs (thirty-three congeners), and 112 % (91–135 %) for ∑PBDEs (five congeners). Internal standard recoveries were 85 % (68–95 %) for PCBs (six ^{13}C-labeled congeners), 70 % (47–106 %) for OCs (18 ^{13}C-labeled compounds), and 150 % (89–214 %) for FRs (five ^{13}C-labeled compounds). Method limits of detection (MLODs) and quantification (MLOQs) were defined as the minimum amount of analyte which produced a peak with a signal-to-noise ratio of 3 and 10, respectively. A blank was run with each batch. No contamination was present in any of the blanks.

Department of Natural Resources (McGill University, Canada)

First, 0.075-0.100 g of blubber was sub-sampled and placed into 2 mL pre-filled bead hard tissue homogenizing tubes (VWR, Mississauga, ON, Canada). Next, each tube was topped with a 1.25 mL aliquot of 20:80 (v:v) ethyl acetate:acetonitrile, and the tubes were then placed in a Precellys Evolution tissue homogenizer (Bertin Instruments, USA) at 6,500 rpm for 4 cycles of 30 seconds each (i.e., 30 seconds of homogenization followed by a 30-second pause) at 0 degrees Celsius. After the homogenization process, the solvent was transferred to a polypropylene (PP)

centrifuge tube and rinsed with ethyl acetate:acetonitrile. A mass-labeled PCB/OC internal standard was added, and the mixture was centrifuged to obtain the supernatant. Next, an elution process was carried out using EMR-Lipid cartridges, and a second round of elution was performed using EMR-Lipid and Bond-Elut Jr PSA cartridges. The resulting mixture was transferred to a heavy-duty glass centrifuge tube, where water and hexane were added and subsequently separated. The upper layer was transferred to a glass tube, and anhydrous MgSO4 was added before centrifugation. The supernatant was collected and evaporated, followed by gravity elution through preconditioned silica cartridges. The eluent was collected in a new glass tube, evaporated, and mixed with isooctane. Each extract was spiked with a mass-labeled PCB-138 normalization standard, transferred to a GC vial, and stored in a freezer or analyzed immediately using GC-MS. Concentrated extracts of target PCBs and OC pesticides were analyzed using a GC-MS system (Agilent Technologies, GC system 7820 A, MSD 5977 B) with selective ion monitoring (SIM) on a fused silica DB-5 capillary column (Pedersen et al., 2023). After being run on our GC, a subset of these samples was sent to the National Wildlife Research Center, ECCC (Ottawa, Canada) for PBDEs/FRs quantification on their GC-MS with electron capture negative ionization (ECNI), as described in the previous section. The data acquisition and processing were performed using Agilent MassHunter. Instrument blanks, internal standard spikes, and calibration standards were run before and after every 12 samples. Method blanks and standard reference materials (SRMs) were included with each batch of 10 killer whale samples. No compounds were detected in the blanks. The NIST accuracies were 125 ± 17 % (PCBs), 80 ± 8 % (OCs) and 110 ± 22 % (FRs). The internal standard recoveries were 91 ± 21 % (PCBs), 66 ± 15 % (OCs) and 63 ± 27 % (FRs).

Center for Environmental Science and Engineering (University of Connecticut, USA)

The samples were accurately weighed and then subjected to established extraction procedures (McKinney et al., 2011b). Each sample was homogenized with diatomaceous earth (Hydromatrix™) and spiked with deuterated surrogates, including 1,2,4,5-tetrachlorobenzene-d2, 2,5-dichlorobiphenyl-d5, 2,3,4,5,6-pentachlorobiphenyl-d5, and 2,3,3′,4,4′,5-hexachlorobiphenyl-d3. Extraction was performed using an accelerated solvent extraction (ASE) system with a 1:1 dichloromethane:hexane mixture for three cycles at 1500 psi and 100 °C. A 10% portion of the extract was used to determine the lipid content through gravimetric analysis. The extracts were then filtered and purified using gel permeation chromatography and solid-phase extraction polar cartridges. The concentrated extracts were monitored for PCBs and OCs using a gas chromatograph coupled with a Quattro Micro tandem mass spectrometer (GC-MS/MS) system, employing a Rxi-5Sil MS GC column (30 m length, 0.25 mm I.D., 0.25 μm film thickness; Restek Corporation, PA, USA) (Provatas et al., 2014). Data acquisition and processing were performed using Waters MassLynx™ software v. 4.1 (Milford, MA, USA). Multiple-reaction monitoring (MRM) was utilized to monitor PCBs, whereas selected ion monitoring (SIM) was employed for monitoring OCs. At the start and every 15 samples, reagent blanks, recovery standards, and calibration standards were run. The standard reference material NIST-1945 (whale blubber) was included in each batch of samples for extraction. Blanks generally yielded results below the detection limit for PCBs and most OC compounds, although occasional detections were made for heptachlor and heptachlor epoxide. Consequently, these detections were subtracted on a batch-by-batch basis. Trace amounts of p,p'-DDE, oxychlordane, and trans-nonachlor were detected in a few blanks, but their levels were more than ten times lower than the concentrations in the samples, so blank subtraction was not performed for these analytes. The full details can be found in Pedro et al. 2017 (Pedro et al., 2017). Internal standard

recoveries were 90 ± 20 % (PCBs) and 77 ± 13 % (OCs) while NIST accuracies were 80 ± 17 % (PCBs) and 73 ± 11 % (OCs).

Table S6-3:Concentrations of legacy contaminant classes (mean ± SE) in ng/g lw, PBDEs (in ng/g lw) and new and emerging flame retardants (in ng/g lw). The ∑PBDEs and non-BDE FRs analyses were conducted only on a subset of individuals.

All individuals from our study (results in ng/g lw) (n = 162)								
	Lipid %	∑PCBs	∑DDTs	∑CHLs	∑HCHs	∑ClBz	Mirex	Dieldrin
Canadian Arctic (n=30)	10.7 ± 1.7	92036 ± 9791	108080 ± 28674	63164 ± 15472	970 ± 176	3324 ± 680	1379 ± 326	8559 ± 1958
Eastern Canada (n=5)	10.9 ± 3.4	96012 ± 31150	63179 ± 31951	22722 ± 7156	521 ± 250	1027 ± 447	389 ± 233	6186 ± 2243
Greenland (n=19)	58.3 ± 3.9	66140 ± 10610	50789 ± 7827	38259 ± 7881	135 ± 24	1819 ± 429	771 ± 77	3964 ± 877
Iceland (n=48)	9.7 ± 1.2	42127 ± 11130	28282 ± 6148	9705 ± 2229	50 ± 14	295 ± 41	433 ± 120	1368 ± 244
Faroe (n=2)	61.3	2953 ± 1483	3320 ± 178	3320 ± 165	23 ± 3	331 ± 24	69 ± 33	272 ± 116
Norway (n=58)	18.0 ± 1.5	12160 ± 10816	6741 ± 1520	2055 ± 369	308 ± 90	55 ± 7	21 ± 6	441 ± 67
Subset of individuals analyzed for PBDEs and non-BDE FRs (results in ng/g lw) (n = 105)								
	Lipid %	∑ PBDE	∑ non-BDE FRs					
Eastern Canada (n=4)	12.7 ± 3.7	7483 ± 2250	894 ± 288					
Greenland (n=3)	23.0 ± 5.9	4196 ± 1585	1320 ± 490					
Iceland (n=48)	9.7 ± 1.2	743 ± 238	307 ± 159					
Norway (n=50)	18.1 ± 1.7	1047 ± 165	185 ± 38					

Table S6-4:Model selection table for the legacy contaminant classes (log-transformed), tested against sex/age (adult females, adult males, juveniles), diet-type (inferred from QFASA analyses) and location, in blubber biopsies from 138 killer whales across the North Atlantic (sampled from 2012 to 2022). AICc: Akaike's Information Criterion corrected for small sample size. Only models with a ΔAICc below 4 are shown. Competing models under ΔAICc = 4 were averaged.

