CONTAMINANT TRACKING THROUGH DENDRO-CHEMICAL ANALYSIS OF TREE-RADII

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' The dreamers of the day are dangerous men, for they may act out their dreams with open eyes, and make it possible. ' T. E. Lawrence 1926, Seven Pillars of Wisdom

DEDICATION

I dedicate this thesis to my partner Christiane Gélinas, with respect and love. Without her constant encouragement, interest, technical advice and probing questions this thesis would never have been realized.

GENERAL ABSTRACT

The research used dendro-chemical analysis of ash tree rings and current year leaf litter to track Cd, Pb, Zn, Cu, Mn, Cr, and Sn spread and cycling from a closed garbage dump-toxic waste site This technique allowed for determination of areal extent, contaminant levels and time period of initial contaminant contact. Only Zn, Sn, and Cu were found in elevated quantities in the xylem wood and Pb in the leaf litter Elemental concentrations of Pb, Sn and Cd in xylem wood and leaves of ash were positively correlated Tin was the only element to demonstrate a clear initial contact period and elemental accumulation with age Significant levels of Cu accumulated in the heartwood while Zn revealed significant but inconsistent accumulation patterns Expected attenuation zones associated with municipal solid waste landfill leachate dispersion were not found, thus the pathway for contaminant dispersion was likely through groundwater flow

An elemental index was developed to facilitate the use of dendro-chemical analysis in periods of suppressed tree growth resulting from environmental pollution.

Nous avons déterminé par une analyse dendro-chimique des anneaux de croissance et de la litière des feuilles tombées durant l'année courante, l'étendue et le cycle de contamination par Cd, Pb, Zn, Cu, Mn, Cr, et Sn provenant d' un ancien dépotoir caractérisé de dépôt toxique L'étude a permis de déterminer l'ampleur de la superficie contaminée, le degré de contamination, et la période initiale de contact entre le milieu et les contaminants Des huit métaux analysés, seulement le zinc, l'étain et le cuivre ont été trouvés en quantité élevée dans le xylème et le Pb dans la litière foliaire Les concentrations de Pb, Sn et Cd dans le xylème et les feuilles du frêne

étaient corrélées positivement. L'étain est le seul élément qui ait démontré clairement la période initiale de contact et une accumulation au cours des années. Une quantité significative de Cu s'est accumulée dans le bois quoique le Zn révèle une accumulation significative mais inconsistante. L'atténuation de la dispersion du lixiviat associée à un lieu d'élimination des déchets solides municipaux n'a pas pu être détectée sur le site à l'étude; par conséquent, les contaminants se sont probablement dispersés avec les eaux souterraines lors de l'écoulement.

Un index des métaux lourds et des oligo-éléments a été développé pour éclaircir la relation entre les concentrations trouvées dans le sol et celles retenues par le bois durant les périodes de croissance ralenties suite au stress causé par la pollution de l'environnement.

General Introduction

Landfills have been, are and will continue to be for some time, the major method of municipal solid waste (MSW) disposal on the North American continent (Bolton et al. 1992). The cleanliness of these sanitary landfills - toxicologically speaking - are functions of management, characteristics of the waste, biological and physico-chemical degradation processes, and age of the site. The normal commingled MSW stream entering a landfill contains varying quantities of toxins from residences (about 1 % by weight [US Congress, OTA, 1989]), businesses and light industries. As with any contaminants placed in the soil environment, there will always be a means and a route for these contaminants to escape (Cairney 1987).

Within Canada and the United States of America, legislation and subsequent regulations have been implemented governing the interim usage of closed landfills. As urbanization pressures escalate, land use changes are focused toward more 'people-oriented pursuits' such as urban greenspaces, housing and industrial developments. The problems associated with contaminant escape and elevated contaminant levels, referred to as environmental pollution (Purves 1977), arising from these closed landfill facilities, then come to the forefront of public awareness.

Within the United States, the US Environmental Protection Agency (US EPA) has forced amendments to the Resource Conservation and Recovery Act (RCRA) which has extended the liability period for closed landfill sites to 30 years and in some cases, to 'in perpetuity' in response to environmental pollution arising from these facilities. This so called 'cradle to the grave' clause, (contained within the RCRA, Subtitle D) echoes scientific reasoning that soil contamination, particularly by trace elements and heavy metals is often an irreversible environmental damage (Kabata-Pendias and Pendias 1984; Purves 1977; Tyler 1981). Questions then arise as to what level of trace element - trace elements being those found in trace quantities within plants - and heavy metal contamination impinges upon man? The demarcation between contamination and the more concentrated contaminant level, pollution, remains nebulous.

The last statement manifests itself in the pollution abatement conundrum " how clean is clean" (Fitchko 1989a). Contamination levels, derived from soil concentrations of trace elements, tend to be arbitrarily set, often based on acceptable potable water standards, and tend to be a broadly based range (Fitchko 1989b).

Determining acceptable ranges of trace element and heavy metal contamination within the soil / plant association - termed contaminant classification or codification - is difficult as these contaminants often behave in broad general patterns due to a multiplicity of influencing factors within the soil matrix (Adriano 1986: Balsberg-Phalsson 1989; Kabata-Penias and Pendias 1984 ; Smith 1973; Tyler 1989). Compounding an individual element's behaviour in the soil is the interaction arising when more than one element is introduced, as is the case when contaminated MSW leachate escapes into the soil. Problems also exist in interpreting relative levels of contamination for different environments because what may be of concern as serious contamination in a rural setting may well be below the ambient concentrations in the urban setting (Fritts 1987; Parker et al.1978).

Garbage dumps and MSW sanitary landfills, both during their exploitation phase or subsequent to closure, are recognized as point sources of groundwater and soil contamination. A cost-effective practical means of monitoring long term trends in environmental change is required. Subtitle D of the RCRA requires the installation of environmental surveillance devices for groundwater and surface water on all existing and operational landfills by 1993. Based on past practices, Canada generally follows the trend set in the United States in the respect to environmental legislation. The impact of this regulation enforcement and the subsequent financial costs will be staggering as indicated by the US National Solid Wastes Management Associations (NSWMA) 1992 survey. Of the over 6,000 operating US landfills, 87 % monitor groundwater and only 58 % monitor surface water. Furthermore, only 88 % of the facillities have the capabilities of testing for heavy metals within the water (Repa et al. 1992). Surveillance of any perturbations of the soil matrix by way of contaminant escape through either groundwater or surface water flow is non existant.

The use of geo-botanical techniques on most annual plants for monitoring contamination of soils will give only a point-in-time and spatial reference rather than a continuum or chronological and spatial history of contaminant spread. This point-in-time analysis of plant tissue offers the best monitoring tool for the soil / plant contamination association, but only if the surveillance program is initiated at commencement of landfill operations and strictly adhers to a yearly schedule. In practice, most landfills within Canada were initiated before environmental legislation came into force, hence, historic monitoring by way of annual or herbaceous plants is impossible. A branch of forestry, dendroecology (Fritts 1987), may offer a possible solution to this chronological environmental monitoring.

Trees are seen by some as an ideal monitoring medium for testing for long term changes in soil trace element concentrations (Barnes et al.1976, Guyette et al. 1987, 89, 91; Kardell and Larsson 1978; Rolfe 1973, Symeonides 1979) for as Lepp (1975) stated in discussion on the subject: " an organism for long-term surveillance must be long-lived, static, must posses mechanisms of metal uptake which can be identified and quantified, must grow on a regular seasonal basis, producing easily detectable and dated annual growth increments, easily and non-fatally sampled". To this statement may be added the term 'ubiquitous in habitat' .

The pairing of growth histories of trees, dendrochronology, with the elemental analysis of tree bole tissue (Ault et al. 1970) for the purpose of chronological environmental monitoring has grown into a valid tool of applied science. This surveillance and analysis system is not without its problems nor critics. To date the majority of the scientific research has focused on elemental aerosol deposits on trees subjected to anthropogenic pollution sources.

The general impetus of this research is two pronged: to further explore the environmental monitoring capabilities of compartimentalized analysis of tree-ring elements coupled with that of growth increments, and to examine the cycling of selected trace and heavy metals from the soil matrix via the tree systems and their return to the ground surface where they may be subjected to human contact. The hypotheses of this thesis would then be: a) that chemical analysis of compartmentalized tree rings in conjunction with tree growth may effectively be used to determine both levels of contamination and the time period of initial contaminant contact; b) that attenuation, the diluting effect of soil binding on the spread of contaminants, will cause the contaminate levels, reflected in compartmentalized tree ring analysis, to decrease with distance from a point source; c) that the tree system is able to draw contaminants from the soil sink and through the metabolic process make these contaminants available by way of natural leaf senescence and abscission to the soil surface. The location chosen to explore these issues for this thesis is a closed garbage dump - toxic waste site located within the boundary of a regional park in eastern Montréal.

Literature Review

I. POINT SOURCE OF POLLUTION : LEACHATE

The knowledge that some, if not all, municipal solid waste (MSW) landfills within Canada have contaminants escaping from their confines is an indisputable fact (Murray and Beck 1990). Leachate is generated by water percolating through MSW and consists of a wide range of soluble organics, dissolved salts and heavy metals (Boyle et al. 1987). By nature, MSW is not homogeneous, as a result any leachate originating from it will incorporate various constituents of the refuse through which it passes. This often pernicious leachate is in constant flux according to: type of refuse; stage of decomposition; biological and physico-chemical processes taking place; and relative amounts of infiltrating moisture. As a result, leachate will vary in composition and concentration both within and between landfill sites as well as over time.

The pernicious leachate is broken into two components, inorganic and organic. Only 18 inorganic and 43 organic compounds are commonly tested for by the US EPA: the actual chemical composition of leachate is as yet unknown, compounded by the fact that its composition is in a dynamic state This research is focused only on the inorganic trace elements and heavy metals Cd, Pb, Zn, Cu, Mn, Ni, Cr, and Sn Generally, these trace elements can be classified into three broad groups based on common plant response characteristics · group I) Cd, Pb; group II) Cu, Zn, Mn, Ni, group III) Cr, Sn (Kabata - Pendias and Pendias 1984). (see appendix 1)

Of the various contaminant escape mechanisms functioning within the MSW landfill, the egress of leachate into the surface layers of the soil by the two prime forces of capillary rise and seasonal mass movement or groundwater rise are of interest. Cairney (1987) noted that the capillary and groundwater rise was greater in the consolidated soil column on the periphery of the landfill than through the unconsolidated waste into the landfill cap. This may move leachate contaminants originating in the waste into the zone adjacent to the landfill. The landfill cap is by design intended to limit the infiltration of water by shedding it onto the periphery zone. The result is a pulse effect as precipitation and / or snow melt shed from the cap periodically flushes contaminants from the landfill periphery into the surrounding soil matrix thus extending the zone of attenuation outwards.

An important consideration is the pH of the leachate, which changes on a continuum with the age of the landfill, from that of acidic to basic, due primarily to concentrations of free volatile fatty acids (Menser et al. 1983). The pH values in the Québec region range from 6.8 to 7.9 (S. Roy, M.Ing.,Ville de Montréal, personal communication, 1992) compared to a mean of 7.1 in Finland (Ettala 1988 b). Experiments using leachates as nutrients applied to the vegetated landfill cap have recorded dramatic increases in soil pH with a concomitant decrease in species diversity of both native and horticultural plants (Chan 1982; Ettala 1988a and 1988b; Gordon et al. 1989a and 1989b; Menser 1981; Menser et al 1983). Saturation with leachate has caused chemical and physical property changes within the soil particularly in the clay fraction causing subsequent clay illuviation (Chan 1982; Menser et al. 1983; Miller and Mishra 1988).

In theory MSW landfill leachate contains many essential plant nutrients but its dynamic state, excesses and unbalanced amounts render its potential use problematic Introduction of leachates with their high COD and BOD into the soil water of the rooting zone of plants impedes the normal physiological functioning of the roots causing physiological abnormalities and often phytotoxic reactions in the plant (Adriano 1986 ; Gilman et al. 1981). Excluding direct leachate contact with leaves, the plants reaction is primarily one of root dysfunction. The high COD and BOD of the pernicious leachate cause both O_2 deficiencies and CO_2 excesses in the root zone. Changes also occur in the water potential and diffusion rates of leaves leading in part to reduced transpiration and photosynthesis rates.

The interactions arising between leachate - with its various components and its chemical and physical properties - and the soil are extremely complex. When the plants are introduced into this milieu the complexities escalate. The interactions resulting from contaminant migration through the soil matrix influence dendroecological monitoring in two aspects: firstly, the growth response of trees as influenced by leachate induced root dysfunction leads to a reduced rate of photosynthesis (Gilman et al. 1981a); and secondly, the elemental makeup of compartmentalized tree-radii as determined by root uptake, translocation and elemental binding within the wood structure of the trees.

II. SOIL SYSTEM

Soil Factors : Influence on Trace Elements and Heavy Metals

A myriad factors interact each exerting an element of control on the ultimate mobility and destination of trace elements and heavy metals. Subordinate to the soil factors of pH, cation exchange capacity (CEC), organic content of the soil are other factors such as soil texture, soil structure and ambient level of pollution (Adriano 1986; Esser et al. 1991; Farrah and Pickering 1978; Kabata-Pendias and Pendias 1984; Pouyat and McDonnell 1991; Tyler 1978).

The increase in soil pH towards alkalinity as affected by leachate egress has major ramification on trace element availability and to some extent their residing time within the soil. The majority of the research in environmental monitoring revolves around soil acidification resulting from acid precipitation, whereas leachate egress increases the soil pH substantially (Chan 1982, Gordon et al. 1989a, 1989b, Menser 1981, Menser et al. 1983) A plethora of information supports the fact that increasing soil acidity makes more heavy metal ions available for uptake by the forest and plant communities but much less information is found on the aspect of heavy metals in situations of increasing alkalinity in the forest environment Uptake is also dependent upon elemental form and soil type

The landfill gas, being a by-product of putrescible refuse and to some extent degradation of leachate, initiates a series of changes within the soil matrix through which it passes. The most significant change in respect to the vegetation within the attenuation zone is the shift from aerobic to anaerobic soil conditions. Concomitant with this is a further O_2 reduction due to methane oxidation causing imbalances, excesses and possibly phytotoxic levels of CO_2 (Barry 1987). Elevated CO_2 levels may lead to decreasing soil pH by the formation of carbonic acid (H_2CO_3). The decreasing levels of pH should make more elements available to plant uptake, but the anaerobic conditions usually negate the anticipated gain in bioavailability.

This transient effect of the landfill gas within the expanding attenuation zone is overshadowed by the arrival of the basic leachate Any decrease in soil pH realized through the passage of the landfill gas is abruptly reversed with the ingress of leachate (Chan 1982; Gordon et al. 1989; Menser 1981; Menser et al. 1983). This leachate-induced increase in soil pH reduces the bioavailability of many elements. Leachate-root contact causes root dysfunction and may result in a reduction in water absorption capacity and possible reduced elemental uptake by the plants (Adriano 1986; Blasberg-Pählsson 1989; Gilman et al. 1981a; Tyler et al. 1989; Vogt and Bloomfield 1991).

At pH approaching 7.0, most heavy metal ions within the soil are usually retained by the clay fraction leading to the conclusion that the sorption process is transformed by decreasing ambient acidity, from a physi-sorption to a chemi - sorption process (Farrah and Pickering 1978). In the management of sludge amended forest and agricultural lands, the mitigation process for heavy metal contaminants within the sludges is to lime the soils to a pH greater than 6.5. This tends to increase the soil retention factor for the heavy metals other than Pb (Adriano 1986, Brady 1984; Harter 1983; Marschner and Wilczynski 1991; Sims and Kline 1991; Weigman 1991).

The CEC is thought to play a more important role in heavy metal availability in acidic soils than in alkaline soils. Harter (1983) indicated that it was not possible to explain the differences in adsorption of heavy metals in alkaline soils based on CEC differences alone.

Organic Matter: Binding Role on Heavy Metals

Organic soils, falling within the broad spectrum of what the US EPA consider as 'wetlands' (Steinberg 1992), form an integral step in either mobilization or occlusion of trace elements. Heavy metal elements, especially Cu, Mn, Ni, Pb, and Zn, have a strong affinity for the organic component within the soil, often forming both water soluble and insoluble complexes (Adriano 1986). Binding of trace elements to organic matter is evidenced in research where mineral soils show strong concentrations of trace elements within the upper 2.5 cm of the soil profile, while organic or 'wetland' soils show a more uniform distribution down the profile (Parker et al. 1978). Organic matter has both cation exchange properties and chelating abilities, leading to an ambiguous role of binding potentially toxic elements while causing mineral deficiencies, especially due to Cu binding.

