

An overview of trace metals in the environment, from mobilization to remediation

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Introduction

This special issue of *Environmental Geology* arose from a symposium on *Trace Metals in the Environment* held at the Geological Association of Canada – Mineralogical Association of Canada (GAC-MAC) Joint Annual Meeting in Winnipeg, Manitoba in 1996. Most of the papers in this special issue are based on presentations made at the symposium. The ultimate goal of the symposium was to bring together workers doing research in diverse fields, all involving metal speciation and transport. More than any other area of scientific endeavor, environmental research demands a multidisciplinary approach. We have much to learn from each other, and together we can move toward a better understanding of metal cycling in the environment.

An environmental model

Our environment, the place where we live, is the planet Earth. The Earth is made up of three reservoirs: the solid Earth or *geosphere*, the liquid layer or *hydrosphere*, and the gaseous envelope or *atmosphere* (Fig. 1). Living organisms (the *biosphere*) inhabit the Earth environment, interacting with its component parts. There is a continuous exchange of matter and energy among the geosphere, hydrosphere and atmosphere. The nature of these reservoirs and the processes involved in transferring energy and matter between them combine to create an environment that is capable of sustaining life in its many forms, a situation unique in our solar system. Metals in the environment may be present in the solid, liquid or gaseous state. They may be present as individual elements, and as organic and inorganic compounds. The movement of metals between environmental reservoirs may or may not involve changes of state.

The geosphere is the original source of all metals (except those that enter the atmosphere from space in the form of meteorites and cosmic dust). Within the geosphere, metals may be present in minerals, glasses, and melts. In the hydrosphere, metals occur as dissolved ions and complexes, colloids, and suspended solids. In the atmosphere, metals may be present as gaseous elements and compounds and as particulates and aerosols. Gaseous and particulate metals may be inhaled and solid and liquid (aqueous-phase) metals may be ingested or absorbed, thereby entering the biosphere. In addition to being the original source of all terrestrial metals, the geosphere may represent a sink for metals. The atmosphere and hydrosphere also constitute sinks for metals; however, from a geological perspective, they are more likely to be considered agents of transport. Whether we define an environmental reservoir as a sink or as a medium for the movement of metals from one site to another depends on the linear and temporal scales of observation. For example, the oceans are a vast reservoir for a variety of chemical elements. However, they also are a conduit for elements derived from weathering of rocks to return to the geosphere through sedimentation. Finally, a reservoir may act as a catalyst for changes of state of metals and metal compounds, without actually having incorporated those metals, as in the case of some biologically mediated reactions.

Figure 1 shows a comprehensive model for metal transport. It takes into account all reservoirs, whether they represent sources, sinks, or agents of transport. This model illustrates the “natural” interactions between environmental reservoirs. Figure 2 shows how the segment of the biosphere occupied by humans interacts with the environment. Figure 2 includes mechanisms and processes similar to those illustrated in Fig. 1. For example, dumping of wastes into lakes and rivers and production of emissions from smelting of ores may be viewed as a form of excretion by society as a whole rather than by an individual cell or organism. Most efforts to remediate metal contamination involve fixing metals in a solid form and returning them to their original source – the geosphere (Fig. 2).

The presentations in the symposium at GAC-MAC and the papers in this special issue illustrate different parts of the model. Some workers focussed on characterization of materials within individual reservoirs, whereas others documented experiments designed to understand mecha-