Models	k	AICc	ΔAICc	loglik	AICwt	R2
∑PCBs ~ sex/age + diet-type + location	11	180.6	0.00	-77.04	0.95	0.55
∑DDTs ~ sex/age + diet-type + location	11	197.3	0.00	-85.48	0.98	0.61
∑CHLs ~ sex/age + diet-type + location	11	197.7	0.00	-85.61	0.99	0.67
∑ClBz ~ sex/age + location	7	217.2	0.00	-100.03	0.68	0.66

∑ClBz ~ sex/age + diet-type + location	11	218.9	1.74	-96.21	0.29	0.69
∑HCHs ~ location	5	251.5	0.00	-119.43	0.67	0.45
∑HCHs ~ sex/age + location	7	253.9	2.41	-118.40	0.20	0.46
∑HCHs ~ diet-type + location	9	255.3	3.76	-116.76	0.10	0.48

Table S6-5:Model coefficients [and 95% confidence intervals] table for the selected models from Table S6-4. Legacy contaminant classes (log-transformed) were tested against sex/age (adult females, adult males, juveniles), diet-type (inferred from QFASA analyses) and location, in blubber biopsies from 138 killer whales across the North Atlantic (sampled from 2012 to 2022).

	∑PCBs	∑DDTs	∑CHLs	∑ClBz	∑HCHs
(Intercept)	4.90 *** [4.51, 5.29]	4.91 *** [4.50, 5.32]	4.70 *** [4.29, 5.11]	3.67 *** [3.22, 4.11]	3.45 *** [2.94, 3.96]
Eastern Canada	0.50 [-0.26, 1.25]	0.04 [-0.76, 0.84]	0.10 [-0.70, 0.90]	-0.16 [-1.02, 0.70]	0.16 [-0.84, 1.16]
Greenland	0.06 [-0.26, 0.38]	-0.16 [-0.50, 0.18]	-0.22 [-0.56, 0.13]	-0.22 [-0.59, 0.15]	-0.88 *** [-1.31, -0.46]
Iceland	-0.08 [-0.47, 0.31]	-0.33 [-0.74, 0.09]	-0.49 [-0.91, -0.08]	-0.71 * [-1.17, -0.26]	-1.12 *** [-1.64, -0.60]
Norway	-0.40 [-0.80, 0.70.00]	-0.82 ** [-1.24, -0.39]	-1.04 *** [-1.46, -0.61]	-1.47 *** [-1.93, -1.01]	-0.48 [-1.01, 0.05]
Adult Males	0.44 *** [0.29, 0.58]	0.56 *** [0.40, 0.71]	0.47 *** [0.31, 0.62]	0.34 ** [0.17, 0.51]	-0.07 [-0.26, 0.13]
Juveniles	-0.11 [-0.35, 0.14]	0.11 [-0.15, 0.37]	0.21 [-0.04, 0.47]	0.33 [0.05, 0.61]	0.17 [-0.15, 0.49]
Mixed-Diet	0.61 *** [0.39, 0.84]	0.54 *** [0.30, 0.78]	0.56 *** [0.32, 0.80]	0.39 * [0.13, 0.64]	0.30 [0.01, 0.60]
Baleen Whale-Dominant Diet	0.06 [-0.90, 1.03]	0.12 [-0.90, 1.14]	-0.06 [-1.08, 0.96]	0.02 [-1.08, 1.13]	-0.33 [-1.60, 0.94]
Pinniped-Dominant Diet	0.83 *** [0.46, 1.20]	0.83 *** [0.43, 1.22]	0.88 *** [0.48, 1.28]	0.44 [0.01, 0.87]	0.46 [-0.03, 0.95]
Toothed Whale-Dominant Diet	0.72 ** [0.29, 1.16]	0.65 * [0.19, 1.11]	0.65 * [0.19, 1.11]	0.55 [0.05, 1.04]	0.46 [-0.12, 1.03]

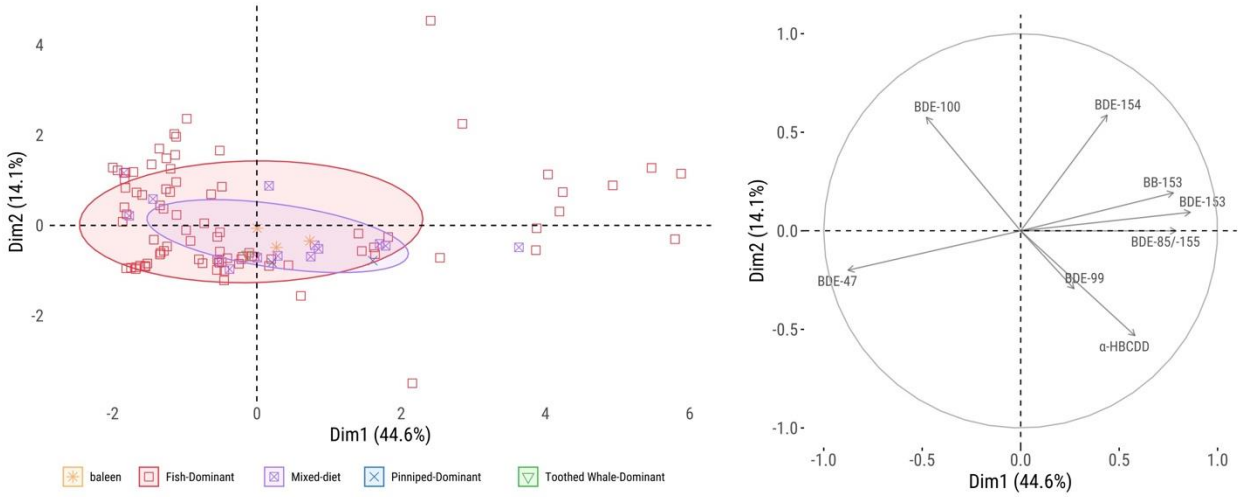

Figure S6-1:PCA on the flame retardant (FR) profiles (% contribution of individual congeners to the ∑FRs) for compounds detected in > 50% of the individual killer whales across the NA (n = 105).