Soil Texture : Influence on the Behaviour of Trace Elements and Heavy Metals

Soil texture influences the availability of elements as related to the CEC properties of the clay fractions within a particular soil. Both percentages and types of clay affect the potential CEC, hence the behavior of trace elements (Farrah and Pickeering 1978; McBride et al. 1981). Illite (2:1) and montmorillonite (2:1) clay types have a higher CEC than kaolinite (1:1) clay type and as such kaolinite clay allows lower bioavailability of elements for plants (Farrah and Pickering 1978; Kabata-Pendais and Pendais 1984). This principle is a design factor for landfill caps and liners in reducing metal contaminant egress and subsequent transfer of these elements to the plants growing on or adjacent to vegetated landfill caps. Along with the CEC of the clay, available surface area and relative amounts of Fe, Al and Mn oxides influence the soil capacity to occlude trace elements (Adriano 1986).

In hydric, inundated (flooded) soils and fresh water sediments, formation of hydrous oxides of Fe and Mn complemented by biological and chemical oxidation of SO_4^{-2} lead to the formation of insoluble sulfides of trace elements both leading to reduced mobility and bioavailability of most trace elements (Adriano 1986).

III. TREE SYSTEM

Tree Root Morphology and Function

The behavior of tree roots in the man-made environment of the landfill cap are drastically different than that of a natural environment. The hostile landfill cap environment is aggravated by moisture stress, severe heat fluctuations, ingress of landfill gases and contaminants resulting in morphological changes and in many cases mortality of vegetation (Adriano 1986; Barry 1987; Cairney 1987; Carpenter 1988; Dreesen and Cokal 1984; Ettala 1988a, 1988b; Ettala et al. 1988; Flower et al. 1981; Gilman et al 1981,1985; Harris 1987; Neal 1989; Parry and Bell 1987; Sheppard and Dzik 1987; Strerns and Petoyan 1984; Sutton 1991; Watson 1989). Tree root morphology and functions become normalized as distance from the point source of contamination increases and contaminant levels decrease.

The areal extent of the root system along with their density to depth ratios have often been greatly underestimated. This collection system can extend in a circular pattern upwards of seven times the area delineated by an imaginary downward projection of the branch tips, and concentrate upwards of 99 % of the roots within the top meter of soil and upwards of 90 % within the first 10 cm, often extending into the litter layer of the soil surface (Perry 1982; Stone and

Kalisz 1991; Sutton 1991). The rooting patterns of trees are of importance in vegetative management of areas adjacent to landfills in respect to leachate spread.

Along with providing support and a system for collection of nutrients and water, tree roots form a first line of defense in contaminant uptake. Only relatively small portions of the pool of trace elements are bioavailable for plant uptake within the upper layers of the soil. The ability to screen or reduce by selective uptake potentially toxic elements is felt to be species and individual specific (Adriano 1986, Sauerbeck 1991; Tyler et al 1989). This selective mechanism for cation uptake has been widely studied but little understood (Baker 1983). Although the differential uptake of trace elements and heavy metals by trees is considered an important part of the plant's strategy for achieving heavy metal tolerance, their selective transport within the plant is thought to be of greater importance (Tyler et al. 1989). However, this selective transport mechanism breaks down on aging or when critical or threshold contamination levels are surpassed, allowing for unrestricted flow into the xylem sap (Lagerwerff 1967). This last aspect is evident in the nonlinear correlations of tissue concentrations of Cd observed between lightly and heavily polluted areas (Tyler et al. 1989). The question of tolerant versus non tolerant species in respect to the mechanism involved is under debate, although non tolerant species appear to have the ability to store less heavy metals in their root systems than tolerant species (Baker 1983; Thurman 1981). Tolerance appears to be related more to phenotype than to genotype in trees.

Roots in addition to forming a screening defense for contaminants within the rhizosphere function to a greater or lesser extent as retention sites for trace elements particularly the heavy metal fraction (Adriano 1986; Baker 1983; Balsberg 1982; Lagerwerff 1967; Rolfe 1973; Sutton 1991; Tyler et al. 1989). Tyler (1989) presented the concept that small roots (<0.5 cm.) functioned as 'refuse dumps' by uptake and subsequent binding of heavy metal ligands in cell walls of the roots before subsequent root mortality and sloughing off. This cost-benefit strategy of plants in response to a contaminant is energy costly but effectively binds the heavy metals to tenacious organic matter making further bioavailability more problematic in the short term.

Variations in pH (micro) adjacent to roots are precipitated by changes in relative quantities of heavy metals (Chainey et al 1986), root respiration and subsequent ambient concentrations of CO_2 (Sutton 1991), presence of root exudates and root exudates induced by heavy metals (Devlin 1966, Thurman 1981). Increased acidity resulting from root exudates affects the ambient pH adjacent to the roots This micro pH change in the rhizosphere appears to enhance the bioavailability in a limited way within neutral to alkaline soils

The age and size of the roots tend to govern the absorption and retention potential (Baker 1983; Balsberg 1982; Lagerwerff 1967; Sauerbeck 1991, Tyler et al. 1989; Van Assche et al. 1988) The fine young roots (< 0 5 mm) have a larger surface area and contain relatively more trace elements than do the older roots. The aging process in the root tissue appears to cause a breakdown or failure in the mechanism inhibiting translocation of these elements (Lagerwerff 1967; Tyler et al. 1989).The age of the root epiderm influences Ca uptake and interacts significantly in the introduction of trace elements into the root system (Baker 1983; Kabata-Pendias and Pendias 198?) Balsberg (1982) felt the greatest barrier to contaminant movement through the root system to the xylem was the larger transport roots. Baker (1983) indicated a cyclic trend in that transport from roots was an energy-dependent process and could be rate-limited by reduced levels of phloem sap flow induced by photosynthetic decreases resulting from heavy metal interference with enzyme function.

Mycorrhizal Influence on Tree Roots

Mycorrhizal association extends the tree root capacity in two ways: by enhancing nutrient uptake of Zn, Cu, Ni, and Cd, especially in nutrient deficient conditions, and acting as a buffer to the roots by adsorption and occluding trace of heavy metals in conditions of excesses thus preventing entry into the root system (Berg et al. 1991; Daneilson 1985; Koomen et al. 1990) Higher concentrations of contaminants are known to suppress and inhibit both colonization of mycorrhizae (and their dependent trees) as well as hindering their function and vitality (Berg et al. 1991; Chappelka et al.1991; Danielson 1985; Koomen et al. 1991; Martin and Coughtry 1981).

Elemental Deposition in Xylem Tissue

Differential root uptake followed by a subsequential selective transport from the roots initiates the process of elemental deposition within the xylem tissue Critical to this process are the forms and isotopes in which elements enter the plant system, with the ratio of complexed to free ions often governing xylem elemental deposition. Some trace elements and heavy metals, especially those found in a divalent form, are preferentially complexed to amino and organic acids and subject to greater mobility within the plant (Baker 1983 ; Lepp and Dollard 1974b) The binding of elements to xylem tissue is, to a degree, seasonally dependent With the onset of flowering the levels of asparagine and arginine rise sharply to levels approaching 75 % of the total amino acids within the xylem sap. This sharp rise in asparagine and arginine levels shows an equally rapid decline at cessation of flowering Of all amino acids, these two readily form metal ligand complexes, in the case of asparagine with, Cd , Ni, Cu, and Zn and with arginine, Cd, Cu, Ni, Pb, and Zn (Lepp 1975). The high flux of complexed heavy metals by amino acids introduces a large pool of potentially toxic elements into the xylem sap. This situation draws lightly bound trace elements from the root system.

The xylem sap is dynamic in nature changing with vertical position in the tree (Frelich et al. 1989, Robitaille 1981) Subsequently, concentrations of the foliar elements are poorly associated with xylem concentrations in the lower tree (at average sampling height, Breast Height 1.3 m.), although stronger correlations between concentrations in the lower xylem tissue and the soils have been recorded (McClenan and Vimmerstedt 1989). This elemental variation is also subject to both tree age and vigor As such, the age and vigor of the sampled trees are critical in the experimental design to obtain a representative and homogenous sampling from a stand (Fritts 1987).

The process of elemental deposition during formation of the yearly xylem tissue is not well understood (Kardell and Larsson 1978). Although elements are laid down within xylem tissue by a process of binding divalent cations to the negatively charged parts of cell wall, the bound elements represent only those that are present at that specific time (Baker 1983; Lepp and Dollard 1974 b). The elemental concentrations may or may not accurately reflect directly soil concentrations, depending on the specific element, but reflect what was transported in the xylem sap of the tree at that point in time

Upon initiation, xylem tissue is composed primarily of polysaccharides and pectic substances As tissue ages formation of cellulose, hemi-cellulose and lignin occurs. Trace elements complexed to ligands are sequestered through translocation of the xylem sap from the transpiration stream, and are layed down before lignin formation is completed The rate of binding within the xylem tissue is dependent on exposure time and concentration (Lepp and Dollard 1974a , 1974b , Lepp 1975) Elemental distribution within the xylem ring does not appear to be sensitive to the direction of pollutant plume or flow (Baes III and Ragsdale 1981)

Upon reaching the leaves the xylem sap may undergo another specific phase of selective accumulation into the mesophyll and other leaf tissue As a result, the xylem sap at the leaf level, due to sequential selective binding to tissue, is dramatically different from the xylem sap in root pressured exudate (Frelish et al. 1989; Robitaille 1981).

Tree Growth Reduction

In ascertaining possible negative effects of trace elements and heavy metals on the general tree growth, it is essential to determine what is 'normal' radial growth for the particular species within given conditions without detrimental influences (Berish and Ragsdale 1985) Age of the tree is important as radial growth progressively decreases with age (Robitaille 1981) although external factors such as changing hydrological patterns are known to severely reduce bole growth (Arp and Manasc 1988). It is well documented that increasing concentrations of trace elements and heavy metals initially induce root dysfuntion before affecting enzyme function in the photosynthetic process, eventually leading to reduction in biomass accumulation and possibly death if concentrations become phytotoxic (Adriano 1986, Baes III and McLaughlin

1984; Balsberg-Påhlsson 1989; Berish and Ragsdale 1985; Symeonides 1979; Thompson 1981; Tyler et al. 1989; Van Assche and Clijsters 1990).

Translocation of Sap

Lateral movement occurs in both directions between xylem and phloem sap leading to redistribution of trace elements both up and down the stem and into the root system (Lepp 1975; Rolfe 1973). This translocation of exported trace elements and heavy metals from the leaves down the phloem with lateral movement into the differentiating xylem tissue is the premise of dendroecological monitoring of aerosol deposition (Fritts 1987; Hughes 1985; Kardell and Larsson 1978; Robitialle 1981; Rolfe 1973; Ward et al. 1974). Lepp (1975) reported that the major barrier to this downward transport of trace and heavy metals is phloem loading. Once the phloem loading mechanism is surpassed, an unrestricted downward flow of ions and organic assimilates from photosynthesis occurs. This barrier breakdown may be equated to a similar process that occurs within the roots (Tyler et al. 1989). Baes III and McLaughlin (1984) observed, as a result of this surpassed phloem loading mechanism, higher metal concentrations in the living phloem and cambium tissue than in xylem tissue.

The general translocation of the various ions via xylem and phloem sap and subsequent lateral movement between xylem and phloem introduce the major debate regarding dendroecological monitoring as a reliable tool. Possible migration of trace elements and heavy metals between the various annual xylem rings has been questioned (Ault et al. 1970; Baes III and Ragsdale 1981a; Barnes et al. 1976; Holtzman 1970; Kardell and Larsson 1978; McClenahen 1989; Robitaille 1981; Stewart 1966; Szopa et al. 1973). The accumulation patterns of trace elements within the xylem tissue are specific to edaphic conditions, trace elements and heavy metal, species, and individual tree. Any movement within the xylem, to a great extent, is metal specific, because some elements have either greater mobility or inversely greater binding propensity.

On the other side of the debate, various researchers have found lateral movement, primarily in Pb (Ault et al. 1970; Barnes et al. 1976; Kardell and Larsson 1978; Szopa et al. 1973). The lateral movement does not appear to be general, but species specific. Baes III and Ragsdale (1981a) found Pb movement in *Carya sp.*, while McClenahen (1989) found major Pb along with minor Cd and Zn movement in *Pinus sp.* Holtzman (1970) found some evidence of Pb movement but its clarity was 'fogged' by background levels of labeled Pb. Some tree species, especially *Quercus sp.*, lacked sensitivity to low levels of Pb, and thus tend to support Holtzman's (1970) finding of 'signal to noise' problem (Baes III and Ragsdale 1981; Fritts 1987).

Natural heartwood formation has been cited as a catalyst for the movement and storage for various trace elements. As accumulation of toxins within living ray parenchyma cells is an integral step towards the death of the cell and heartwood formation, it has been suggested that the movement of toxic excreta from the live cells across the dead xylem cells supports the 'mobility of metals school' (Lepp 1975; Rolfe 1974). Stewart (1966) postulates that the outer most ring of heartwood forms a barrier to toxins and that these excluded and accumulating toxins will then form an outwardly expanding band of toxins called the heartwood. Only 'excesses' of aromatic and other extraneous substances are subject to movement towards the forming heartwood. Added to this is the observation by Guyette (1989) that moisture content of the heartwood may play a critical role in any possible toxin movement. This issue is further complicated considering that the movement of these toxins may depend on the ionic or complexed form of elements, and the trees' propensity for certain isotopic forms of the elements (Holtzman 1970; Lepp and Dollard 1974b).

The possibility of lateral movement of elements towards the heartwood area of the bole may be complicated by problems or errors in interpretation of data. The illusion of higher concentrations in the older rings in comparison to the younger rings is similar to the failure to 'standardize' for tree growth or basal area increment (Fritts 1987). The annual growth, expressed in tree rings, may be viewed as a sequential addition of growth cones on a vertical axis. With standardizing growth rates the critical factor is the sampled biomass: as a constant growth rate applied to an expanding tree circumference results in newer rings representing a lower proportion of conducting xylem biomass than does the older rings. It is necessary to standardize concentrations based on a per unit of biomass to obtain accurate measurements (Baes III and McLaughlin 1984, Berlish and Ragsdale 1985, Guyette et al. 1989; Robitaille 1981).

Guyette (1989) discussed the compartmentalization of trace elements and heavy metals, subject to tree species, whereby lateral movement may take place but only within restricted distances (or years). This compartmentalization is most evident in conifer trees as they have fewer and shorter ray parenchyma cells and have tracheids rather than vessels (Guyette et al. 1989). Diffuse porous trees appear to present a more complex picture in respect to metal movement within the xylem, across the xylem, or binding to the xylem, than ring porous trees (Baes III and Ragsdale 1981; Kardell and Larsson 1978; Lepp 1975).

Although the lateral movement is not 'zero', some consider that it is small enough to render the xylem tissue as a reliable environmental accumulation indicator (Baes III and Ragsdale 1981; Baes III and McLaughlin 1984; Berlish and Ragsdale 1985; Rolfe 1973, 1974; Sheppard and Funk 1975; Symeonides 1979).

Aerosol Deposition on Trees

An external source that influences the relative concentrations and movements of trace elements and heavy metals within the tree is aerosol deposition. Most aerosol deposition comes from anthropogenic sources associated with metal smelters and vehicular traffic. By nature, trees are excellent filters for aerosols. Decreasing heavy metal concentrations in vegetation are strongly correlated with the increasing distance from the point of pollution (Ault et al. 1970; Barnes et al. 1976; Robitaille 1981; Smith 1971, Ward et al 1974). It was found that the concentration of metals on the vertical axis of the tree was highest between 1.2 and 1.8 m above ground (Barnes et al. 1976; Smith 1971).

Debate exists over the method of entry into the tree of elements contained in aerosol deposits. Ward et al. (1974) observed that heavy metal concentrations decreased rapidly from outer bark to inner bark and concluded that only limited amounts of heavy metals entered the tree through the bark. Ward et al. (1974) suggested that the tree's main uptake of heavy metals was through the roots. Similar results were found by Baes III and Ragsdale (1981) Direct foliar uptake is remote (Arvik and Zimdahl 1974; Sheppard and Funk 1975) where apoplastic movement across the bark into the cambium and subsequent tree cells is supported by others (Barnes et al 1976; Lepp 1975, Lepp and Dollard 1974a). Tree dormancy was not an issue as no effect of season variations in across bark movement was noted (Lepp and Dollard 1974a; Lepp 1975).