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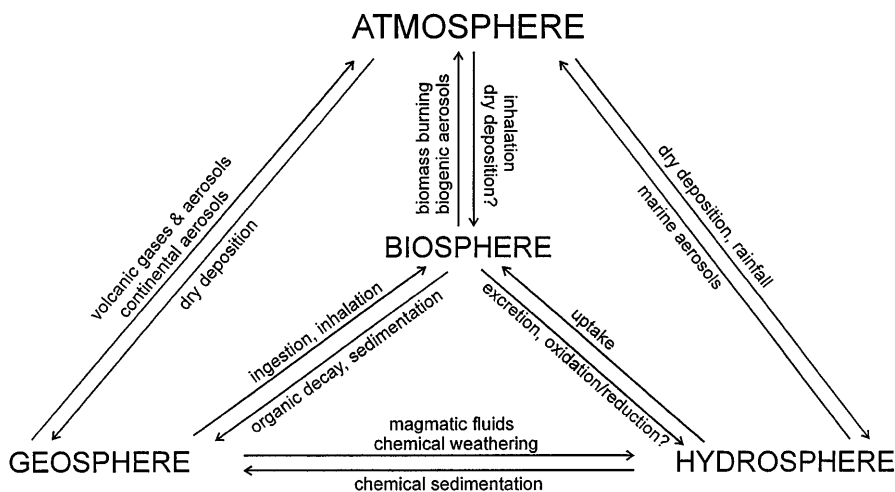


Fig. 1

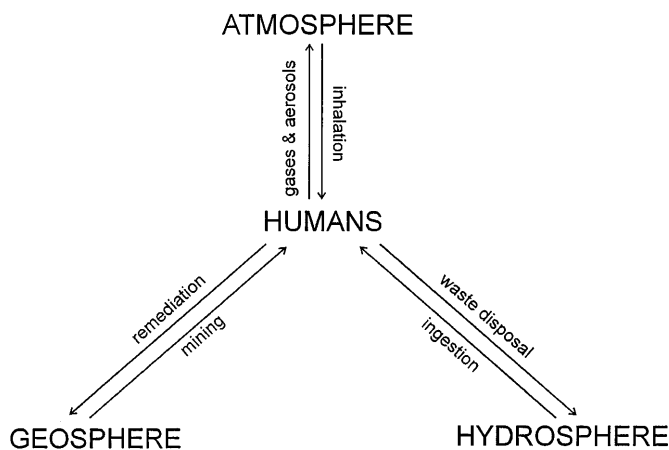


Fig. 2

nisms of exchange between environmental reservoirs. This overview is not intended to be comprehensive, but rather to identify some areas of current research, and to place the papers in this issue in a wider context.

Geosphere – hydrosphere interactions

One area of considerable environmental concern is the weathering of mine tailings and generation of acid mine-drainage. Tailings environments are the site of metal flux from the geosphere to the hydrosphere through dissolution of minerals, and vice versa, through precipitation of minerals and adsorption and exchange of metals (Fig. 1). The presentations on this topic delivered the clear message that decisions regarding disposal and reclamation require an understanding of metal behaviour specific to the local conditions – including climate, mineralogy, geology, geochemistry, and topography. Shaw and others (1998) described laboratory and field-lysimeter experiments to determine optimal dry-cover and dam construction materials. Their study of oxidation behaviour of Fe-sulphide minerals in tailings showed a positive correlation between the sulphur content of tailings and their acid-generating capacity. McGregor and others (1996) reported on the mobilization and attenuation of heavy me-

tals in the Copper Cliff Ni-mine tailings near Sudbury, Ontario. Oxidation and dissolution of sulphide minerals release Fe, Ni, and Co to pore water in the shallow tailings. Subsequently, pH-buffering reactions result in precipitation of these metals in secondary-phase minerals (Jambor 1994). The authors reported that goethite, jarosite, and vermiculite are sinks for Ni. Chemical-extraction experiments demonstrated that significantly higher concentrations of metals are removed in reducing experiments than in acid-leaching experiments. The implication is that some remediation measures involving acid-buffering may inadvertently destabilize secondary minerals which host heavy metals.