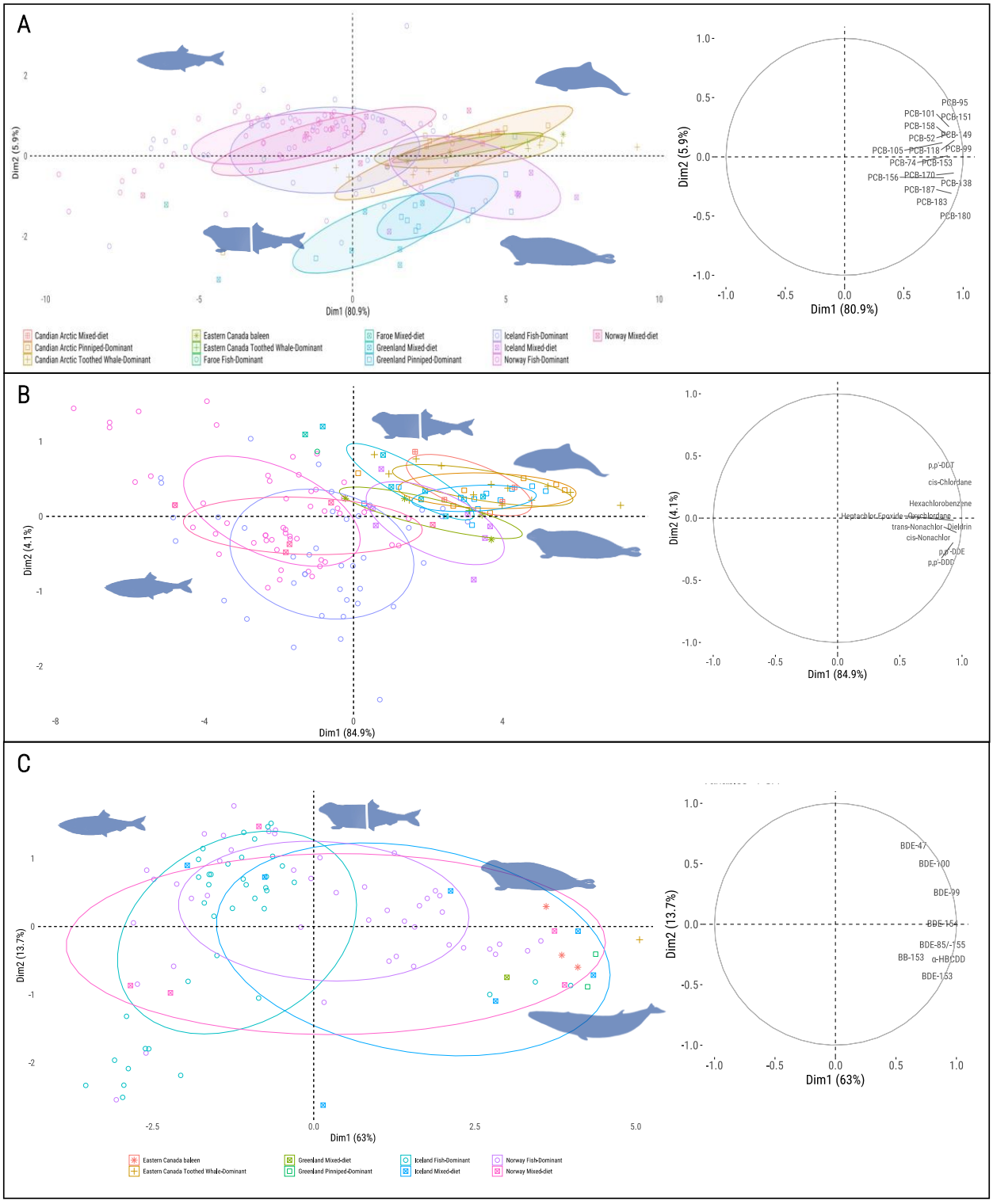

Figure S6-2:Principal component analysis on the individual compounds for A) polychlorinated biphenyls (PCBs) and B) organochlorine pesticides (OCs) and C) flame retardants (FRs) in the blubber of North Atlantic killer whales. Panels A and B share a legend. Only the PCB and OC compounds detected in > 70% of the individuals were included, and only the FR compounds

detected in > 50% of the individuals were included. Each point represents an individual killer whale. Raw congener concentrations (not percentages) were log-transformed and the PCA was scaled. The number of individuals feeding on marine mammals were too low to compute ellipses for panel C, but they can be located on the right side of the panel, showing higher concentrations overall, than fish-feeding killer whales.

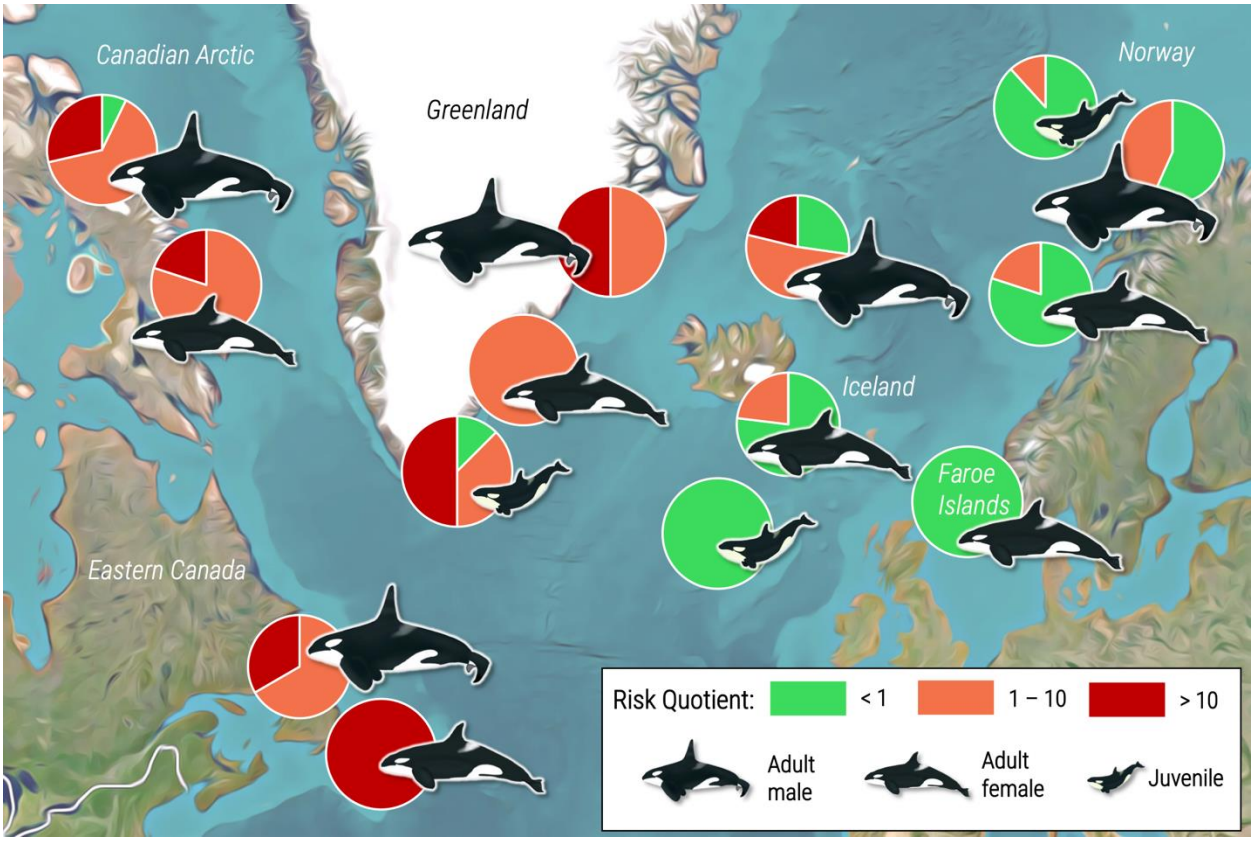

Figure S6-3: Risk Quotient estimated for ∑PCBs based on a conservative 10 mg/kg lw threshold for immunotoxic and hormonal imbalance effects (from Dietz et al. 2019) in North Atlantic killer whales divided by location and sex. Note: The juvenile category in Norway includes individuals identified in the field as "females or juveniles". See Table S6 for more information.

Table S6-6:Risk quotients (RQs) for ∑PCBs calculated for each of the 162 individuals in our study.