Leaf Fall and Leaf Litter

As a consequence of the tree sequestering trace elements and heavy metals within the leaves, the onset of leaf senescence and abscission results in various trace and heavy metals completing the cycle to the forest floor Due to the variability in heavy metal tolerance and accumulation rates within individual species, Van Hook et al. (1977) indicated that heavy metal concentrations in leaf litter vary with species association. Although concentrations of trace elements and heavy metals in leaf litter are often low they are significantly high enough to warrant further studies (Rolfe 1974). Bergkvist (1989) indicated that leaf litter and leaf leachates falling to the forest floor accounted for more than half of the heavy metals taken up by the trees.

Bergkvist (1989) found that the canopy retains only 25 % of the precipitation, the remainder - through fall - is enriched in heavy metal concentrations by 2 to 4 times. The forest floor litter would absorb, in addition to precipitation, trace elements and heavy metals in direct aerosol deposit, through fall, and stem flow. Stem flow accounts for less than 10 % of the precipitation catchment but contains the highest concentrations of metals (Bergkvist 1989).

As freshly fallen leaf litter begins to decompose the leaf cuticle breaks down exposing the cellular structure to passive sorption. Consequently, recently fallen leaves often have higher concentrations of heavy metals than does older litter (Hughes 1981). Berg (1991) noted that the partially decomposed leaf litter adsorbed heavy metals from the soil This situation reverses itself as the leaf litter progresses through the various stages of decomposition (Hughes 1981; Tyler 1981).

Heavy metal pollution within the litter and humus has proven extremely detrimental to the biological activity often causing a complete shift in microbial population structure (Berg et al. 1991; Martin and Coughtrey 1985; Strojan 1978; Tyler 1981, Williams et al. 1977). Pollution causes incomplete humification and a slowdown in remobilization of the trace elements and minerals from the litter (Berg et al. 1991; Martin and Coughtrey 1981; Strojan 1978). The organic matter, particularly the O_2 litter level, resulting from litter humification, contains the highest levels of heavy metals in the soil profile (Adriano 1986; Balsberg 1982; Harter 1983; Lagerwerff 1967; Pouyat and McDonnell 1991; Tyler 1978; Tyler 1981; Van Hook et al. 1977). Apparently there are slight seasonal fluctuations in heavy metals contained within the litter layer possibly dependent on moisture content in conjunction with the water soluble heavy metal fraction (Balsberg 1982; Tyler 1981).

IV. ENVIRONMENTAL MONITORING

As the science of environmental monitoring expands, demands from industries and governments for environmental audits also rise. With many pure sciences migrating towards the realm of applied sciences, the venue will shift from obtained results being technically defensible into the arena of being not only technically but legally defensible (Fitchko 1989b).

Trees, as with plants in general, fall into three broad categories in respect to contact with trace and heavy metals. They are either; excluders, index plants, or accumulators (Peterson 1983). With individual species, response to a pollutant is dependent on the specific pollutant and as such should always be analyzed on a particular soil-plant system basis (Kabata-Pendias and Pendias 1984). Combined elements are confounded within the soil matrix and may render synergystic, antagonistic or cumulative results (Kabata-Pendias and Pendias 1984; Van Assche and Clijsters 1990). In general, the elemental makeup of the tree reflects the rooting medium. Ambient environmental conditions influence the spectrum of availability of nutrients within the rooting medium, yet the effects of the elements once within the tissue are relatively constant (Davis and Beckett 1978)

Trees do not show a rapid response to environmental change, whether it be stand thinning or pollution. The response time, termed lag time, varies with species and may range from one year to upwards of a decade (Kozlowski 1979; Schweingruber 1987; Sheppard and Funk 1975). This situation presents an interpretative grey zone as to when actual pollutant contact is evidenced and best circumvented by compartimentalizing or bulking within a number of years. Complicating this is the degree of pollutant exposed to the tree when viewed on the tree's response continuum from a simple facultative adaptation to acute phytotoxicity (Dickson et al. 1991).

CHAPTER I. DETERMINATION OF DISPERSION, HEAVY METAL CONCENTRATION AND INITIAL CONTACT PERIOD OF ESCAPING LANDFILL LEACHATE FROM A CLOSED GARBAGE DUMP-TOXIC WASTE SITE BY DENDRO-CHEMICAL ANALYSIS OF TREE-RADII

Abstract

A case study was undertaken using dendro-chemical analysis of ash tree rings to track trace and heavy metal contaminant spread from a closed garbage dump-toxic waste site. The use of dendro-chemical analysis allowed determination of areal extent, contaminant level and time period of initial contaminant contact. Analysis was conducted on Zn, Cd, Pb, Ni, Sn, Mn, Cr, and Cu, although only Zn, Sn, and Cu were found in elevated concentrations in the xylem wood at this site. Tin was the only element to demonstrate a clear initial contact time period and elemental accumulation with age. Initial contact marking was not evident with either Cu or Zn, although significant levels of Cu accumulated within the heartwood of the ash while Zn revealed neither a definite nor a consistent accumulation pattern within the xylem wood based on age. An anticipated attenuation zone associated with municipal solid waste landfill leachate dispersion was not found at this site. It was determined that the main pathway for contaminant dispersion was through groundwater flow.

An elemental index was developed to elucidate the relationship between elemental concentrations in the soil and those bound within xylem wood especially during periods of suppressed tree growth as a result of stress due to environmental pollution.

Introduction

Landfilling of municipal solid waste (MSW) has been and will continue to be a major method of waste disposal within North America (Bolton et al. 1992, Cossu 1989; U S Congress [OTA] 1989). Past practices often sited MSW landfills as close as practical to the municipal centre it served As urban sprawl consumes land, sites that contain closed garbage dumps or landfills come under increasing developmental pressure (Bryant et al 1982, Janelle and Millward 1982; Russwurm 1978). Within Canada and the United States of America, legislation and subsequent regulations have been implemented governing the interim use of such closed landfills (Loi sur la qualité de l'environnement 1972 [L.R Q. c, Q-2 sect 65]). As urbanization pressures escalate, changes in land use patterns focus toward more people - oriented pursuits such as urban parks, greenspace and housing developments. Thus, contentious issues concerning public safety associated with contaminant escape from these closed landfill facilities come to the forefront of public concern (Ministère de l' Environnement du Québec [GERLED] 1986, 1991).

During the rapid post-war urban expansion period, regulatory controls regarding siting and environmental safeguards of landfills tended to be lax (Bryant et al. 1982; Burton 1972; Mitchell 1980; Woodrow 1980). As a result a series of problems are now surfacing, best exemplified in the legacy of environmental problems resulting from landfilling of industrial and toxic wastes in the Love Canal region of Niagara Falls, New York. With land development encroaching on areas of closed landfills, the levels and extent of contaminant escape are often unknown and open to speculation. The period of landfill exploitation is, at times, also open to question as is the precise location of dumping areas and possible toxic contents of these sites. The temporal aspect has legal ramifications if liability for site clean-up is questioned during a subsequent environmental land audit (Saxe 1990).

It is generally recognized that the egress of trace and heavy metal contaminated leachate from a closed landfill is a problem. On older above-surface unlined waste disposal sites the area and soils adjacent to the periphery - the attenuation zone - can be biologically, chemically and physically altered by the ingress of pernicious leachate and the landfill gas that is displaced and driven in advance of it (Robinson 1989). Thus the attenuation zone expands with age of the disposal site contingent on the relative quantities of leachate entering the zone and the areal extent of dispersion. Leachate is both diluted and disseminated through this expanding attenuation zone by the flushing effect of water being shed (by design) from the landfill cap.

Various methods exist for monitoring trace (those elements usually found in trace quantities within plants) and heavy metal concentrations particularly in soil and water, but the temporal aspect is absent. A bioassay of herbaceous plant material offers a means of tracking and recording the bioavailability of contaminant spread within the soil but the temporal aspect is contentious as few plants present a long lived medium for chronological histories. Trees, being ubiquitous within the peri-urban region fulfil the criteria for long-term spatial and temporal surveillance as laid down by Lepp (1975). As such, Ault et al. (1970) postulated that chemical analysis of tree rings, compartmentalized into 5 year growth segments, may well reflect ambient trace and heavy metal levels during that time period. This dendro-chemical analysis has been used for air pollution studies on a regional (Baes III and McLaughlin 1984; Bondietti et al. 1989; Dewalle et al. 1991; Frelish et al. 1989; McClenahen et al. 1989) and point or linear source (Barnes et al. 1976; Szopa et al. 1973; Ward et al. 1974) bases as well as a stream pollutant monitoring (Sheppard and Funk 1975). The use of this analysis system in the field of waste management to track contaminant leachate spread is relatively new.

Results obtained by various researchers both substantiate (Baes III and McLaughlin 1984; Berish and Ragsdale 1985; Guyette et al. 1989, 1991; Kardell and Larsson 1978; McClenahen et al. 1989; Robitaille 1981; Rolfe 1974; Sheppard and Funk 1975; Smith 1966) and refute (Ault 1970; Baes III and Ragdsale 1981; Barnes et al. 1976; Hampp and Höll 1974; Lepp and Dollard 1974a; Szopa 1974) the use of dendro-chemical analysis as a valid tool for environmental monitoring of the onset and subsequent levels of contamination by trace and heavy metals. Whatever the source of contaminant, a series of factors interact, and may confound the process of yearly deposition of elements within the xylem tissue (Figure 1 1). The relative amount of elements bound within the xylem tissue are contingent on a number of factors: age of individuals and tree species chosen for analysis, rates and mechanisms of uptake of that particular species, soil factors such as pH, CEC, organic matter content and texture; ambient contaminant level and duration of exposure; variations in binding potential of individual elements, and possible translocation and redistribution within the wood (Adriano 1986; Holtzman 1970; Kardell and Larsson 1978; Lepp and Dollard 1974 a, b; Lepp 1975; Rolfe 1973, Schweingruber 1987; Stewart 1966; Symeonides 1979). Not withstanding the problems associated with the preceding, translocation and redistribution of elements appear to be the most contentious issue negating accurate environmental monitoring by tree ring analysis.

Because of the difficulties involved in quantifying levels of contamination within living trees, various approaches have been utilized. Baes and McLaughlin (1984) presented the concept of xylem accumulation rates, referred to as elemental burden. This was refined by Frelish et al. (1989) from an overall growth rate burden to a sampled biomass burden taken at breast height. The introduction of elevated levels of trace and heavy metals into the soil-root environment by leachate initially results in root dysfunction leading to reduced levels of photosynthesis (Adriano 1986; Balsberg-Påhlsson 1989; Tyler et al. 1989). Reduction in the photosynthetic process is reflected in suppressed increment growth. The previous methods failed to adequately account for elevated elemental concentrations coupled to reduced or suppressed xylem growth.

As such, this study was conducted to: a) evaluate the potential of tree ring analysis for contaminant tracking in both space and time in the vicinity of a closed MSW landfill, and b) explore an elemental index for tree ring analysis that was developed to assess the level of environmental (soil-tree) contamination particularly in periods of suppressed tree growth.

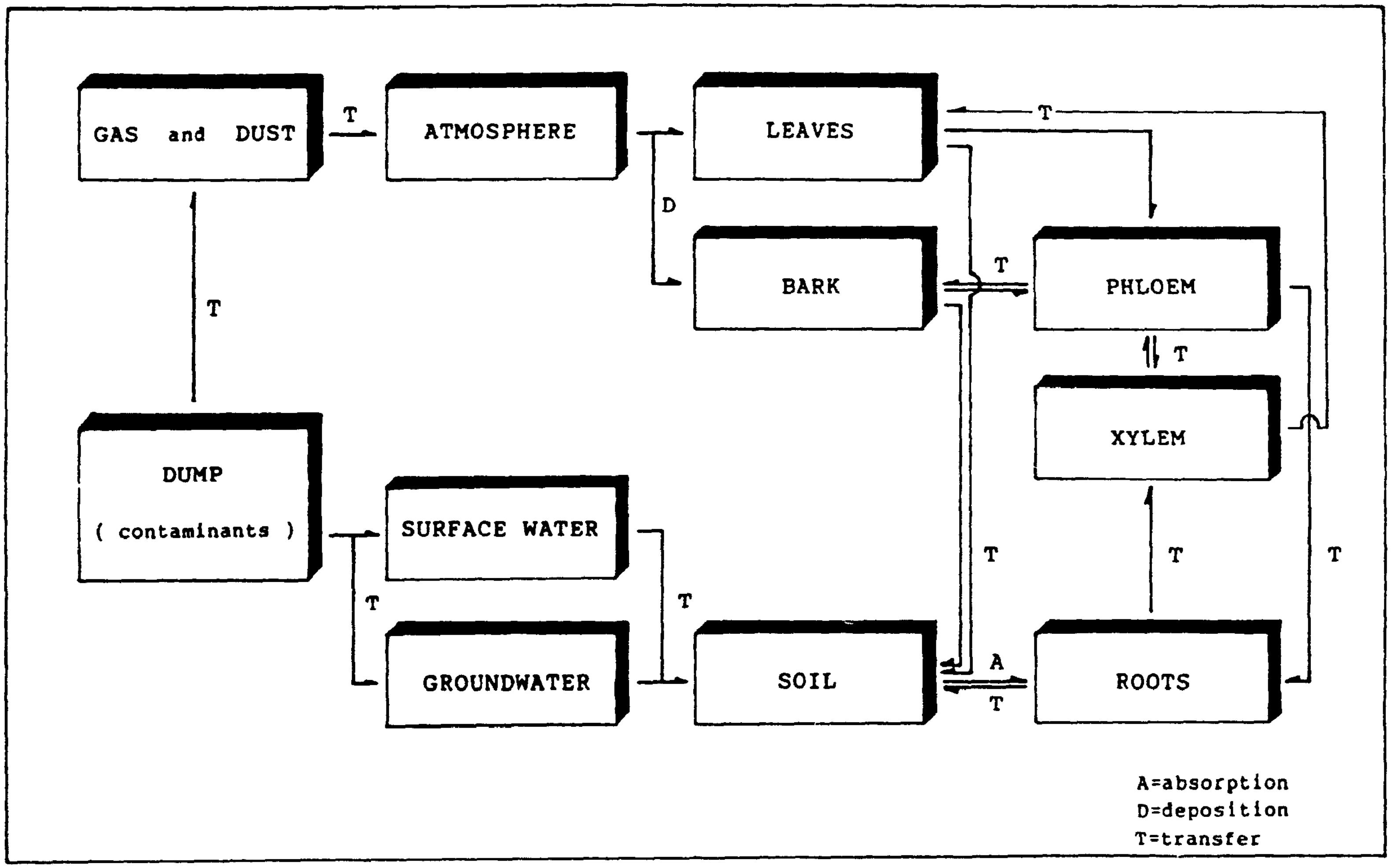

Figure 1.1, Schematic of possible routes of contaminant entry into the tree (after Robitaille 1981).

Materials and Methods

The Landfill

The point source of contamination for this study is a closed garbage dump, classified as a Toxic Waste Site, Category II (Ministère de l'Environnement du Québec [GERLED] 1986, 1991), that lies within the conurbation of Montréal. It is incorporated within the Parc régional de la Pointe-aux-Prairies situated at the northeastern tip of the Island of Montréal (45° 41' N lat., 73° 31' W. long.) This area is noted for its geographical propensity to receive urban solid waste and assorted industrial waste generated within the Communité Urbaine de Montréal (C U M) The waste site came into existence in the early 1960's, first as an unauthorized dumping area, termed Dépôt sauvage within Québec, and as a result of dumping pressure, evolving into an authorized dump in 1969. The site was officially closed in 1972. It is differentiated from a sanitary landfill in that it lacked compaction and daily (working period) covering. The absence of regulatory controls during its exploitation resulted in the disposal of a wide range of domestic and industrial waste. Consequently, various major toxic elements entered the site during this period and escaped into the groundwater leading to the present designation as a Toxic Waste Site. Speculation is that the toxic heavy metal component originates with waste coming from the paint and electroplating industries. Studies conducted by the Ministère de l'Environnement du Québec (1983 - 85) on soils, surface water and the groundwater in the vicinity of the northwest corner of the dump site indicated elevated levels of Zn and Pb (11 700 and 200 $\mu g\ g^{-1}$ dry weight, with critical levels set at 500 and 200 $\mu g\ g^{-1}$, dry weight, respectively in soils / sediment).

The Geology and Soils of the Study Site

The geology tends to confound the study area as the site is situated over the Sainte-Rose fault at the interface of two different ordivician aged sedimentary rock groups: those of the major Trenton limestone group, sub-group Tétrauville, that lie under the southern portion, and the Lorraine clay-schist group under the northern part (Clark 1952). The bedrock is covered by varying depths of gravel till, containing sand and sand-gravel lenses, grading into alluvial clay and silt clay loam. The topography is flat with the soils tending to be imperfectly drained (LaJoie and Baril 1956; Ville de Montréal 1966). On the periphery of the dump, the pH of the soil averages 7.5, concurrent with the effect of leachate flow into the soil at this point The pH decreases rapidly within the initial 50 meters, from the high of 8.3 to a mean of 6 0. The predominant geomorphological feature of this site is the depressional filiform swamps that run in a northwest-southeast direction The pH adjacent to the ephemeral swamps and the major swamp is 6.0; there is apparently no increase in the pH associated with the swamps. The major swamp within the study area appears to coincide, both in position and direction, with the Sainte-Rose fault. As a result of the filiform nature of the swamps, surface water drains in a northwesterly direction from both the forested area and dump site. Groundwater, on the other hand, has been found to be flowing to the northeast at a speed of 220-330 meters per year (Tecwatco Inc. 1986).

The Forest

The dominant vegetation on the northern flank of the dump site is a mixed deciduous forest of primarily a red maple association (*Acer rubrum,* L., *Acer saccharum,* Marsh.; *Fraxinus americana,* L., *Fraxinus pennsylvanica* Marsh.; *Quercus rubra,* L.; *Carya cordiformis* (Wang.) Kosh.). Low herbaceous shrubs and grasses cover the formerly cultivated land lying to the west of the site while the filiform swamp complex contains an association of cattail and purple loosestrife (*Typha latifolia* L.; *Lysimachia salicaria*).

The area used as a control was the Morgan Arboretum of McGill University, located on the western portion of the Island of Montréal some 50 km southwest of the study area. The control site was chosen to match, as closely as possible, both edaphic and biotic conditions, as found in the study area.

Field Sampling

To investigate the extent of contaminant spread through the soil matrix in areas adjacent to this closed garbage dump, a series of three transects 100 meters apart were established commencing at the edge of the landfill cap These transects of 250 meters in length were sub-divided into 25 plots, 10 meters apart A permanent plot centre was established for future reference and as an integral part of the mapping process

Given the composition of the forest stand adjacent to the dump, ash (*Fraxinus sp.*) was chosen for its ubiquitous nature and its ring porous characteristics. The ring porous nature of the wood facilitated ease of reading and measurement for calculation of the basal area increment (BAI) and had suitable elemental binding properties (Kardell and Larsson 1978, Lepp 1975) To obtain samples for botn growth and chemical analysis of the trees, two corings from each of two selected ash trees were taken with a standard 4 3 mm forestry increment bore. Corings were taken at 1 3 meters above ground (Breast Height, BH) on the northwest quadrant of the tree To avoid contamination between core samples, the increment corer and extractor were cleaned with methyl alcohol after each sample Diameter measurements of the trees were also taken at BH for BAI calculations. Due to the nature of the mixed hardwood forest stand, sampling of the ash trees at plot centre was not always possible. In such circumstances, to maintain the distance parameter, trees sampled were at distance and 90° from the transect line Maximum lateral sampling distance considered was 25 m from line centre Site characteristics such as slope, drainage and general vegetative cover were also noted Soil samples were taken concurrently from the top 10 cm of the profile for analysis of pH (in water) (Table 1.1) (Kalra and Maynard 1991)

The annual increment of each tree was measured to the nearest 0 01 mm for calculation of BAI. Subsequently, the core samples were cut into five year segments starting from the outside ring, and the four core samples per plot were pooled for chemical analysis. The bulked core segments of each of the seven age classes covering a 35 year period, were then dried to a constant weight and weights recorded in order to determine the magnitude of bulking required to obtain sufficient material for the chemical digestion process From the review of the literature it was concluded that there was insufficient organic material from the bulked cores to accomplish the wet oxidation digestion method that was chosen to precede the chemical analysis, thus, the samples were again bulked providing a single sample for 0 to 50 meters, 50 to 100 meters etc. while maintaining the established age classes. The pooling of samples negated the possibility of replication of distance on each line.

Laboratory Methods and Analyses

With anticipated low concentrations of heavy metals within the wood, various precautions were initiated to minimize contamination that may affect the results. All glassware and laboratory equipment that came in contact with the samples was acid washed then rinsed three times each in demineralized water then deionized water before the final three rinses with NANOpure water. Samples remained within closed containers unless needed for immediate use. Surgical gloves and clean forceps were used for handling of the wood cores. Core measurements and the sectioning of cores with a surgical steel microtome blade was carried out in an area removed from the soil laboratory where contaminating ambient dust may have been present. Working within the laboratory, the work area was kept as clean and dust free as possible.

Core samples were digested in a nitric-perchloric acid mix (Smith, 1953, 1957). A 0.5 g sample of organic material was placed into standard 100 mL digestion tubes along with 4 mL of HNO_3 (Trace Element Grade) and allowed to stand overnight. This partially digested solution was then heated to 150°C for one hour in the aluminium digestion block heater before adding 1 mL of $HClO_4$ (Trace Element Grade). The temperature of this 4:1 acid digestion mix was slowly raised to 235°C and held for two hours before being allowed to cool. The digested solution was brought to a volume of 50 ml in a graduated cylinder using hot NANOpure water. Digests were analyzed on an Inductively Coupled Plasma - Atomic Emission Spectrometer (Perkin-Elmer Plasma 40, ICP - AES) for Pb, Cd, Zn, Cu, Mn, Ni, Cr and Sn .

The soil pH was measured potentiometrically in a saturated paste method designed for organic soils using a 1:4 soil-to-liquid (10 g soil sample in 40 ml of deionized water) mixture (Kalra and Maynard 1991).

Calculations and Mapping

Ault (1970) indicated that chemical analysis of xylem wood from tree rings may well reflect the ambient levels of elements present in the soils on which the trees grew. This may be the case under normal tree growth, but when environmentally induced stress suppresses tree growth the elemental concentrations bound within the xylem tissue of the trees may not be a fair indicator of ambient soil concentrations. A methodology was developed by Baes and McLaughlin (1984) to express "xylem accumulation rates" or elemental burden in trees. Assuming that BAI is linearly proportional to growth rates, elemental burden is then the product of growth rates and elemental concentrations. This method, however, does not adequately measure the effect of loading of the xylem tissue in periods of suppressed growth coupled with high elemental concentrations. In an attempt to circumvent the effects of growth on elemental concentration and burden an elemental index has been developed. The calculations were as follows:

$$EI = \left[\frac{EC}{MCEC} \times \frac{MC\,BAI}{BAI} \right]$$

Where: E C = Elemental Concentration   M C E C = Mean Control Elemental Concentration
M C BAI = Mean Control Basal Area Increment   BAI = Basal Area Increment

In the following discussion growth rates (expressed as BAI) will be classified as suppressed, near normal and above normal in comparison to the mean growth rate of the control plot. In times of above normal growth rates, high BAI, coupled with moderate elemental concentrations, the index would reflect loading values lower than that of suppressed growth rates coupled with similar elemental concentrations. Two similar BAI rates coupled with a high and low elemental concentrations will give higher and lower index values respectively. By utilizing the control ratios it was felt that a more accurate chronology of the elemental loading of the soil as reflected in the tree rings during periods of fluctuating growth or growth stress could be achieved.

A base map was produced from field measurements taken with a compass and chain, and position corrected using aerial photographs. The map image was produced using standard cartographic techniques. Contour images of the elemental indicies were manually plotted with contour intervals and locations confirmed with computer assisted mapping using Claris™ Resolve™. Analysis of maps included mathematical straight line representation and line softened contours to explore any variations within the visual map representation.

Statistical Analysis

Data were analyzed on the Statistical Analysis System (SAS, Institute Inc. 1985) computer programme. The experimental design was a nested pattern with lines nested within distances, and ages within lines and distances; thus the ANOVA analysis was executed using a nested model. As the assumption of homogeneity of variance in levels of elemental concentrations was not met, data were transformed using Log 10. This calculation reduced the magnitude of the variability of the data but did not completely eliminate lack of homogeneity. Regression analysis was performed between elemental index and age class data to confirm the increase over time in elemental index from the minimum recorded. This regression analysis was conducted only on the newest 5 age classes as determined by the minimum recorded values to confirm the period of initial contamination.

Results

Lines and Distances

The effects of the 3 pooled lines when nested within the 5 distances were significant for Sn and Cu indices, while differences between distances were significant for Zn and Cu indices but not for Sn (Table 1.2).

Table 1.1 Mean soil pH at various distances on different lines.

Distance	Line 1	Line 2	Line 3
25 m	6.6	7.9	7.9
75 m	6.4	5.7	5.8
125 m	6.7	6.0	5.5
175 m	5.8	6.1	5.9
225 m	6.1	6.2	5.9

Table 1.2 Probability levels associated with the effects of distance and transect line on elemental concentrations.

	Zn Index	Sn Index	Cu Index
Distance	0.01	0.13	0.02
Line (Distance)	0.38	0.01	0.04
Age Class	0.75	0.01	0.01
Age class * Distance	0.08	0.08	0.53

Table 1.3 Duncan's new multiple-range test conducted on Zn Index at specified distance intervals on pooled lines.

Distance	Zn Index Means *
25 m	2.66 b
75 m	3.22 b
125 m	1.08 c
175 m	5.01 a
225 m	2.38 b

Zn Index means with a common letter are not significantly different from each other at the 0.01 level of significance

Table 1.4 Linear regression analysis conducted on pooled line data for the five most recent age classes as defined by the minimum recorded value for Sn and Cu.

	Tin (Sn)			Copper (Cu)		
Distance	Pr. > F	Intercept	Slope	Pr. >F	Intercept	Slope
25 m	0.11	3.46	-0.15	0.01	1.20	0.20
75 m	0.30	3.57	-0.10	0.07	1.43	0.22
125 m	0.20	3.52	-0.09	0.01	1.65	0.08
175 m	0.01	3.65	-0.07	0.03	2.26	0.05
225 m	0.01	3.90	-0.09	0.83	2.92	0.01

Table 1.5 Duncan's new multiple-range test on pooled line mean concentrations of the various age classes.

Age class	Sn $\mu g\ g^{-1}$	Cu $\mu g\ g^{-1}$
1991 - 1987	5.87 a	1.49 b
1986 - 1982	1.30 b	2.49 ab
1981 - 1977	0.88 c	2.53 ab
1976 - 1972	0.82 c	2.60 ab
1971 - 1967	0.59 c	5.70 a

Means with a common letter are not significantly different from each other at the 0.01 level of significance

Table 1.6 Variations in elemental concentrations, calculated elemental burdens and elemental indexes selected from Sn data.

Distance	Age Class	Basal Area Increment	E.C. $\mu g\ g^{-1}$	E.B. $\mu g\ g^{-1}$	E.I. $\mu g\ g^{-1}$
1	1991-1987	16.8	14.9	250.0	2.4
1	1986-1982	20.3	6.9	139.7	0.9
1	1981-1977	13.1	6.1	79.3	1.3
1	1976-1972	7.8	0.0	0.0	0.0
1	1971-1967	6.6	6.1	40.2	2.5
1	1966-1962	8.2	3.1	25.0	1.0
1	1961-1957	9.5	5.4	51.0	1.5
2	1991-1987	27.7	7.0	192.8	0.7
2	1986-1982	24.9	3.3	81.3	0.4
2	1981-1977	24.7	0.5	11.6	0.1
2	1976-1972	25.9	3.4	87.2	0.4
2	1971-1967	22.0	2.2	47.8	0.3
2	1966-1962	18.8	9.2	172.6	1.3
2	1961-1957	20.1	4.4	87.7	0.6
3	1991-1987	3.1	46.7	144.3	40.5
3	1986-1982	4.0	4.1	16.4	2.8
3	1981-1977	7.3	2.0	14.3	0.8
3	1976-1972	13.9	6.9	95.4	1.3
3	1971-1967	11.3	0.0	0.0	0.0
3	1966-1962	7.5	7.1	53.0	2.6
3	1961-1957	8.5	4.0	33.9	1.3

E.C. = Elemental Concentration E. B. = Elemental Burden E. I. = Elemental Index

$$E.\ I. = \left[\frac{E.C.}{C.E.C.} \times \frac{C.B.A.I.}{B.A.I.} \right]$$

where: C.E.C. = Control mean E.C. with value of 5.40 ; C.B.A.I. = Control mean BAI with value of 14.65

E.B. = (BAI x E.C.)

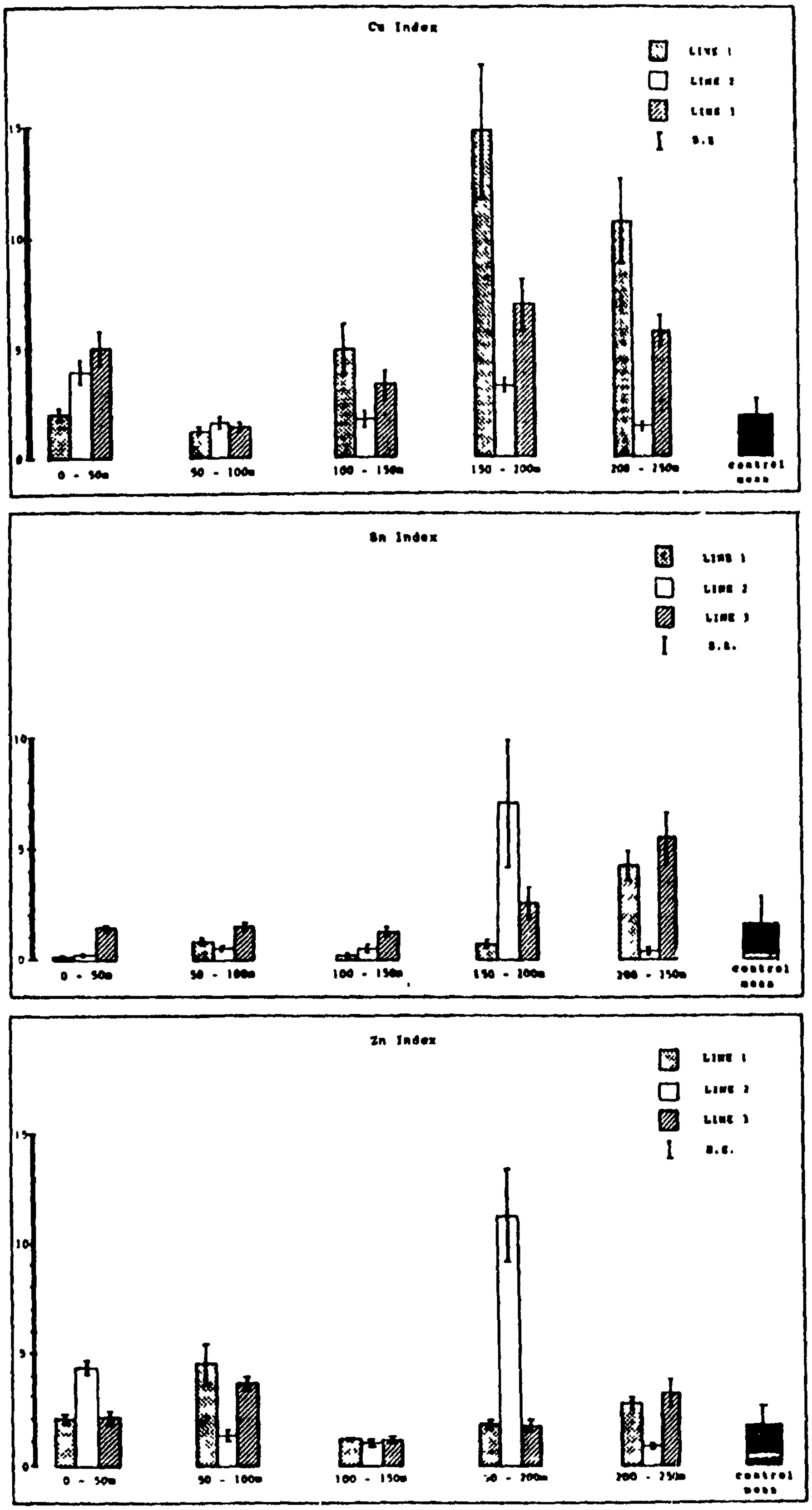

Figure 1.2 Calculated indices for Cu, Sn and Zn plotted at 50 m intervals on the three transect lines.