Population	Sex	Det-type	ID	Risk Quotient	RQ category
Norway	male	Fish-Dominant	17001	0.56	0 – 1
Norway	male	Fish-Dominant	17002	1.44	1 – 10
Norway	female or juv	Fish-Dominant	17003	1.16	1 – 10

Norway	female or juv	Fish-Dominant	17004	0.97	0 – 1
Norway	female or juv	Fish-Dominant	17006	0.54	0 – 1
Norway	female or juv	Fish-Dominant	17007	1.05	1 – 10
Norway	female or juv	Fish-Dominant	17008	0.72	0 – 1
Norway	female or juv	Fish-Dominant	17009	0.26	0 – 1
Norway	male	Fish-Dominant	17010	1.04	1 – 10
Norway	male	Mixed-diet	17011	0.91	0 – 1
Norway	female	Fish-Dominant	17012	0.69	0 – 1
Norway	female	Fish-Dominant	17013	0.06	0 – 1
Norway	juvenile	Fish-Dominant	17014	0.32	0 – 1
Norway	female or juv	Fish-Dominant	17015	0.61	0 – 1
Norway	female or juv	Fish-Dominant	17016	0.89	0 – 1
Norway	female or juv	Fish-Dominant	17017	0.10	0 – 1
Norway	female or juv	Fish-Dominant	17019	0.68	0 – 1
Norway	female or juv	Fish-Dominant	17021	0.18	0 – 1
Norway	male	Fish-Dominant	18001	4.51	1 – 10
Norway	male	Fish-Dominant	18005	7.42	1 – 10
Norway	male	Fish-Dominant	18006	0.36	0 – 1
Norway	male	Fish-Dominant	18007	0.56	0 – 1
Norway	male	Fish-Dominant	18008	1.12	1 – 10
Norway	male	Fish-Dominant	18009	0.97	0 – 1
Norway	female	Fish-Dominant	18010	0.60	0 – 1
Norway	male	Fish-Dominant	18011	0.17	0 – 1
Norway	male	Fish-Dominant	18012	0.91	0 – 1
Norway	male	Fish-Dominant	18014	0.82	0 – 1
Norway	female	Fish-Dominant	18015	0.21	0 – 1
Norway	female	Fish-Dominant	18016	0.59	0 – 1
Norway	male	Fish-Dominant	18017	3.50	1 – 10
Norway	male	Fish-Dominant	18020	1.44	1 – 10
Norway	female	Fish-Dominant	18021	0.07	0 – 1
Norway	juvenile	Fish-Dominant	18022	0.19	0 – 1
Norway	juvenile	Fish-Dominant	18023	0.40	0 – 1

Norway	male	Fish-Dominant	18024	0.74	0 – 1
Norway	male	Mixed-diet	18025	6.93	1 – 10
Norway	male	Fish-Dominant	18026	1.19	1 – 10
Norway	male	Fish-Dominant	18027	1.45	1 – 10
Norway	male	Fish-Dominant	18029	0.43	0 – 1
Norway	female	Mixed-diet	19000	1.79	1 – 10
Norway	na	Fish-Dominant	19002	0.73	0 – 1
Norway	female	Mixed-diet	19005	1.18	1 – 10
Norway	male	Fish-Dominant	19007	6.91	1 – 10
Norway	female	Fish-Dominant	19008	0.60	0 – 1
Norway	juvenile	Mixed-diet	19009	0.17	0 – 1
Norway	male	Fish-Dominant	19010	0.80	0 – 1
Norway	male	Fish-Dominant	19011	0.06	0 – 1
Norway	male	Fish-Dominant	19013	5.24	1 – 10
Norway	male	Fish-Dominant	19014	1.36	1 – 10
Norway	male	Fish-Dominant	19015	0.50	0 – 1
Norway	juvenile	Fish-Dominant	19016	0.33	0 – 1
Norway	male	Fish-Dominant	19017	0.13	0 – 1
Norway	female	Fish-Dominant	19018	0.52	0 – 1
Norway	male	Fish-Dominant	19019	0.36	0 – 1
Norway	female or juv	Fish-Dominant	19020	0.34	0 – 1
Norway	male	Fish-Dominant	19021	0.52	0 – 1
Norway	male	Fish-Dominant	19023	0.69	0 – 1
Greenland	female	Mixed-diet	35143	4.60	1 – 10
Faroe Islands	female	Mixed-diet	40888	0.15	0 – 1
Faroe Islands	female	Fish-Dominant	40889	0.44	0 – 1
Greenland	female	Mixed-diet	48335	6.69	1 – 10
Greenland	female	Mixed-diet	48336	5.12	1 – 10
Greenland	na	Pinniped-Dominant	48337	10.53	10 – 100
Greenland	female	Mixed-diet	48338	2.66	1 – 10
Greenland	male	Pinniped-Dominant	48339	19.60	10 – 100
Greenland	male	Pinniped-Dominant	48340	13.33	10 – 100

Greenland	male	Pinniped-Dominant	48732	6.00	1 – 10
Greenland	na	Mixed-diet	51607	0.63	0 – 1
Greenland	female	Pinniped-Dominant	48733	1.78	1 – 10
Greenland	female	Pinniped-Dominant	48735	4.73	1 – 10
Greenland	female	Pinniped-Dominant	48736	5.96	1 – 10
Greenland	male	Pinniped-Dominant	51601	11.31	10 – 100
Greenland	na	Mixed-diet	51606	2.78	1 – 10
Greenland	male	Pinniped-Dominant	51610	4.58	1 – 10
Greenland	male	Pinniped-Dominant	51613	5.26	1 – 10
Candian Arctic	female	Toothed Whale-Dominant	ARPI-2013-4001	1.50	1 – 10
Candian Arctic	female	Pinniped-Dominant	ARPI-2013-4002	3.84	1 – 10
Candian Arctic	female	Toothed Whale-Dominant	ARPI-2013-4003	4.54	1 – 10
Candian Arctic	female	NA	ARPI-2013-4004	4.41	1 – 10
Candian Arctic	male	Toothed Whale-Dominant	ARPI-2013-4005	6.42	1 – 10
Candian Arctic	female	Mixed-diet	ARPI-2013-4006	3.26	1 – 10
Candian Arctic	male	Toothed Whale-Dominant	ARPI-2013-4007	1.41	1 – 10
Candian Arctic	na	Pinniped-Dominant	ARPI-2013-4008	1.01	1 – 10
Candian Arctic	male	NA	ARPI-OO-01-2019	0.43	0 – 1
Candian Arctic	male	Pinniped-Dominant	ARPI-OO-02-2019	9.26	1 – 10
Candian Arctic	male	Pinniped-Dominant	ARPI-OO-03-2019	16.82	10 – 100
Candian Arctic	female	Toothed Whale-Dominant	ARPI-OO-04-2019	16.13	10 – 100
Candian Arctic	male	NA	ARPI-OO-05-2019	16.33	10 – 100
Candian Arctic	female	Pinniped-Dominant	ARPI-OO-06-2019	9.55	1 – 10
Candian Arctic	male	Pinniped-Dominant	ARPI-OO-08-2019	15.63	10 – 100
Candian Arctic	female	Toothed Whale-Dominant	ARPI-OO-09-2019	56.93	10 – 100
Candian Arctic	male	Toothed Whale-Dominant	ARPI-OO-10-2019	21.14	10 – 100
Candian Arctic	female	Mixed-diet	ARPI-OO-11-2019	2.27	1 – 10
Candian Arctic	female	Toothed Whale-Dominant	ARPI-OO.012018	3.96	1 – 10
Candian Arctic	male	Toothed Whale-Dominant	ARPI-OO.022018	7.86	1 – 10
Candian Arctic	male	Toothed Whale-Dominant	ARPI-OO.032018	8.23	1 – 10