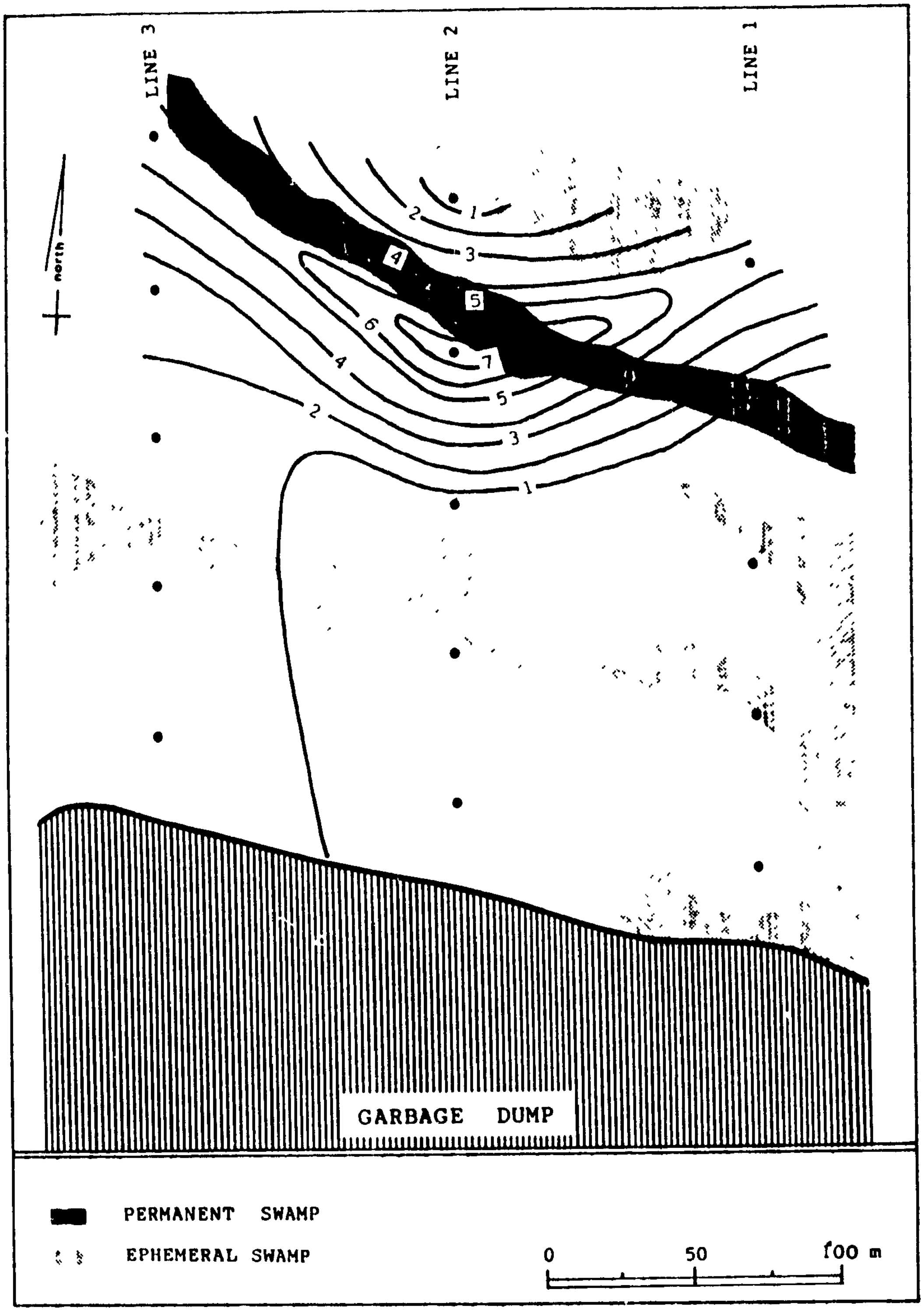

Figure 1.3 Map of the vegetated northern quadrant of the MSW site illustrating the calculated Sn index plotted with one unit interval isobars.

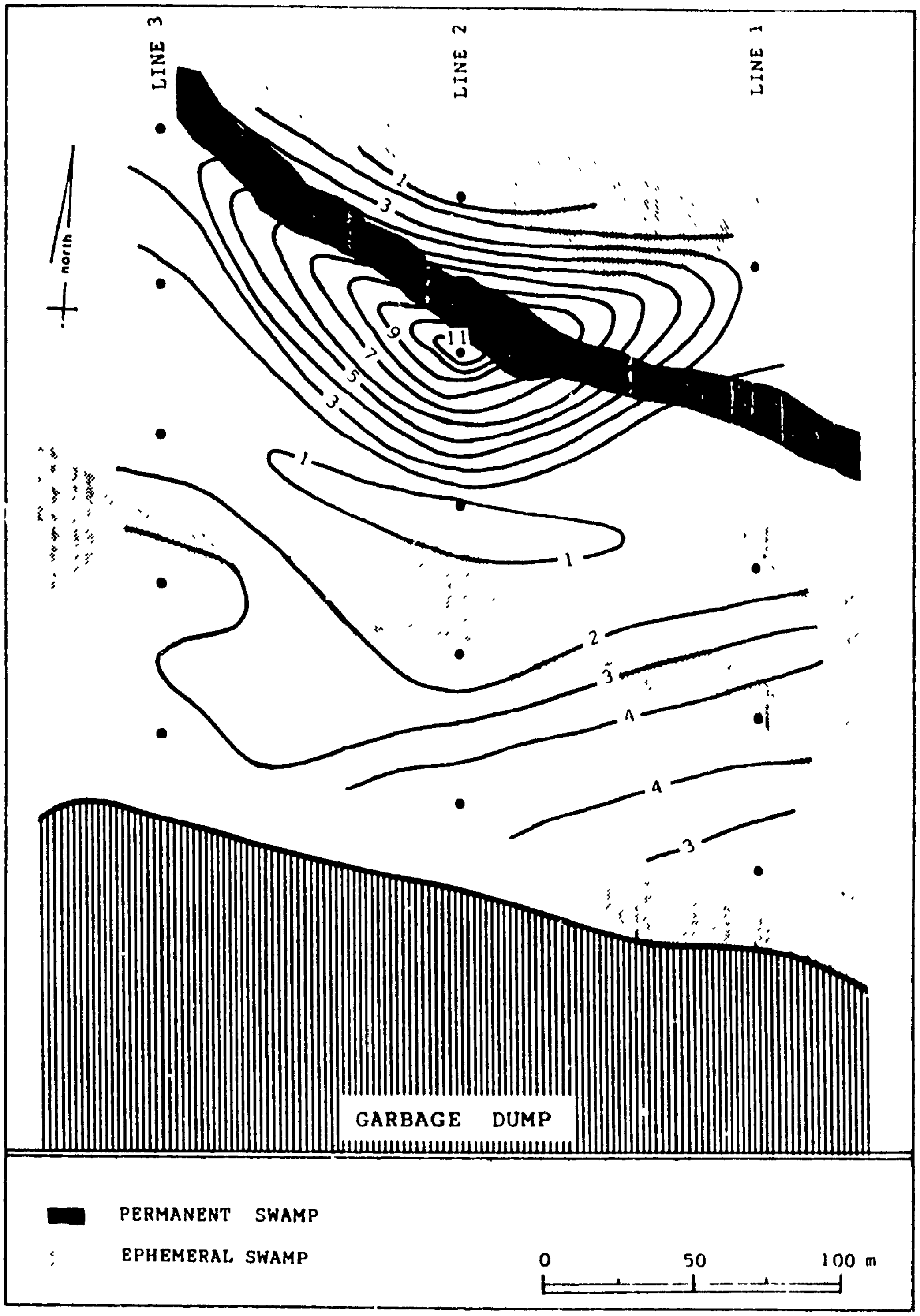

Figure 1.4 Map of the vegetated northern quadrant of the MSW site illustrating the calculated Zn index plotted with one unit interval isobars.

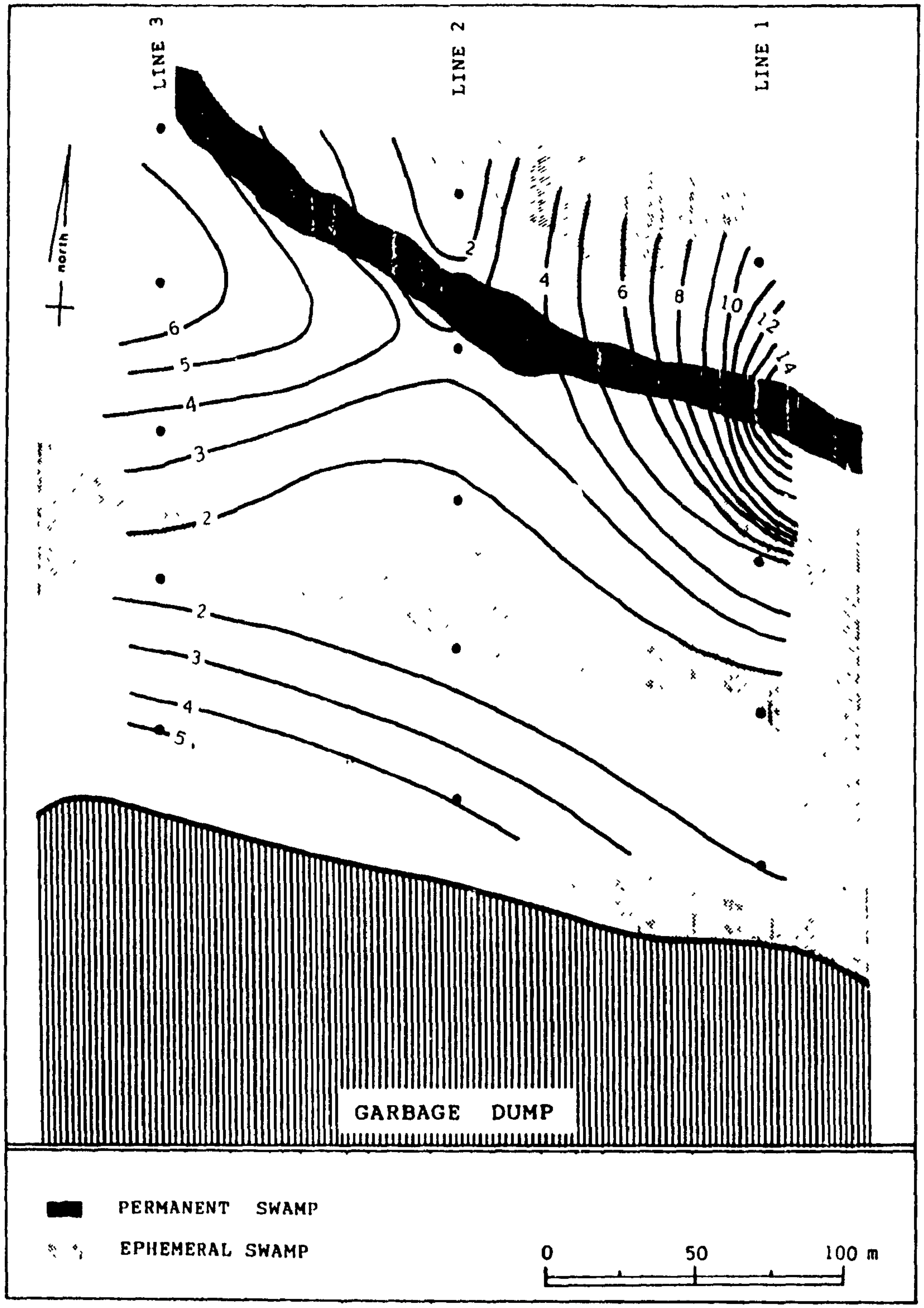

Figure 1.5 Map of the vegetated northern quadrant of the MSW site illustrating the calculated Cu index plotted with one unit interval isobars.

Tin (Sn)

An increase in the Sn index levels occur after 125 m from the dump face, culminating in a crescent shaped ridge of elevated index values centering on line 2 - 175 m and seconded to line 3 - 225 m (Figures 1 2 and 1 3) This ridge parallels the southern boundary of the major swamp on the site but on line 1 - 225 there is also a minor peak. A considerable depression occurs on line 2 - 225 m, located across the swamp from the major elemental peak.

Zinc (Zn)

A depression occurs on the pooled lines at distance 125 m with a high occuring at distance 175 (Table 1.3 , Figure 1.4). The plotted Zn index, similar to the Sn index, reveals a pattern related to the swamp (Figures 1.2 and 1.4) A possible ridge of elevated values, although not statistically significant, may be seen running from the dump face, commencing in the vicinity of line 2 - 25 m and passing through line 1 - 75 m (Figure 1. 4). Similar to Sn, a low point occurs across the swamp at a distance of 225 m on line 2.

Copper (Cu)

Copper patterns deviate from both Sn and Zn indexes although the loci for Cu both occur on the southern boundary of the swamp similar to Sn and Zn (Figures 1.2 and 1.5). Copper forms two peaks; the primary running along line 1 distances 175 to 225, and the secondary peak on line 3 at distances 175 and 225 (Figures 1.2 and 1.5). Line 2 forms a sharp valley between the peaks on lines 1 and 3. A minor gradient, although not statistically significant, possibly exists adjacent to the dump face vectoring in a slight northeasterly direction, parrallel to the groundwater flow. Similar to Sn and Zn, a depressional point exists across the swamp on line 2 - 225 m.

Age Classes

Differences between age classes were significant for the Sn and Cu indices but not for Zn (Table 1.2). Linear regression analysis carried out on the 5 most recent age classes revealed that age class using pooled line data was significant for Sn at distances 175 and 225 m (Table 1.4) with elemental concentrations increasing with age (Table 1.5). For Cu, age class was significant at distances 25, 125 and 175 m (Table 1.4) with elemental concentrations decreasing with age (Table 1.5).

Elemental Index

The results presented in Table 1 6 highlight the differences between elemental concentrations, elemental index and elemental burden. A scenario that has similar elemental concentrations while the BAI varies between suppressed (dist 1, 1971-1967), near normal (dist 1, 1981-1977) and above normal (dist 2, 1991-1987) growth has an elemental index that will be seen to decrease with increasing growth rates while with the elemental burden system the level progressively increases. A relatively constant high BAI coupled with a low (dist 2, 1981-1977), moderate (dist 2, 1986-1982) and higher (dist 2, 1991-1987) elemental concentration lead to both an elemental index and burden steadily increasing but not as much for the elemental index (7 x) compared to the elemental burden (16 x). The effects of a suppressed growth rate coupled with low, moderate and higher elemental concentrations (dist 3, 1981-1977, dist 1,1966-1962, dist 3, 1966-1962) show that both elemental burden and elemental index rise, but with suppressed growth the elemental index would be a higher value.

Other Elements

Although previous research indicated that elevated levels of both Zn and Pb were present in the northwest sector of the attenuation zone - in the vicinity of line 3 distance 0 to 50 m - (Ministère de l'Environnement du Québec, closed file), this study found that concentrations of Pb were not sufficiently different from control levels to be considered. Likewise, Cd, Ni, Cr, and Mn in the tree rings were not significantly higher than the control levels.

Discussion

Age Effect

Tin was the only element to support the hypothesis of initial marking and a progressive accumulation pattern. The most pronounced conformity was at distances of 125 and 225 meters (Table 1 4) where Sn indicated contaminant onset in the 1969 age class (Table 1 5) with a subsequent steady accumulation pattern. The onset of Sn contamination in the 1969 age class (1967 - 1971) would coincide with the supposition (Ministère de l'Environnement du Quebec, closed file) of contaminant dumping of waste from the electroplating and paint industries in and around 1967 - 69 period at this site It may be speculated that dendro-chemical analysis of ash (*Fraxinus sp*) shows potential for determining the onset of Sn contamination moved by leachate Ash also demonstrates an ability to reflect concentrations of other heavy metals within the soil matrix.

Plants respond to the introduction of heavy metals in their rhizosphere in one of three ways; by excluding the heavy metal uptake via various root mechanisms, by reflecting ambient soil concentrations of contaminants within the plant's tissue, or by accumulating the heavy metals within the plant tissue The plants then are known as e cluders, index plants, or accumulators (Peterson 1983). The question arises as to whether ash is an index plant or an accumulator of such heavy metal elements, especially in the case of Sn It is only through further experimentation on Sn uptake and xylem wood binding within by various tree species that this question will be answered

A review of the literature indicates that there have been relatively few studies that have examined Sn with particular reference to the terrestrial environment. Within the aquatic environment, Sn contamination as a result of scaling of marine paints is of growing concern in some estuaries (Bryant and Langston 1992).