Candian Arctic	female	Toothed Whale-Dominant	ARPI-OO.052018	14.36	10 – 100
Candian Arctic	female	Toothed Whale-Dominant	ARPI-OO.062018	1.67	1 – 10
Candian Arctic	female	Toothed Whale-Dominant	ARPI-OO.102018	3.65	1 – 10
Candian Arctic	female	Pinniped-Dominant	ARPI-OO.132018	4.19	1 – 10
Candian Arctic	female	NA	ARPI-OO.142018	1.65	1 – 10
Candian Arctic	male	Toothed Whale-Dominant	ARPI-OO.152018	9.29	1 – 10
Candian Arctic	male	NA	ARPI-OO.162018	7.45	1 – 10
Candian Arctic	male	NA	ARPI-OO.172018	8.02	1 – 10
Candian Arctic	male	Toothed Whale-Dominant	ARPI-OO.182018	8.64	1 – 10
Greenland	female	Mixed-diet	GL1	5.36	1 – 10
Greenland	female	Pinniped-Dominant	GL2	3.38	1 – 10
Greenland	male	Pinniped-Dominant	GL3	10.43	10 – 100
Iceland	male	Mixed-diet	IS003	19.55	10 – 100
Iceland	male	Fish-Dominant	IS008	1.28	1 – 10
Iceland	male	Fish-Dominant	IS010	0.82	0 – 1
Iceland	male	Fish-Dominant	IS011	9.51	1 – 10
Iceland	male	Mixed-diet	IS015	38.65	10 – 100
Iceland	male	Fish-Dominant	IS018	1.94	1 – 10
Iceland	male	Mixed-diet	IS028	1.86	1 – 10
Iceland	female	Fish-Dominant	IS045	0.58	0 – 1
Iceland	male	Fish-Dominant	IS046	1.66	1 – 10
Iceland	male	Fish-Dominant	IS047	0.63	0 – 1
Iceland	female	Fish-Dominant	IS062	0.95	0 – 1
Iceland	male	Fish-Dominant	IS067	0.91	0 – 1
Iceland	male	Fish-Dominant	IS068	1.49	1 – 10
Iceland	male	Fish-Dominant	IS071	1.86	1 – 10
Iceland	male	Fish-Dominant	IS078	0.49	0 – 1
Iceland	female	Fish-Dominant	IS104	0.13	0 – 1
Iceland	male	Fish-Dominant	IS117	0.98	0 – 1
Iceland	male	Fish-Dominant	IS122	2.07	1 – 10
Iceland	male	Fish-Dominant	IS136	6.54	1 – 10

Iceland	male	Fish-Dominant	IS139	3.81	1 – 10
Iceland	male	Fish-Dominant	IS143	0.76	0 – 1
Iceland	male	Fish-Dominant	IS151	6.14	1 – 10
Iceland	female	Fish-Dominant	IS152	2.92	1 – 10
Iceland	female	Fish-Dominant	IS154	0.17	0 – 1
Iceland	male	Fish-Dominant	IS155	1.31	1 – 10
Iceland	male	Mixed-diet	IS159	1.44	1 – 10
Iceland	male	Fish-Dominant	IS169	1.05	1 – 10
Iceland	male	Mixed-diet	IS172	13.00	10 – 100
Iceland	female	Fish-Dominant	IS174	0.30	0 – 1
Iceland	male	Fish-Dominant	IS183	1.02	1 – 10
Iceland	male	Fish-Dominant	IS211	3.86	1 – 10
Iceland	male	Mixed-diet	IS241	19.13	10 – 100
Iceland	male	Fish-Dominant	IS243	18.46	10 – 100
Iceland	male	Fish-Dominant	IS251	10.69	10 – 100
Iceland	female	Mixed-diet	IS253	3.76	1 – 10
Iceland	male	Fish-Dominant	IS254	0.54	0 – 1
Iceland	male	Mixed-diet	IS256	20.51	10 – 100
Iceland	female	Fish-Dominant	IS257	2.14	1 – 10
Iceland	female	Fish-Dominant	IS262	0.39	0 – 1
Iceland	male	Fish-Dominant	IS267	0.86	0 – 1
Iceland	female	Fish-Dominant	IS271	0.14	0 – 1
Iceland	female	Fish-Dominant	IS274	0.24	0 – 1
Iceland	female	Fish-Dominant	IS279	0.20	0 – 1
Iceland	male	Fish-Dominant	IS280	1.69	1 – 10
Iceland	female	Fish-Dominant	IS288	0.67	0 – 1
Iceland	male	Fish-Dominant	IS306	1.02	1 – 10
Iceland	juvenile	NA	IS351	0.34	0 – 1
Iceland	juvenile	Fish-Dominant	IS423	0.71	0 – 1
Eastern Canada	male	Baleen Whale-Dominant	KWNFL22	7.47	1 – 10
Eastern Canada	male	Toothed Whale-Dominant	SPM2	2.85	1 – 10
Eastern Canada	female	Baleen Whale-Dominant	SPM3	12.61	10 – 100

Eastern Canada	male	Baleen Whale-Dominant	SPM4	20.21	10 – 100
Eastern Canada	na	Baleen Whale-Dominant	SPM5	4.86	1 – 10

7 CHAPTER SEVEN: COMPREHENSIVE DISCUSSION

7.1 REFLECTION ON NORTH ATLANTIC ECOTYPES

Over the course of four and a half years, I have devoted my efforts to studying North Atlantic killer whales and their dietary habits. Through my research, a significant finding has emerged: there is no distinct categorization of "type 1" or "type 2" North Atlantic killer whales based on their feeding ecology. Coincidentally, Dr. Foote, the author of the 2009 study that originally proposed these ecotypes, recently published a note in Marine Mammal Science advocating for the retirement of the "type 1" and "type 2" classification in future research (Foote, 2022). Ever since I began my research on North Atlantic killer whales, I eagerly anticipated the publication of this paper by Dr. Foote. I deeply appreciate Dr. Foote's humility in revisiting his previous research findings a decade later. Dr. Foote's contributions have been fundamental in shaping our understanding of North Atlantic killer whale diets and have provided a strong foundation for further research in this field. His original 2009 study was instrumental in expanding our knowledge of North Atlantic killer whales and has inspired numerous subsequent research initiatives (Foote et al., 2009). In that study, Dr. Foote observed morphological differences between museum specimens and stranded killer whales, leading to the identification of two distinct types of North Atlantic killer whales. Type 1 killer whales were described as generalist feeders, primarily consuming fish but occasionally preying on marine mammals like seals. These individuals showed heavy tooth wear, usually associated with feeding on fish. Indeed, “sucking” (for lack of a better term) herring causes the scales to rub against the teeth resulting in significant wear in older individuals. It is worth noting that Type 1 killer whales encompassed most Northeastern Atlantic individuals. Since then, research has revealed the prey-switching abilities of Icelandic and Norwegian killer whales, highlighting that some individuals

exhibit mixed diets, incorporating both seals and fish, while the rest specializes on herring or other fish species like mackerel or lumpfish (Eve Jourdain et al., 2019; Olafsdottir et al., 2019; Samarra et al., 2017a). In our studies, mixed-diet killer whales exhibited higher contaminant concentrations, indicating a long-standing preference for marine mammals in their feeding habits, compared to an occasional consumption of marine mammals.

Dr. Foote subsequently identified type 2 North Atlantic killer whales as specialists in feeding on marine mammals. These whales exhibited minimal tooth wear, indicating a diet primarily focused on marine mammals. The classification of type 2 was based on stranded killer whales found in the Faroe Islands and Scotland. The limited sample size of only five individuals posed a significant challenge when considering type 2 killer whales as a potential ecotype. Dr. Foote acknowledged this limitation, emphasizing the need for more extensive data to establish meaningful classifications. He encouraged researchers to collect samples, particularly from the isolated killer whale populations in Greenland and Canada, to further our understanding of North Atlantic killer whale ecology. In the meantime, Dr. Foote suggested temporarily retiring the use of the "type 1/type 2" classification until sufficient data becomes available.