Copper, on the other hand, showed accumulation patterns consistent with translocation and accumulation in the heartwood. This translocation of Cu from the newly formed annual xylem tissue into the heartwood is contrary to the findings of Robitaille (1981) where Cu was higher in the heartwood rings than in the newer rings of the control trees, but the tree rings of the polluted stand showed higher concentrations than in the newer rings. As these values were unadjusted for sampled biomass, the results show a definite accumulation pattern in the newer rings. Although Guyette and McGinnes (1987) analysed only heartwood samples, their findings indicated that there was a definite Cu accumulation pattern in the newer rings. Symeonides (1979) concluded that lateral translocation within the bole is of minor significance for Cu and that the increasing levels were strongly correlated with a reduction in growth. All three researchers were working on coniferous trees while this research was conducted using deciduous trees. It is not known if the anatomical characteristics described by Guyette et al. (1991) for the conifers have a major bearing on the lateral translocation of Cu in *Fraxinus sp.*.

The findings of Robitaille (1981) and Guyette and McGinnes (1987) indicate a definite accumulation pattern with Zn, while Symeonides' (1979) research indicated significant lateral translocation. Zinc, in this study, demonstrated neither a definite nor consistent pattern of accumulation based on age.

A 'spiking' phenomenon, as described by McClenahen et al. (1989) and McLaughlin (1987), was evident in analysis of the age class data for Sn, Zn and Cu. These isolated accumulation peaks or high spots were randomly dispersed throughout the data but appeared unrelated to outside or laboratory contamination. The cause of these isolated spikes is unknown.

Attenuation Zone

The clay soil found on our site provides numerous cation exchange sites that increase the contaminant residence time due to binding, and tend to retard the spread of contaminants. An attenuated design landfill site is founded on this principle (Robertson 1989). The effect of leachate causing a dramatic rise in pH appears to have dissipated within 50 meters of the landfill periphery. Rising pH possibly reduced the bioavailability of the elements under study (Brady 1984; Lepp 1975). On lines 2 and 3 the pH near to the dump drops 2.2 and 2.1 units respectively over a distance of 50 meters (Table 1.1). The two pH values of 7.9, lines 2 and 3, (including a reading of 8.3) probably suppressed the uptake of elements in this area. There appears to be little evidence of the effect of landfill gas, possibly due to the almost 20 year time span, on the soil pH and other parameters at the 50 meter demarcation line, although subsequent tree growth analysis revealed a growth reduction to a distance outwards for 100 meters (unpublished data). An anticipated increase in soil pH associated with the swamps was not realized, with the reasons for this being unclear, but it is speculated that surface water may have disseminated leachate through the swamps.

Line Distance Attenuation

It was anticipated that there would be a definite gradient or attenuation effect on the contaminated leachate escaping from the periphery of the dump site and that soil-caused attenuation of the trace and heavy metals would be subsequently reflected in the uptake and sequestering process in the tree rings. The aspect of the higher soil pH values adjacent to the landfill periphery with the attending reduction in bioavailability of the heavy metal elements confound the principle of the attenuation zone and its effect on elemental uptake. Although examination of the mean elemental concentrations recorded at the various distances on pooled lines by the Duncan's new multiple range test revealed no significant differences, Cu did demonstrate a minor but not statistically significant gradient that is visible on the map (Figure 1.5). The minor gradient in Cu was overshadowed by the major contamination highs evidenced between 175 - 225 m.

When elemental indices (to be discussed later) were plotted on a topographic map (Figures 1.3, 1.4 and 1.5), specific spatial patterns emerged indicating a strong association with the surface drainage system. The permanent swamp, running roughly parallel to the dump face, some 175 meters out, appears to either present a barrier or provide a method for contaminant dissemination. From a ground survey of the drainage system, it may be speculated that this concentration of elements is related more to groundwater movement than to surface water flow as surface water movement is limited for the greater part of the year. Furthermore, the concentration may be the result of an upwelling effect caused by the Sainte-Rose fault deflecting groundwater into the upper rooting zone of the trees.

The loci for both Sn and Zn is the 175 meter mark on line 2, with a secondary or lower centre for Sn on line 3 distances 175 - 225. This secondary locus for Sn may be due to lateral movement caused by surface drainage. Copper has different loci than either Sn or Zn, on line 1 distances 175 followed by 225 meters. Although these loci are divergent for the various elements, all lie along the southern boundary of the permanent swamp. This swamp is speculated to be coincidental with the Sainte-Rose fault.

Further examination of the map reveals a possible contaminant movement or stream for both Zn and Cu emanating from the commencement of line 2 and flowing in a northeasterly direction. This point of commencement of the aforementioned suspected contaminant stream supports both the findings regarding the direction of the groundwater flow and its role in contaminant dissemination, and the conjecture that toxic liquid wastes from the paint and electroplating industries were deposited in the northwestern quadrant of the dump site (Ministère de l'Environnement du Québec, closed file). Copper, being the only element that expresses a gradient, shows a distinctly, but not statistically significant , visible slope to the northeast on the map.

Zn - Pb Contamination

The elevated levels of Zn and Pb observed in soils, surface and groundwater (Ministère de l'Environnment du Québec, closed file) were not detected in the tree ring analysis conducted in the vicinity of the boundary zone. Although the anticipated decrease in soil pH caused by the presence of the landfill gas that precedes the leachate should have increased the bioavailability of both Zn and Pb, the possible short duration and effect of the anaerobic soil conditions may have negated this. With increased soil pH induced by the introduction of the leachate a subsequent decrease in bioavailability is noted. Rolfe (1973) indicated that significantly higher concentrations than the 200 $\mu g\ g^{-1}$ (soil Pb) was needed to accumulate significant quantities of Pb in plant material. In addition Guyette et al. (1989) indicated that tree uptake of Pb was negligible at soil pH > 5.0. It should be noted that this 200 $\mu g\ g^{-1}$ level of soil-Pb at our site was only at the critical level. Concentrations above the critical or trigger level are of concern to environmental managers.

Although the levels of Pb recorded within the tree rings were generally below mean control concentrations and as such were not sufficiently elevated in comparison to the control to be considered within this study, a few interesting observations may be drawn from the data Based on distances, Pb showed similar concentration patterns as observed with Sn, Zn and Cu indices (Figures 1.3, 1.4 and 1.5). Elevated levels of Pb were recorded at distances 175 and 225 on line 1; distances 125, 175 and 225 on line 2 , and at distances 75 and 225 on line 3. The phenomenon of isolated 'spiking' in the age class analysis that was observed in Sn, Zn and Cu was also noted with Pb.

Soil concentrations of Zn in the vicinity of the northwestern corner of the dump were in a magnitude of 23.4 times higher than the critical level (Ministère de l'Environnement du Québec , closed file). Anticipated uptake of Zn by the trees was not observed possibly because of the increased soil pH levels caused by the leachate influence. Boyle and Fuller (1987) found that Zn migration through the soil was more dependent on the organic C concentration of the leachate than with the soil characteristics. In determination of the ultimate fate of the recorded high levels of Zn in the soils, a chemical analysis of the tree roots could reveal the degree of sequestering occuring there.

Elemental Index

The relationship between the bioavailability pool of elements in the soil and the rate of sequestering of elements within the xylem tissue of trees is subject to variation due to tree performance or growth. Hence when the tree is subjected to stress related growth impairment a truer evaluation of this soil-tree uptake relationship is achieved when the elemental index is used.

Elemental concentrations and their bioavailability within the soil matrix are generally not subject to rapid yearly fluctuations and as such may be viewed on the long-term basis as either increasing, stable or decreasing. An influx of elements or leachate into the rooting zone may initially increase tree growth due to a fertilizer effect; highly elevated elemental levels ultimately reduce the trees' vigour and growth through a process initiated by root dysfunction. This inital surge in elemental concentration, the fertilizer effect, may best be seen through examination of the elemental burden; when the effects of root dysfunction are expressed in a growth reduction, however the elemental index will be the most advantageous.

Data Interpretation

Various approaches are used to interpret crude numerical data. The foremost method is the use of statistical analysis. With contaminant tracking particularly from a known source, the thematic map fulfils an intermediate step between the actual site and statistical analysis of the collected data. A thematic map is a visual interpretation of a data set applied to an existing base map - an approach either performed manually or via a computer (Geographical Information System) - and, as such, is in itself subject to interpretation of both the maker and the viewer. The prime advantage that thematic maps offer over numeric data presented in a table is the incorporation of the dimensional and subsidiary information comprised in the topographical base map. This additional information may expose associated or subtle trends coincidental with ground features that are not readily apparent in the numeric data. Subsequent statistical analysis may validate these apparent map trends. Conversely, false interpretations of the map by either the maker or viewer may lead to erroneous assumptions. As such, thematic maps are only one of a series of tools that may be utilized to obtain a clearer understanding of the data that has been collected.

Conclusion

The use of dendro-chemical analysis of tree radii for environmental monitoring of heavy metal concentrations and onset of contaminantion in soil is weakly valid. The ubiquitous ash (*Fraxinus sp.*) at this site demonstrated potential for monitoring both levels and onset period of contaminant for Sn but, although expressing contaminant concentration levels for Zn and Cu, it failed to adequately mark the point of onset of the contamination. Copper was found to concentrate in the heartwood while Zn expressed no propensity for accumulation in either the heartwood or newer wood of the ash Anticipated Pb contamination failed to materialize within the tree radii presumably due to the relatively low initial soil contamination levels (200 $\mu g\ g^{-1}$) coupled with near neutral soil pH.

The development and use of the elemental index in conditions of reduced tree growth show potential over elemental burden when utilized in vegetative mapping applied to environmental contaminant monitoring.

On this site the natural attenuation zone as seen in many of the soil based landfills, is poorly defined. The major pathway for contaminant escape appears to be through groundwater flow rather than by surface drainage. Elemental concentrations of Sn, Zn, and Cu, the so called 'hot spots' , are closely associated with the permanent filiform swamp. This swamp may be coincidental with the Sainte-Rose fault; therefore, it may be theorized that contaminants that moved in the groundwater flow may be forced upwards by the fault into the rooting zone of the vegetation near the swamp boundary.

Very few landfills are free from the eventuality of contaminant escape into the surrounding environment. With more and diverse toxins entering the waste stream bound for deposition within sanitary landfills, a greater emphasis must be placed on operational and post-closure monitoring of contaminant escape. Many waste disposal sites within Québec do not have adequate monitoring facilities. Environmental monitoring is a multifaceted operation, with no single method excluding all other surveillance techniques.

Dendro-chemical analysis works well in situations of relatively high contamination, but in low or near-ambient levels concomitant factors such as soil conditions and elemental characteristics may render the results suspect. An extended system of groundwater monitoring wells, once established, provides a cost-efficient multi-range monitoring network especially when contaminant flow data is coalesced with the results of surface water, soils and vegetation monitoring. A sound knowledge of the extent of contaminant spread is a foundation for a contaminant mitigation programme.

Linking Paragraph

Contaminant escape from a closed garbage dump or a MSW sanitary landfill by way of leachate migration is a generally accepted fact. This escaping MSW leachate may or may not present a potential or real risk to the health of the human population or to the quality of the environment. When environmental monitoring acknowledges elevated levels of trace and heavy metals in the soil environment and in the boles of the forest trees surrounding a point source of pollution, questions then arise as to what quantity of these trace and heavy metals are actually made available to human contact through cycling in vegetative material. With closure of garbage dumps and MSW sanitary landfills, the land use function usually shifts to a people-oriented vocation such as urban or peri-urban greenspace This land use shift brings together the land and the people, forcing proximity and hence increasing the potential for contaminant contact. The second paper examines some of these ramifications.

CHAPTER II. POTENTIAL FOR TREES TO CYCLE SELECTED TRACE AND HEAVY METAL ELEMENTS FROM THE SOIL TO THE FOREST FLOOR IN AN AREA ADJACENT TO A CLOSED GARBAGE DUMP - TOXIC WASTE SITE.

Abstract

Trace and heavy metal content of current year leaf litter samples were determined to examine the potential for cycling these elements from the vicinity of a closed garbage dump-toxic waste site. Analysis was conducted using ash and other associated tree species leaves for Zn, Cd, Pb, Ni, Sn, Mn and Cu. Correlation analysis between the elemental concentration of ash xylem wood and ash leaves was also performed, revealing that Pb, Sn and Cd were positively correlated. Heavy metal elements found in elevated quantities in the current year leaf litter were Pb in ash leaves (11.6 times control) and Pb and Zn in other leaves (9.2 and 1.5 times control, respectively). The presence of elevated levels of Pb in the leaf litter nearly two decades after site closure indicates a persistent problem with Pb contamination at this site.

Introduction

The legacy of our industrial society is the waste we produce. Past and current practices are to bury our wastes in landfills. As urban expansion progresses, land once exploited for landfilling operations comes under increasing development pressure for use as urban greenspace, housing and industrial sites (Gilman et al. 1981 b). Present Québec environmental regulations (Loi sur la qualité de l'environnement 1972 [L. R. Q. c, Q-2 sect. 65]) restricts construction of buildings on a closed landfill site for a period of twenty-five years. The interim period often sees the development of people-oriented facilities such as golf courses, botanical gardens, and parks, collectively known as urban greenspaces. Often, the original land use of these greenspace facilities is unknown to the general public (Department of the Environment 1986; Entraco 1990). Rising public awareness over the urban environment coupled with redevelopment often exposes the history of this urban land use mosaic. As such, concern may be raised over public health and safety in the event any contaminants should escape from these closed municipal solid waste (MSW) landfill sites.

It is commonly recognized that the major health hazard associated with commingled MSW and light industrial waste landfills is leachate production and migration (Boyle and Fuller 1987) . This MSW leachate is the result of water percolating through putrescible refuse and inorganic wastes. Leachate contains a wide range of soluble organics, dissolved salts and heavy metals. It is the heavy metal fraction that is of importance in this study (Boyle and Fuller 1987).

On terrain that has been exploited for a landfill site, often only a small percentage of the land base has been denuded of vegetation and actually has waste deposited on it. Thus, a closed landfill site can have varying proportions of unoccupied land on which vegetation may or may not be growing (Ballestero et al. 1990). Upon land use conversion to an urban greenspace it is the existence and quality of this vegetation that has ramifications on a restoration scheme (Meade 1992). The prime attraction of these urban greenspaces is the vegetation. If the existing indigenous vegetation is extensive and deemed acceptable for greenspace development a more reactive management plan will prevail. If the existing vegetation is unsatisfactory or non existent, landscaping will be developed based on a proactive design.

The greater the site disturbance, the greater the scope for a proactive landscaping scheme for restoration of the area. Regardless of the engineering expertise and management care expressed during exploitation of a landfill, "ultimately it is the standard of restoration and the long-term satisfactory performance of the restored land upon which the acceptability of the landfill will be judged" (WHO-IRIS 1991). As such, the amenity use of the vegetation coupled with potential cycling properties must be examined. Inherent in trees is the reaction to heavy metals that might be moved within leachate, as such the plants ability to either exclude or accumulate these toxins are of importance (Peterson 1983). A tree, either indigenous or introduced, that excludes or limits uptake of heavy metals results in these heavy metals remaining in the soil profile (Baker 1983; Thruman 1981) whereas accumulator species may transfer relatively higher quantities, via leaf fall, onto the ground surface and potential human contact.

The purpose of this study was to examine: 1) the potential of trees to cycle heavy metals from the vicinity of a closed MSW landfill and by way of leaf abscission, expose them to possible human contact and 2) the possible correlation between heavy metal content of the leaves and the xylem wood of the originating trees and 3) whether leaf fall can be used for delineating areas of sub-toxic heavy metal concentrations.

Materials and Methods

The Landfill

The point source of contamination for this study is a closed garbage dump, classified as a Toxic Waste Site, Category II (Ministère de l'Environnement du Québec [GERLED] 1986, 1991), that lies within the conurbation of Montréal. It is incorporated within the Parc régional de la Pointe-aux-Prairies situated at the northeastern tip of the Island of Montréal (45° 41' N. lat., 73° 31' W. long.). This area is noted for its geographical propensity to receive urban solid waste and assorted industrial waste generated within the Communité Urbaine de Montréal (C. U. M.). The waste site came into existence in the early 1960's, first as an unauthorized dumping area, termed Dépôt sauvage within Québec, and as a result of dumping pressure, evolving into an authorized dump in 1969. The site was officially closed in 1972. It is differentiated from a sanitary landfill in that it lacked compaction and daily (working period) covering. The absence of regulatory controls during its exploitation resulted in the disposal of a wide range of domestic and industrial waste. Consequently, various major toxic elements entered the site during this period and escaped into the groundwater leading to the present designation as a Toxic Waste Site. Speculation is that the toxic heavy metal component originates with waste coming from the paint and electroplating industries Studies conducted by the Ministère de l'Environnement du Québec (1983 - 85) on soils, surface water and the groundwater in the vicinity of the northwest corner of the dump site indicated elevated levels of Zn and Pb (11 700 and 200 $\mu g\ g^{-1}$, dry weight, with critical levels set at 500 and 200 $\mu g\ g^{-1}$, dry weight, respectively in soils / sediment).