Our results of QFASA modeling, qualitative fatty acid analyses and contaminant concentrations based on ~200 killer whales spanning from the west to the east NA Ocean provided a panoramic view of the complex feeding strategies across the NA, as well as within-population individual feeding specialization (Remili et al., 2022; Remili et al., 2023; Remili et al., 2021). Our findings emphasize the importance of shifting our focus from population-level studies to individual-based investigations when studying the ecology of North Atlantic killer whales. Increasing evidence suggests significant dietary variations within different killer whale populations globally. Over the past few years, this approach of recognizing interindividual differences has gained momentum in marine mammal research (Bories et al., 2021; Jourdain et

al., 2020; Jourdain et al., 2017; Newsome et al., 2015; Samarra et al., 2017c; Vongraven et al., 2014). By considering individual whales as unique entities rather than mere constituents of larger populations or types, we can gain a deeper understanding of their intricate interactions with the marine environment (Araujo et al., 2011; Bolnick et al., 2011; Bolnick et al., 2003). This perspective shift holds great potential for enhancing our comprehension of the ecological dynamics between killer whales and the oceans they inhabit (Johnson et al., 2019; Pontbriand et al., 2023; Zhao et al., 2022). For killer whales, further research should investigate this dietary plasticity from a genetic approach to understand how population structure may arise from this dietary variation (de Bruyn et al., 2013; Tavares et al., 2018). Nevertheless, our discoveries offer fresh insights into identifying prey species and estimating the diet of killer whales, which can contribute to understanding the predatory effects of these apex predators across various ocean basins worldwide, including the North Atlantic.

7.2 THE FUTURE OF QFASA IN MARINE MAMMAL STUDIES

QFASA was developed by Sara Iverson in 2004 and was primarily used on various pinniped studies. The CCs developed by Iverson and her team were obtained through feeding trials on harbor seals, grey seals, and mink (Iverson et al., 2004). Then, in 2008, Thiemann et al. applied QFASA to polar bears (*Ursus maritimus*) using mink-derived CCs, which was then followed by other polar bear QFASA studies by McKinney et al. and Bourque et al., in 2013 and 2020 respectively (Bourque et al., 2020; McKinney et al., 2013).

While QFASA research has made significant progress in the realm of marine mammals, particularly with the refinement of the model and the introduction of new functionalities by Bromaghin in his 2017 QFASAR package, relatively few researchers have ventured to adapt the

model for cetaceans (Bromaghin, 2017). The first limitation came from the challenges associated with conducting feeding trials, which can be both costly and ethically complex when applied to cetaceans. The second limitation stemmed from the stratification of blubber in cetaceans, rendering many CCs unsuitable for use with biopsy samples that typically collect the outer layers of blubber. However, Choy et al. demonstrated that CCs derived from mink yielded the most accurate estimations of diets for two captive beluga whales when applied to the inner blubber fatty acids (Choy et al., 2019). They used these identical CCs to estimate the diet of wild belugas, by applying them only the inner blubber of harvested individuals (Choy et al., 2020). While promising, these two studies were only conducted on the inner blubber, which, for wild cetaceans, is only available in deceased individuals, not live healthy individuals.

Following these two studies, we established our own killer whale CCs using deceased captive individuals from SeaWorld that had been consistently fed a controlled diet with a fixed prey composition. When applied to the corresponding fatty acid layer, these CCs produced highly accurate diet estimates for the captive individuals and credible estimates for harvested Greenlandic killer whales. Notably, the QFASA models consistently identified that the predominant prey species were as harp and hood seals, which aligned with the findings of stomach content analyses in the sampled individuals. The publication of our research encouraged other research groups to embark on developing QFASA for their respective species. In fact, a research team is currently preparing a publication that examines the performance of their own CCs on bottlenose dolphins in Sarasota Bay, Florida (Tatom-Naecker et al., 2023). In addition to our efforts with killer whales, our research team is currently testing the applicability of QFASA on small cetaceans off Saint-Pierre and Miquelon in collaboration with Florida International University, harbor porpoises in collaboration with NOAA, as well as on sperm whales in collaboration with the Department of Fisheries and Oceans (DFO). The QFASA modeling

publications we released for killer whales have received considerable acclaim within the marine mammal community, and praise through online communications and conference accolades. I expect that these publications will serve as an inspiration for numerous new projects focusing on the development of QFASA for other species, further expanding the application and impact of this methodology.

One key mystery pertains to the turnover and integration of fatty acid signatures within the blubber. While Choy et al. approximated that changes in diet would take approximately two weeks to be reflected in the inner blubber, there is currently a lack of precise estimates regarding the integration of these signatures into the outer blubber, which is typically collected through skin biopsies (Choy et al., 2019). Addressing this aspect will contribute to a more comprehensive understanding of the dynamics and temporal aspects of fatty acid integration within different layers of the blubber. In Chapter 5 of this thesis, we hypothesized that fatty acid signatures obtained from biopsies could potentially reflect a feeding period ranging from a few weeks to several months, taking into account the observed skin turnover in stable isotope studies. To advance our understanding in this area, future investigations should prioritize determining the rate of tissue turnover and precisely determining the specific feeding period represented by the biopsies. As an additional note, I am excited to share that in the upcoming fall of 2023, I will be embarking on a postdoctoral project in collaboration with NOAA. This project will focus on examining seasonal variations in the diets of Southern Resident killer whales. By investigating these seasonal changes, we aim to gain valuable insights into tissue turnover dynamics and enhance the accuracy of our estimations regarding the feeding periods of these killer whales.

An additional limitation arises from species-specific variations in the metabolism associated with the deposition of fatty acids from the diet into the blubber. As illustrated in Chapter 4, it became evident that species-specific CCs are necessary to accurately estimate the

diet of killer whales. This notion was further supported by a recent poster presentation at the European Cetacean Society, highlighting that QFASA modeling for bottlenose dolphins requires the use of bottlenose dolphin-specific CCs to achieve accurate results. In comparison, the CCs utilized in our QFASA studies for dolphins in Sarasota Bay yielded less precise estimations of their true diets. These findings underscore the significance of employing species-specific CCs to ensure the accuracy of diet estimations in QFASA modeling (Tatom-Naecker et al., 2023). The authors of the conference poster recommended using multiple CCs from different species to investigate potential diet estimate variations in species where CCs are not available. We hypothesized in Chapter 5 that our calibration coefficients could likely be applied to other species with a similar stratification index as killer whales. These species include pilot whale, belugas and narwhals (Koopman, 2007). It is important to recognize that even the fatty acids initially identified as "dietary" by Iverson undergo modifications during the transition from the diet to the predator's fatty storage. As a result, despite the similarities in the blubber structure among killer whales, belugas, pilot whales, and narwhals, these species may exhibit variations in metabolism and the deposition of specific dietary fatty acids. Consequently, it becomes imperative for future investigations to delve into this potential avenue.

These two factors emphasize the extent of our current knowledge gaps regarding the fate of various fatty acids. The objective of this discussion is to inspire future studies dedicated to understanding the lipid metabolism in killer whales and other cetaceans. Through the application of innovative lipidomic techniques, researchers may gain valuable insights into these transformations and explore potential variations among individuals (Bories et al., 2021). By addressing these knowledge gaps, we can improve the accuracy of diet estimations in cetacean research.