The Geology and Soils of the Study Site

The geology tends to confound the study area as the site is situated over the Sainte-Rose fault, interfaced by two different ordivician aged sedimentary rock groups; those of the major Trenton limestone group, sub-group Tétrauville, that lie under the southern portion, and the Lorraine clay-schist group under the northern part (Clark 1952). The bedrock is covered by varying depths of gravel till, containing sand and sand-gravel lenses, grading into alluvial clay and silt clay loam. The topography is flat with the soils tending to be imperfectly drained (LaJoie and Baril 1956; Ville de Montréal 1966). On the periphery of the dump, the pH of the soil averages 7.5, concurrent with the effect of leachate flow into the soil at this point. The pH decreases rapidly within the initial 50 meters, from the high of 8.3 to a mean of 6.0. The predominant geomorphological feature of this site is the depressional filiform swamps that run in a northwest-southeast direction. The pH adjacent to the ephemeral swamps and the major swamp is 6.0; there is apparently no increase in the pH associated with the swamps. The major swamp within the study area appears to coincide, both in position and direction, with the Sainte-Rose fault. As a result of the filiform nature of the swamps, surface water slowly drains in a northwesterly direction from both the forested area and dump site. Groundwater, on the other hand, has been found to be flowing to the northeast at a speed of 220-330 meters per year (Tecwatco Inc. 1986).

The Forest

The dominant vegetation on the northern flank of the dump site is a mixed deciduous forest of primarily a red maple association (*Acer rubrum*, L., *Acer saccharum*, Marsh.; *Fraxinus americana*, L., *Fraxinus pennsylvanica* Marsh.; *Quercus rubra*, L ; *Carya cordiformis* (Wang.) Kosh,.). Low herbaceous shrubs and grasses cover the formerly cultivated land lying to the west of the site while the filiform swamp complex contains an association of cattail and purple loosestrife (*Typha latifolia* L.; *Lysimachia salicaria*).

The area used as a control was the Morgan Arboretum of McGill University (Morgan Arboretum control), located on the western portion of the Island of Montréal some 50 km southwest of the study area. The control site was chosen to match, as closely as possible, both edaphic and biotic conditions, as found in the study area.

Field Sampling

To investigate the amount of contaminant cycling by trees in the area adjacent to this closed garbage dump, three transects 100 meters apart were established radiating from the edge of the landfill cap. These transects of 250 meters in length were sub-divided into 25 plots, 10 meters apart. A current year leaf litter sample (1991), 1 m^2 (0.404 m radius) in area, was collected from the forest floor at each plot centre. A permanent plot centre was established for future reference. Site characteristics such as slope, moisture and general vegetative cover were also recorded.

Laboratory Methods and Analyses

The leaf samples were forced-air dried to 70° C to retard decomposition and to facilitate both cleaning and sorting. The hand-cleaned leaves were sorted into two classes; ash (*Fraxinus sp.*), and all other leaves. The leaves were redried to a constant weight, weighed to determine if pooling was necessary and ground in a cyclone grinder.

With anticipated low concentrations of heavy metals within the leaves, various precautions were initiated to minimize outside contamination that may skew the results All glassware and laboratory equipment that came in contact with the samples were acid washed then rinsed three times each in demineralized water then deionized water before the final three rinses with NANOpure water. With the ever present problem of cross contamination by leaf dust during the grinding procedure, extreme caution was taken to clean the grinding equipment after each sample Samples remained within closed containers unless needed for immediate use Surgical gloves and clean spatulas were used for handling the ground leaves While working within the laboratory, the work area was kept as clean and dust free as possible

The ground leaf samples were digested in a nitric-perchloric acid mix (Smith 1953, 1957). A 0.5 g sample of organic material was placed into standard 100 mL digestion tubes along with 4 mL of HNO_3 (Trace Element Grade) and allowed to stand overnight. This partially digested solution was then heated to 150° C for one hour in the aluminium digestion block heater before adding 1 mL of $HClO_4$ (Trace Element Grade). The temperature of this 4:1 acid digestion mix was slowly raised to 235° C and held for two hours before being allowed to cool. The digested solution was brought to a volume of 50 mL in a graduated cylinder using hot NANOpure water. Digests were analyzed on an Inductively Coupled Plasma-Atomic Emission Spectrometer (Perkin-Elmer Plasma 40, ICP - AES) for Pb, Cd, Zn, Cu, Mn, Ni and Sn .

The soil pH was measured potentiometrically using a saturated paste method designed for organic soils using a 1 : 4 soil-to-liquid (10 g soil sample in 40 ml of deionized water) mixture (Kalra and Maynard 1991).

Calculations

Working from a leaf litter elemental concentration, an annual cycling budget was calculated to estimate the load of elements returned to each hectare of ground surface per year

The relationship between the actual trace and heavy metal content of the ash leaves and those found within the recent ash wood (1987 - 1991 year period) was also examined. A detailed wood analysis was carried out in 1993 (Reeves 1993, unpublished).

Statistical Analysis

Mean, ' t ' test and standard errors along with Pearson's coefficients of correlation between the ash leaf elemental concentrations and the current ash xylem wood (1987 - 1991 period) elemental concentration were calculated using the Statistical Analysis System (SAS, 1985) computer programme. A probability level of 0 05 was used unless otherwise specified.

Results

The mean concentrations of the trace and heavy metals examined appear in Table 2.1 . The elements found in elevated quantities in our study are MSW site (ash) Pb, and MSW site (other) Zn and Pb (Table 2.1 and Figure 2.1). Manganese levels in the MSW site (ash) and Cu in the MSW site (other) are significantly lower than their respective Morgan Arboretum controls.

When the various elements presented in Table 2.1 were ranked in descending order of concentrations, the following pattern emerged:

Morgan Arboretum Control (ash & other)	Mn > Zn > Cu > Ni > Sn > Pb > Cd
MSW Site (ash)	Mn > Zn > Cu > Pb > Ni > Sn > Cd
MSW Site (Other)	Mn > Zn > Cu > Pb > Ni > Sn > Cd

Correlation coefficients between elemental concentrations found in the current five year segment of ash xylem wood (1987 - 1991) and the freshly fallen ash leaves (1991) were not significant for Zn, Cd, Ni and Mn but were significant for Pb, Sn and Cu ($p < 0.15$) (Table 2.2).

When Table 2.3 is viewed in conjunction with Table 2.4, the results indicate comparable quantities of elements returned to the soil surface at the MSW site and Morgan Arboretum control locations except for Pb which is elevated in our MSW study site.

Table 2.1 Mean elemental concentrations in leaf litter sampled in the fall of 1991. Mean ± S.E.

	Elements ($\mu g\ g^{-1}$)						
	Zn	Cd	Pb	Ni	Sn	Mn	Cu
Ash Leaves							
MSW site	32.7 ± 3.1	0.23 ± 0.0	6.5 ± 0.1	4.2 ± 0.3	1.1 ± 0.2	127 ± 14	24.3 ± 0.5
Morgan Arboretum (control)	26.3 ± 3.5	0.19 ± 0.1	0.5 ± 0.1	2.7 ± 0.4	0.8 ± 0.4	276 ± 38	30.0 ± 1.4
Other Leaves							
MSW site	41.6 ± 1.4	0.28 ± 0.0	6.3 ± 0.1	2.6 ± 0.1	1.1 ± 0.1	433 ± 38	15.5 ± 0.3
Morgan Arboretum (control)	28.7 ± 0.8	0.38 ± 0.1	0.7 ± 0.3	2.0 ± 0.5	1.1 ± 0.4	429 ± 57	23.1 ± 0.8

Table 2.2 Correlation between ash xylem wood (1991 - 1987 time period) and ash leaf litter (1991) elemental concentrations.

	Elements						
	Zn	Cd	Pb	Ni	Sn	Mn	Cu
Pearson Correlation Coefficient	-0.02	-0.01	0.38	-0.15	0.47	-0.01	0.48
Pr.> |R| under Ho	0.96	0.96	0.15	0.60	0.07	0.97	0.06

Table 2.3 Quantities of selected trace and heavy metals cycled to the surface in 1991 at the MSW site and the Morgan Arboretum control area in leaves of ash and other species.

	Elements (in g ha^{-1} yr^{-1})						
	Zn	Cd	Pb	Ni	Sn	Mn	Cu
Ash Leaves							
MSW site	90.7	0.6	18.0	11.7	2.9	351.0	67.4
Morgan Arboretum (control)	72.8	0.6	1.6	7.5	2.1	764.5	83.3
Other Leaves							
MSW site	115.3	0.9	17.3	7.1	3.0	1 203.5	42.9
Morgan Arboretum (control)	74.5	1.5	1.9	5.7	3.0	1 191.0	63.9
Total							
MSW site	206.0	1.4	35.3	18.8	5.9	1 554.5	110.3
Morgan Arboretum (control)	152.3	2.1	3.5	13.2	5.1	1 955.5	147.2

Table 2.4 Comparison of elemental concentrations in autumn foliage of the Morgan Arboretum control, MSW site, Turkey Lakes Watershed and Hubbard Brook Experimental Forest.

	Elements ($\mu g\ g^{-1}$)						
	Zn	Cd	Pb	Ni	Sn	Mn	Cu
MSW Site	37.1	0.3	6.4	3.4	1.1	280.3	19.9
Morgan Arboretum (control)	27.5	0.3	0.6	2.4	0.9	352.6	26.5
Turkey Lakes Watershed	30.0	0.3	0.9	6.6	-	841.0	9.8
Hubbard Brook Experimental Forest	273.0 (40*)	0.6	-	-	-	2,256.0	7.3

* concentration without the influence of Betula sp.

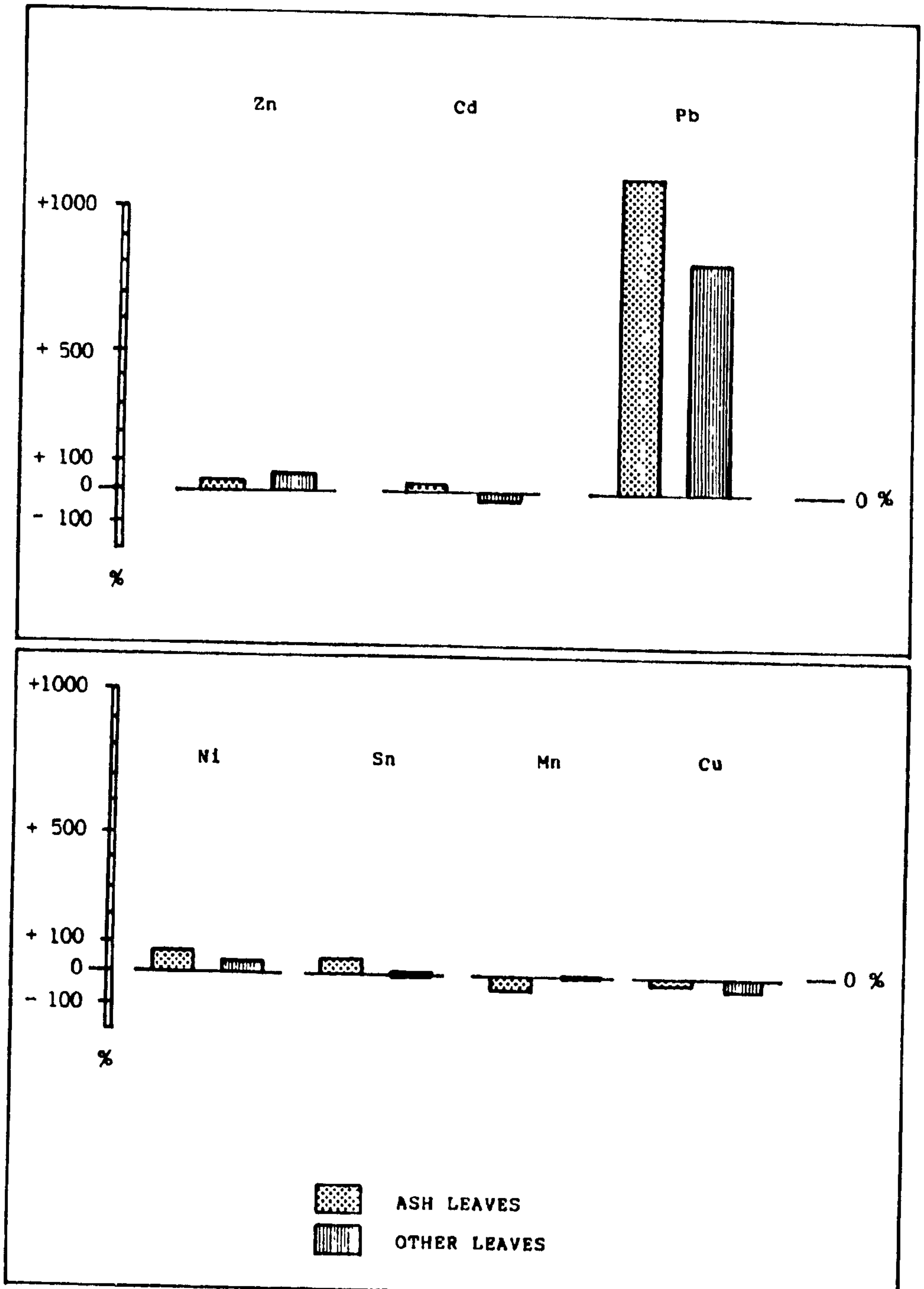

Figure 2.1 Variations in leaf elemental concentrations for the MSW site ash and other leaves given in a percentage above or below a zero based control.

Discussion

A certain degree of contamination of the leaves may be assumed to have occurred through direct atmospheric deposition. Small amounts of Cd and Pb at the Morgan Arboretum control may have originated from the light and heavy industries located in the Beauharnois-Melocheville, Québec, areas in the vicinity of the control site (Elkem Metal, Chromasco, Alcan Ltd., Tricil Ltd.) (Zayed and Loranger 1992). The study area will likewise be affected by limited deposits originating from the petroleum refining industries located in eastern Montréal Both areas are situated about one kilometre from a major autoroute. For this reason there was no attempt to delineate between the relative amount of ground or atmospheric contaminants with this study. The original assumption for this was that the level of air borne contaminants would be similar at the site and at the control area.

The overall elemental pattern for the Morgan Arboretum control is similar to the pattern at Turkey Lakes Watershed, Ontario (Hogan and Morrison 1988) conducted on the distribution of trace metals within the above ground phytomass. As for the study site, the major change in elemental concentration is found with Pb.

From Table 2.1 it appears that the major contaminant of the ash leaves at the MSW site is Pb, being 11.6 times that of the Morgan Arboretum control (ash) levels. Analysis of all the other leaves collected at the MSW site reveals elevated levels of Pb and Zn, respectively being 9.2 and 1.5 times higher than the Morgan Arboretum control (other). Levels of Mn in MSW site (ash) , and Cu in MSW site (other) are significantly lower than their respective control values.

Correlation

As samples of xylem wood for species other than ash were not taken, correlation coefficients were determined between ash leaves and ash wood only. Between the ash wood and leaf concentrations, the elements Pb, Sn and Cu are all positively correlated. The major contaminants found in the xylem wood of the ash trees at the MSW site were Zn, Sn and Cu (Reeves 1993, Chapter 1). These contaminants were not found in elevated levels in the ash leaf analysis. This variation between the wood and leaf analysis may indicate that Zn, Sn and Cu are preferentially bound by the woody tissue of the ash trees before reaching the foliage.

If the correlation coefficient was strong it could eliminate sampling problems encountered with taking corings of xylem wood for chemical analysis. This lack of a strong correlation coupled with leaf drift negates the possibility of contaminant mapping of the soil concentrations of heavy metals via the leaf litter.

Redistribution of Elements within the Heavy Metal Profile

Leaves, in reality, contain only a small portion of the trace and heavy metal pool contained within trees. The observations of Van Hook (1977) revealed that the foliage made up around 3 % of the above surface vegetation biomass, compounded with the findings of Parker et al. (1978) that the total above ground vegetation accounts for < 1% of the total metal pool in the urban ecosystem leads to the conclusion that the foliage will contain relatively small amounts of heavy metals. Parker et al. (1978) states " living vegetation will not be effective as sinks for heavy metals or as pumps to remove metals from contaminated sites." Data from Table 2.3 appears to confirm the statement by Parker as the quantities of elements returned to the surface were relatively small. The concept that vegetation will cycle large quantities of trace and heavy metals from a landfill or adjacent areas onto the surface where possible human exposure exists, must be viewed with caution.

Site Comparisons

The information from Turkey Lakes Watershed (TLW) (Hogan and Morrison 1988) and Hubbard Brook Experimental Forest (HB) (Wittaker et al. 1979) presented in Table 2.4 reveals that concentrations of Zn at the control, MSW site and TLW are comparable. Hubbard Brook concentrations for Zn are listed at a mean of 273 $\mu g\ g^{-1}$ although this figure is skewed by the high stand composition of *Betula sp.* which is an acknowledged Zn accumulator (Hogan and Morrison 1988; Wittaker et al. 1979). If the *Betula sp.* are removed from the calculations the Zn level is reduced to a comparable level of 40 $\mu g\ g^{-1}$. Zinc is not an element that is strongly back translocated into the tree upon senescence of the leaves in the autumn (Guha and Mitchel 1966). Although Zn is slightly elevated in the leaves at the study site, it is not considered to be in quantities that will present problems to humans or the vegetative community.

Copper concentrations at the Morgan Arboretum control and the MSW site show a two fold increase over TLW and HB values. As Cu availability generally increases with decreasing soil pH and as both the Morgan Arboretum control and the study area have equally high levels of Cu, it may be speculated that the high levels of Cu recorded at the Morgan Arboretum control and the MSW site (soil pH of around 6.0) may be due to a higher levels of Cu in the parent rock material and the soils of the Montréal region and less to a contamination problem at the MSW site.

The Mn levels in the leaves vary considerably between the various sites with a low value recorded in the MSW site (ash). The reasons for this may lie with different pH and Redox potential in the soils at the various sites.

Guha and Mitchel (1966) indicated that Pb was progressively accumulated in the leaves over the summer with slight back translocation upon leaf senescence in the autumn. Freshly fallen leaves at the MSW site have Pb that is elevated significantly in comparison to either the Morgan Arboretum control or TLW. Hogan and Morrison (1988) indicated that their Pb findings at TLW were approximately 1/3 to 1/4 of those recorded at HB. As both HB and TLW have soils listed as acidic the elevated levels found at our site are of interest. The mean soil pH at the MSW site was 6.0, with more basic values recorded near the periphery of the garbage dump. These recorded pH values are representative of conditions leading to reduced Pb uptake by the plants. As such, the recorded high levels of Pb in the freshly fallen leaves may be indicative of a Pb contaminant problem at the MSW site.

Previous xylem wood analysis (Reeves 1993, chapter 1) revealed that Pb was not significantly elevated in the bole even though Pb was an acknowledged contaminant at the MSW site (Ministère de l'Environnement du Québec, closed file) . Various researchers found however that foliage contains lower relative quantities of Pb than is found in the bole (Adriano 1986 ; Van Hook 1977). The high leaf concentrations of Pb compared to the non-significant levels found in the xylem wood counter the findings of both Adriano (1986) and Van Hook (1977). The reason for these conflicting results is not clear.

The question over the collection date of the leaf samples as influencing the relative content of Pb must be considered in future studies such as this one. Lead content found in the fallen leaves may have been augmented by leaf leachate and subsequent absorption from the ground surface Lead concentrations in the O_2 litter level are generally higher than those in the O_1 level (Adriano 1986; Van Hook 1977). Berg et al. (1991) found that newly fallen leaf litter absorbs heavy metals from the soil and lower leaf litter, O_2 level, on the forest floor during the early stages of leaf decomposition thus augmenting the heavy metal concentration in the O_1 level. Tukey et al. (1958) reviewed the aspect of leaf leachates and found that the loss of elements through leaching increased with leaf age culminating with the largest quantities lost as leaves approached senescence. With approaching senescence and subsequent leaf abscission the leaves are prone to leach more elements such as Pb onto the existing leaf litter, providing an above-surface pool of available heavy metals for absorption into the freshly fallen leaves that will form the O_1 litter level. Although differentiation between the Pb content at the moment of leaf abscission and few weeks after incorporation onto the existing litter surface has not been examined, it would be of interest to study, particularly in the light of the higher levels of Pb existing at our MSW site.

Each particular forest stand or landscaped tree association will be comprised of one or all of the classes - excluders, index plants and accumulators (Peterson 1983) - depending on the species composition. Therefore, levels of soil contamination will be reflected in leaf analysis differently depending on the percent species composition of the trees overlying that area. On abscission, leaves are effected by both a flutter effect and wind drift. The relative weight, shape, texture and surface area of the leaf - essentially the aerodynamic design of the leaf - has ramifications on its descent or flutter path. Leaves are also subject to secondary movement on the earth's surface due to wind movement. Leaf dissemination, especially from trees located on or near a point source of contamination, has the effect of dispersing the contaminant over a larger area. This may also result in a further diluting effect as leaves of accumulator species may mix with those of excluder or index plant. This factor may also mask or hide potential contaminant 'hot spots' or present false 'hot spots' due to leaf drift and accumulation patterns. The trace element uptake response by the plants along with leaf drift negate mapping possibilities that may be evident with soil or tree radii contamination.

The localized contaminant 'hot spots' found through dendro-chemical analysis of the trees (Reeves 1993, chapter 1) failed to correlate with leaf contaminant concentration possibily due to leaf drift and dilution effect of leaf spread. This lack of visual correlation in contaminated areas leads to the conclusion that leaf contaminant concentrations are of little value in contaminant tracking. This does not infer that concentrations of contaminated leaves may not be of value within the sphere of management of parks situated on or near contaminated sites.

Conclusion

The only element significantly elevated in leaf litter of ash and other species at the MSW site was Pb. This Pb level, being 11.6 times the Morgan Arboretum control value, deserves management consideration within the Parc régional de la Pointe-aux-Prairies and should be further investigated to determine the full extent of contaminant spread (Pb) and mitigation procedures to be implimented. As the dump site has been officially closed for almost two decades this elevated Pb level is indicative of a persistent Pb contaminant problem.

A lack of a strong correlation between the wood and leaf elemental concentrations essentially rules out the possible use of leaf analysis as an indicator of the level of contaminant loading of the woody plant tissue.

Although trees do not exhibit a strong propensity to cycle heavy metals, they do provide a proven attributes for leachate mitigation. Any vegetation - as required by Québec environmental regulations (Regulation respecting solid waste Q-2 r.14 sect. 45) - planted on a landfill cap, in addition to providing a measure of erosion control, will reduce water infiltration into the landfill cap through evapotranspiration, and in turn, reduce the relative quantities of leachate generated. On areas peripheral to the landfill, trees through evapotranspiration tend to lower the ambient water availability in the soil. Consequently, it lowers the water table hindering the rapid upward movement of leachate through the saturated soil. As moisture levels are reduced, the leachate movement in the soil is transposed from that of a mass flow to a more restricted transport by capillary action. This moisture draw down by the trees lengthens the residency time that heavy metals spend in the lower soil profile allowing a longer time period for the natural attenuation processes of the soil to function.

Management by selection of specific tree species, either through a reactive or a proactive scheme, is useful in mitigating contaminant uptake. Indigenous or introduced tree species that exclude heavy metals uptake should be used in land restoration projects adjacent to closed landfills as they tend to retain relatively higher proportions of the heavy metals in their root systems where they are less prone to come into human contact.

Wind patterns and the resulting leaf accumulations are also worthy of note when design criteria are being considered for parks in areas of potential contamination. As such, leaf accumulations, by design, may be removed and dealt with in a safe manner, eliminating potential problems with contact with the park users.

Thesis Conclusion

Using a closed garbage dump - toxic waste site as a known point source of soil contamination, research was conducted in order to explore the use of dendro-chemical analysis of compartmentalized tree radii as a monitoring tool to determine areal spread, time period of initial contaminant contact and bioaccumulation of the contaminants. Elemental concentrations bound within the xylem wood tissue during periods of suppressed tree growth resulting from environmentally induced stress failed to accurately reflect the ambient soil elemental concentrations, as such an index was developed to overcome this problem.

Using this calculated index it was found that concentrations of Sn, Zn and Cu were elevated within the xylem tissue of the ash trees (*Fraxinus sp.*) at this site. Tin (Sn) was the only element to show a definite marking of initial contaminant contact, as well as demonstrating elemental accumulation with age. Initial contaminant contact marking was not evident with either Cu or Zn, although significant levels of Cu accumulated within the heartwood of the ash while Zn revealed neither a definite nor consistent accumulation pattern with age.

An attenuation zone usually found in association with above-surface MSW landfills failed to materialize at this site; accumulation of elements occurred at a point distant from the point source of contamination. This accumulation of elements occurred in association with a permanent swamp. It is felt that the contaminants were vectored into this area by groundwater flow.

An analysis of the current year leaf litter indicates a persistent problem with Pb contamination at this site, especially considering it is 20 years since cessation of landfill operations. Lead (Pb) cycled to the soil surface via ash tree uptake and natural leaf abscission was 11.6 times that of the control. Although the concentrations of Pb in the leaves was significantly elevated, it was not sufficiently elevated in the xylem tissue to render it significantly different from the control. The reason for this, at present, is unknown.

A series of questions arose from this thesis research and may stimulate further research:

What is the extent and what are the bioeffects of accumulations of Sn within the terrestrial environment ?

Of the indigenous tree species found within Canada, and with specific orientation to Québec, which are excluder, index or accumulator plants in reference to the uptake and binding of trace and heavy metals, and secondly which tree best reflects these individual properties for each element?

With leachate migration being an acknowledged problem associated with MSW landfills, which tree species are most effectively planted on the periphery of landfills so as to be able to withstand the ingress of the leachate into the rhizoshpere?

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APPENDIX

The major elemental groupings follow those found in the research presentation of Kabata-Pendais and Pendais (1984). The information on each of the elements has been extrapolated and conceptualized from the numerous references used in preparing the thesis. As such, the following information is rendered in a form that is as concise as possible; the sources, other than specifically cited in the synopsis, are contained in the bibliography.

Elemental Characterization

The research in the preceding thesis focuses only on the inorganic trace and heavy metals Cd, Pb, Zn, Cu, Mn, Ni, Cr, and Sn as found in MSW leachate. As a rule of thumb, concentrations of these metals taken up and bound in the xylem wood are around 1% of those in the soil sink. The effects of these elements are specific to element, species, organ, and often enzyme. With many of the essential trace elements, the difference between deficiency and excess (luxury consumption) is a very narrow margin : likewise, an equally narrow range exists between suppressed, normal growth and death. The accumulating non-essential trace elements function on a continuum between normal growth and death of the plant.

Generally these trace elements can be classified into three broad groups based on common response characteristics: group I) Cd, Pb; group II) Cu, Zn, Mn, Ni; group III) Cr, Sn.

Group I

The elements of this group, Cd and Pb, are of most concern to man as they are both cumulative toxins in the mammalian system. Only Pb has an established Biological Exposure Index. The medical community intends to establish a similar index for Cd (Hall 1989).

Cadmium (Cd)

Non-essential and toxic to both plants and man, it is known to concentrate in the liver and kidneys of man Most sources of Cd appear to be anthropogenic in origin arising within the past 40 years Natural concentrations are often related to argillite and shale sediments Within soils it tends to be occluded in the organic matter, with mobility changing from high in acid soils to moderate in alkaline soils. Cadmium is considered toxic in soil concentrations ranging from 3 - 8 ppm. Cadmium takes on a contradictory role in relation to and interaction with other elements It is in a constant state of flux between enhancing, reducing and competing with other elements Within plants the primary area of concentration is in newer roots, often causing root dysfunction, internal water deficit, reduced conductivity and transpiration, and interfering with enzyme function.

Lead (Pb)

Non-essential and toxic to both plants and man, although naturally occurring Pb appears to be non-toxic to plants (Smith 1973). Lead is readily assimilated in both the bone and soft tissue of mammals generally gaining entry by way of ingestion. In soils it tends to accumulate in organic layers and has low mobility in both acid and alkaline conditions. Lead is considered toxic in soil concentrations of around 100 ppm with its toxicity generally decreasing with increasing alkalinity. It forms synergistic relationships with Cd and possibly antagonistic reactions with Zn within the plants. The main concentration area in plants is the newer roots although movement takes place as age breaks down the binding mechanisms. Lead causes general stress especially evidenced in stomatal opening and interferes with several enzymes.

<u>Group II</u>

The members of this group are generally essential but non-cumulative elements in man.

Copper (Cu)

Essential to both plants and man and toxic to plants especially in the cuprous form at pH < 7.0 Mobility is low in both acidic and alkaline soils. Availability is intimately related to soil pH with Cu being strongly adsorbed onto silicate minerals and chelated to organic matter Copper is considered toxic to plants in soil concentrations that range from 60 - 100 ppm. Copper has an antagonistic relationship with Zn within the plant, competing with and inhibiting it. In relation to Cd, Cu is competitive, often to the extent that Cu uptake is inhibited. A possible synergistic relationship between Cu and Cr exists. Copper toxicity is often mitigated with the presence of Zn and Mn in the soil. Toxicity within plants is by way of interference, as it promotes oxidation of ferrous to feric ions thus obstructing Fe translocation.

Zinc (Zn)

Essential but non-toxic for both plants and man. In soils Zn is strongly adsorbed by silicate minerals and to a lesser extent, chelated on organic matter. Mobility is classified as extremely mobile on acid soils and moderate on alkaline soils. Zinc initially tends to concentrate in the root system until luxury levels are obtained. At this point it is highly mobile within the plant with uptake reflecting in a linear manner the soil concentrations. Zinc is considered toxic in soil concentrations that range from 70 - 400 ppm. Zinc is competitive with and often acts catalytically with Cd. Cadmium influences Zn root to shoot redistribution and enhances Zn toxicity. In excessive levels, Zn interferes with photosynthesis, phloem transport, transpiration and to a lesser extent respiration.

Manganese (Mn)

Essential to both plants and man with toxic reactions observed in plants at pH<5.0. Elevated environmental levels of Mn are of growing medical concern as a result of the switch from Pb to Mn derivatives as anti-knock compounds in gasoline. It is found throughout the soil profile but may accumulate in surface layers as it chelates with organic matter Toxicity for Mn in soil concentration is listed at between 1500 and 3000 ppm Extractable Mn is related to soil moisture, hence Redox potential, with poorly aerated soils leading to increased uptake. It is known to be antagonistic with Fe, interfering with Fe function.

Nickel (Ni)

Non-essential to plants although essential to man, being toxic to plants and marginally toxic to man. Often associated with Cr, it creates problems for plants growing on serpentine soils. Mobility of Ni is high in acid soils and low in alkaline soils with availability inversely related to soil pH Distribution in the soil profile is somewhat nebulous, although possible chelation with organic matter has been suggested. It is considered toxic in soil concentrations of around 100 ppm. Nickel has antagonistic and / or synergistic relationships with both Zn and Cd. It also has an antagonistic relationship with Fe resulting in Fe suppressing Ni uptake. Many plants show less tolerance to Ni than to Pb.

Group III

This group is the least understood with few definite assumptions. These elements vary dramatically depending on chemical speciation.

Chromium (Cr)

Non-essentiai to plants and marginally essential to man. Depending on the species of plant, Cr demonstrates various degrees of toxicity. Hexavalent Cr (Cr^{6+}) is highly toxic and carcinogenic to man, with other forms being relatively non-toxic. Mobility is moderate in acid conditions dropping to low in alkaline soils. It is considered toxic in soil concentrations that range from 75 to 100 ppm. A possible synergistic relationship exists with Cu. Luxury levels interfere with the normal functioning of glucose and cholesterol metabolism.

Tin (Sn)

Non-essential to plants and marginally essential to man. Toxicity is reported varying from highly toxic to higher plants and fungi, to undefined. In the organo-tin form it is highly toxic to man, while inorganic forms of tin appear relatively non-toxic It is listed as fairly mobile in the soil in conditions of near neutral pH Tin toxicity in some plants occurs when soil concentrations of 60 ppm are surpassed Although readily taken up by the plants, it appears to be sequestered in the roots with little movement. Interaction with other elements is undefined.