7.3 REFLECTION ON CONTAMINANT EXPOSURES AND ASSOCIATED RISKS

In chapter 6, we presented the results of our transatlantic contaminant study on killer whales. We measured ∑PCB concentrations far beyond the highest thresholds in multiple populations, including the Canadian Arctic, Eastern Canada, and Greenland. The highest threshold known for ∑PCBs is currently set at 41 mg/kg kw. It was calculated in 1976 for female seals in the Baltic Sea that showed a high rate of fetal resorption and uterine occlusions (Helle et al., 1976). Kannan et al. (2000) estimated a 17 mg/kg lw threshold for marine mammals which would translate to lymphocyte proliferation reductions, reduced natural killer cell activity and lower vitamin A concentrations. Desforges et al. (2016) estimated the threshold for lymphocyte proliferation reduction to be at 10 mg/kg lw ∑PCBs in the blubber for belugas, based on in vitro studies, which would cause enough stress at the population level to impact its healthy growth. They then reported that realistic contaminant mixtures used in in vitro studies on killer whale and polar bear immune cells were by far more toxic than a single compound exposure (Desforges et al., 2017). For ecotoxicologists, including myself and many others, this particular issue becomes worrisome, especially considering how concentrated other compounds can be in killer whales (∑DDTs, ∑CHLs, etc.). Currently, there is a lack of defined thresholds for other types of contaminants and mixtures of pollutants that marine mammals are exposed to. To address this knowledge gap, further research employing "omics" approaches, such as metabolomics and transcriptomics, can provide valuable insights into the impacts of different contaminants on marine mammals. By using these advanced techniques, we can better understand the health risks associated with contaminant mixtures and work towards establishing thresholds for these effects. This research is essential for gaining a comprehensive understanding of the impacts of contaminants on marine mammal populations (Simond et al., 2020; Simond et al., 2022).

Throughout my early career, I have observed passionate debates surrounding the significance of thresholds in marine mammal risk assessment. In 2018, a study conducted by Desforges et al. (2018) and published in *Science* generated controversy as it employed a population dynamics model for killer whales based on the thresholds mentioned in the previous paragraph. The study predicted that the accumulation of PCBs could lead to a significant decline and potential extinction of half of the killer whale populations within the next century. However, despite the study's diligent data consolidation efforts, it faced swift and explicit criticism from within our field, targeting the authors, the modeling approach, the conclusions, and other related aspects. This critical response, evident in the publication of six eLetters in response to the study, has created an atmosphere of skepticism and uncertainty in the field of marine mammal risk assessment. Personally, I experienced the remnants of this animosity when presenting the results of Chapter 3 at an international conference. I believe that more productive conversations could have focused on advancing marine mammal conservation and risk assessment instead of incessantly fixating on the outcomes of a peer-reviewed and non-retracted study. I must admit that the idea of publishing Chapter 6 is already causing me anxiety, primarily due to the intense negative response regarding the previous study on global killer whale contamination.

7.4 DEALING WITH BAD NEWS AND GLIMPSES OF HOPE

The Stockholm Convention will likely fail to meet its 2025 and 2028 targets for the phase-out of hazardous substances and safe waste disposal (Melymuk et al., 2022). This failure may be attributed to challenges in implementation, lack of political will, global trade complexities, enforcement issues, technological limitations, and emerging contaminants. The management of the remaining global stock of PCBs and other legacy contaminants is challenging, particularly due to a significant portion that is considered "unmanageable." These

unmanageable stocks, already released into the environment or lacking proper documentation, pose risks of environmental releases (and human exposures), worsened by aging infrastructure containing these contaminants. Addressing these challenges will require international cooperation, capacity building, resource mobilization, and sustained commitment from member states (Melymuk et al., 2022).

Addressing the more specific issue of pollution in marine mammals will necessitate a pragmatic and systematic approach to mitigate its adverse effects. First, enhancing monitoring programs in the North Atlantic and elsewhere is crucial to gather reliable data on pollutant levels in marine mammal populations. For example, the establishment of dedicated and year-round monitoring program for killer whales on the Eastern coast of Canada seem like a feasible operation. Additionally, a summer monitoring program based in Nuuk or Tasiilaq (Greenland) could allow for killer whale photoidentification during the summer months and measure recapture indexes within Greenlandic killer whales. It would also enable comparisons between multiple locations and identify migration patterns in the North Atlantic. Second, interdisciplinary and international collaboration among ecotoxicologists, conservation biologists, policymakers, and other stakeholders is crucial. These collaborations may facilitate knowledge exchange and resource sharing, enabling the development of targeted strategies for pollution mitigation. It may involve implementing stricter regulatory frameworks, convincing stakeholders to safely dispose of the existing contaminant stocks, developing programs to help countries lacking the proper infrastructure to dispose of their contaminated waste, and raising public awareness through education and media campaigns.

It can be disheartening to come across or share discouraging news about the concerning future faced by such charismatic creatures. Alongside chemical pollution, killer whales, like other marine mammal species, encounter a range of additional threats, including plastic

pollution, overfishing, bycatch, reduced prey availability, climate change, chronic stress from noise pollution, vessel traffic, whale watching, and tourist activities like swimming encounters (as observed in Norway). Adding more bad news to the growing list of threats to these killer whales is not something that makes me particularly happy. But I do believe that there are ways to communicate with the public and stakeholders to improve our conservation of the species.

When the public asks me what we can do to "save the whales" from pollution, I usually answer in two parts: first, we can make sure to engage local governments in reducing future pollution and disposing of contaminated waste (as stated previously). Spreading the word, and ensuring peers are aware of the current issues can also help. But there is a second part to this answer: we need to ensure the whales do not go hungry. Starvation alone weakens the immune system of killer whales, increasing their vulnerability to diseases and infections, which can have detrimental effects on their survival and well-being. Additionally, inadequate food availability and malnutrition can lead to reduced reproductive success, lowered fertility rates, and impaired calf survival, posing challenges to the long-term viability of killer whale populations. But the story does not end here… If a killer whale goes hungry, it will have to rely on its own fatty storage for energy, *i.e.*: blubber. Using its own blubber for energy will induce what ecotoxicologists call "remobilization": accumulated pollutants stored in the blubber layer of marine mammals are released back into the animal's circulation and tissues. After entering the bloodstream, the contaminants can be distributed to different organs and tissues throughout the whale's body, which may lead to harmful effects. This process of remobilization can cause elevated levels of these chemicals in essential organs like the liver, kidneys, and reproductive system, posing risks to the overall immune health and reproductive capabilities of the animal. Ensuring the whales have enough food so that they do not remobilize their stored contaminants is crucial, if we wish to avoid this snowball effect.

I believe that we can do good through science communication and outreach. I started my own scientific communication journey in 2020, during the pandemic. I founded my platform, Whale Scientists, to inform the public on various whale conservation issues worldwide. Through consistent efforts and a commitment to providing reliable information, our platform has earned a reputation as a trusted source of knowledge in the field of marine mammal science. This accomplishment fills me with a sense of pride and serves as a testament to the power of effective science communication. We have communicated about multiple species that are relatively unknown and significantly at risk in Southeast Asia for example. Through our efforts, we have successfully inspired individuals to contribute to local non-profit organizations in India and China. These donations now serve a vital purpose in supporting the rescue and conservation efforts aimed at protecting the endangered Yangtze finless porpoises and Irrawaddy dolphins.

I firmly believe that every researcher possesses the ability to effectively communicate their findings to the public. We hold a passion and enthusiasm that has the potential to captivate listeners, conveying the significance and relevance of their research. Our fresh perspectives and innovative ideas can lead engaging discussions and inspire the next generation of scientists. By doing so, we can not only disseminate valuable information but also offer potential solutions for the conservation of marine mammals. Through engaging and accessible communication, researchers can bridge the gap between scientific knowledge and public understanding, fostering a sense of connection and shared responsibility towards the conservation of our ocean's remarkable creatures. Communications with stakeholders and decision makers is also crucial, if we wish to make a difference and advocate for the future of these charismatic creatures (LeFlore et al., 2022; Toomey et al., 2017). Possible ways of communicating our finding are press releases, educational recaps of our scientific findings (for example through infographics), webinars with a public audience, blog posts, podcast interviews, etc. (Wall et al., 2017).

Throughout my doctoral studies, I have made consistent efforts to effectively communicate the findings of my research papers using various communication tools and channels. My aim has been to ensure that both the public and stakeholders gain a clear understanding of our discoveries and the actionable steps that can be taken to assist these animals. Drawing from my three and a half years of experience in science communication, I have compiled a visual guide (Figure 7-1) to assist fellow academics in effectively communicating about the importance of whale conservation.

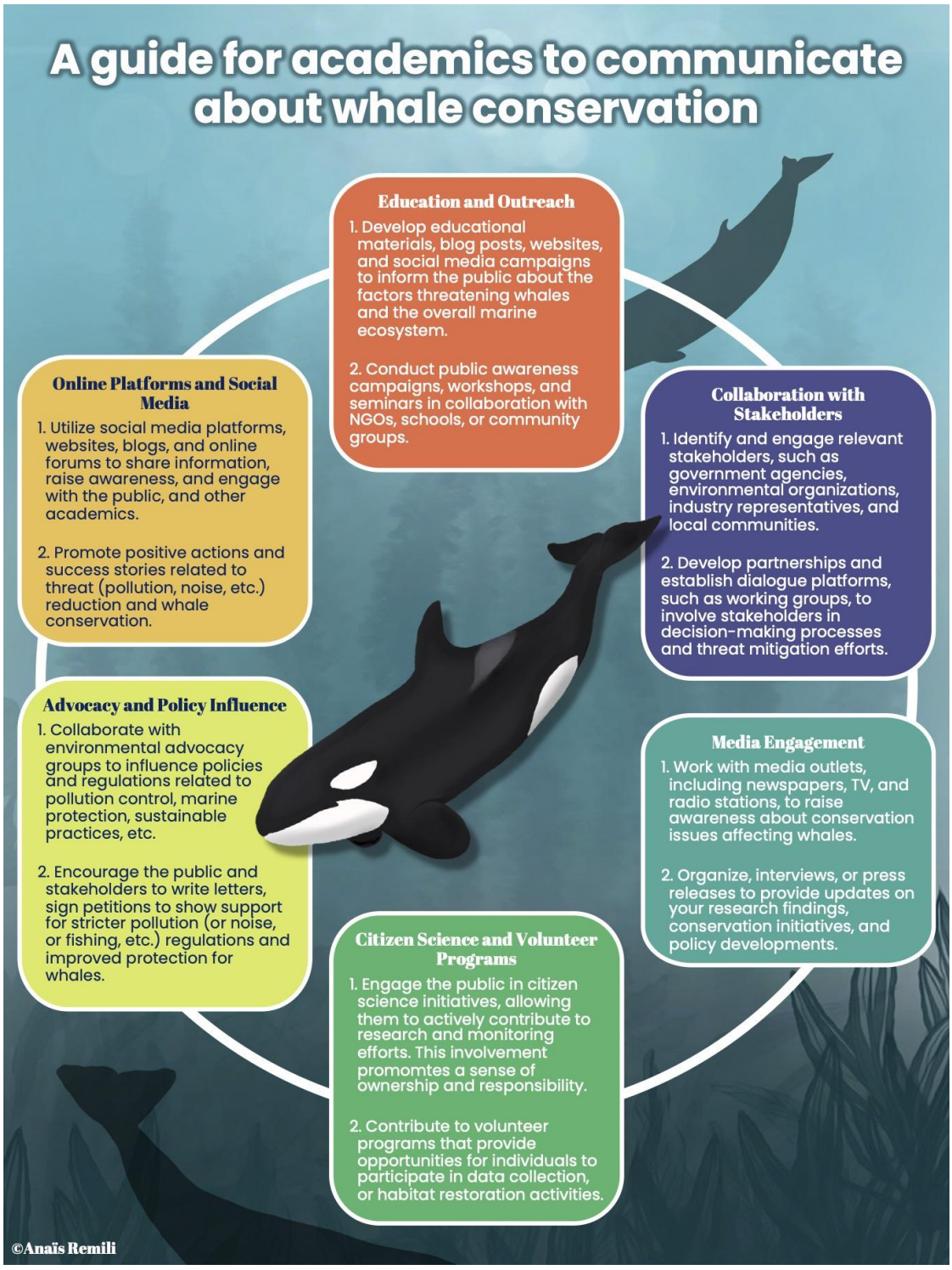

Figure 7-1: An overview of the different strategies academics may use to help with whale conservation. These strategies can be employed individually or in conjunction with others. (Illustrations: A. Remili)

8 CONCLUSION AND SUMMARY

Throughout my PhD journey, my primary objective revolved around comprehending how inter-population and intra-population variations in feeding ecology influenced the risks associated with persistent organic pollutant exposures in North Atlantic killer whales. To tackle this significant issue, I employed multiple high-resolution chemical tracers in samples collected simultaneously across various NA regions, with the aim of deepening our understanding of killer whale feeding ecology.

During my research, several key findings emerged. Firstly, I established that individual feeding specializations (measured through stable isotopes and photoidentification) had a discernible impact on contaminant exposures within the Icelandic killer whale population. Building upon this discovery, I developed a robust method that would allow us to estimate the dietary compositions of wild killer whales, relying on the measurement of fatty acid signatures in skin biopsies. I then applied this innovative method to study approximately 200 North Atlantic killer whales, revealing substantial variations in feeding habits both between and within locations. In this study, I was not only surprised by the noticeable contrast in marine mammal consumption between the Western and Eastern North Atlantic regions, but also by the significant variations in feeding patterns observed within each location. This finding countered the notion of a strict two-ecotype separation among North Atlantic killer whales, challenging previously held assumptions.

Most significantly, our research demonstrated the direct and striking impact of these feeding habits on the accumulation of contaminants in killer whales. The implications are grave, indicating that killer whales relying on marine mammals for sustenance, regardless of whether it occurrs occasionally or regularly, face significantly heightened risks of health issues, including reproductive impairments. These findings underscore the urgent need to consider the health

implications for North Atlantic killer whales in future decision-making processes concerning contaminant management and the conservation of these apex predators.

In conclusion, my extensive and dedicated research journey encompassed a comprehensive exploration of the intricate relationships between killer whale feeding ecology, persistent organic pollutant exposures, and associated health risks. The significance of these findings cannot be overstated, as they provide crucial insights that should inform future conservation strategies and guide policy decisions aimed at safeguarding the well-being and preservation of North Atlantic killer whales.

As a last note, I would like to thank you for reading this doctoral thesis, which encompasses four and a half years of intensive research efforts to understand feeding patterns and contaminants in killer whales. Throughout this journey, I have experienced tremendous growth, both in my scientific knowledge and on a personal level. Pursuing this PhD project was a dream come true, and I poured my heart and soul into it. Today, I am excited to venture into exciting new projects, including the development of QFASA for the critically endangered Southern Resident killer whales, in collaboration with NOAA and DFO. Additionally, I am eagerly looking forward to starting a postdoctoral fellowship in Vancouver next year, where I will explore the impact of contaminants on killer whale health through metabolomic profiling. This is a logical progression of my research, but I remain strongly attached to QFASA and feeding ecology and will keep developing new ways to estimate marine mammal diets (I am currently working on compound specific stable isotope analyses). This is me signing off, and closing this chapter, one that changed my life forever. Thanks again.

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