In contrast to temperate regions, areas of low precipitation undergo arid weathering. Chávez and others (1996) documented mineral-weathering reactions under arid conditions in the southwest USA, where tailings are exposed to alternating periods of oxidation (during dry seasons, spring and fall) and leaching (during snow melt and rainy seasons). Their examination of paragenetic relationships of sulphides and oxides in mine tailings indicated that some sites are not actively releasing metals, and thus require different remediation techniques than those that generate acidic drainage. *Careful mineralogical characterization is a necessary step in the evaluation of tailings*, as their current metal content may have been influenced by grain-size variation, addition of lime, and variations in residual-metal concentration resulting from differing extraction efficiencies. The importance of these factors has been documented by Boulet and Larocque (1998) in a comparative study of sulphide mine tailings at two sites in New Mexico. The young and relatively unaltered Cyprus-Pinos Altos tailings show variations in metal concentrations that reflect the above factors. In contrast, current metal distributions in the extensively weathered Cleveland mine tailings are a function of the distribution and stability of secondary phases. For example, acidic drainage from the Cleveland tailings has much lower concentrations of Pb than other metals because secondary Pb-sulphate is highly insoluble in tailings environments.

Percival and others (1996) reported that total dissolved As concentrations in surface waters draining mine waste near Cobalt, Ontario vary from 11 to 20000 $\mu\text{g/l}$, with strong seasonal variation. Arsenate species predominated in strongly oxidizing surface waters, whereas arsenite species were observed in relatively reducing groundwater. Strong seasonal variations in surface-water concentrations are related to the formation of secondary efflorescent arsenate minerals. During dry periods, mineral crusts containing secondary arsenates form; during wet periods, these efflorescent minerals are readily dissolved and flushed into surface run-off.

Kwong (1996) documented natural acidification of a lake in Central Yukon. Clear Lake is adjacent to a buried massive sulphide body, and receives metalliferous, acidic, subsurface drainage. Due to the presence of carbonate rocks flanking the lake, neutralization occurs within 1 km downstream. Elevated metal concentrations and authigenic sulphides are found in the reducing environments of bogs and lake-bottom sediments. In contrast to the acidic conditions in the Clear Lake area, surface waters of the Keno Hill Mining District, Central Yukon, are neutral to slightly alkaline (Roots and Kwong 1996). The climate is cold and semi-arid. Here, a large volume of unvegetated rock waste has accumulated over 75 years of mining, and has caused elevated metal concentrations in nearby streams. Metal transport in these streams is limited due to cryogenic precipitation, and coprecipitation and sorption onto suspended particulate matter and the stream substrate. Acid drainage is not widespread in this area due to the slow rate of sulphide oxidation and continuous neutralization by local carbonate minerals.

Geosphere – atmosphere interactions

Continental aerosols and volcanic gases, aerosols, and particulates carry metals from the geosphere to the atmosphere. Metals may be returned to the geosphere through dry deposition (Fig. 1). Rasmussen (1998) discussed the current debate over the relative significance of natural and industrial emissions of metals to the atmosphere on the global scale. Geoscience perspectives are needed to address the large uncertainties and information gaps associated with attempts to distinguish airborne metal species arising from industrial sources from other species of metals in the atmosphere. These include volcanic particles and gases (Stimac 1996), windblown soil and sediment particles, forest-fire debris, biogenic particles (waxes, pollen, spores), seaspray, low-temperature degassing, and methylation products. Existing estimates of the annual global flux of metals from natural sources vary by orders of magnitude. Thus, conclusions about the relative significance of industrial emissions can depend entirely on which natural flux estimate is chosen. For example, one emissions inventory for Pb indicated that only 4% of the global Pb flux arises from natural sources (Nriagu and Pacyna 1988), whereas another indicated that at least 90% arises from natural sources (Kownacka and others 1990). This disparity is due entirely to differences in the

estimation of the natural Pb flux, as the estimates of anthropogenic emissions in the two inventories are virtually identical.

Stimac and others (1996a, b) presented evidence of the significance of natural volcanic emissions by documenting the redistribution of Pb and other volatile metals during eruption and devitrification of the Bandelier Tuff, New Mexico. Lead and other incompatible trace metals become strongly enriched in silicic magmas. During eruption, they are partitioned into the vapor phase, and thus may be released directly into the atmosphere in large amounts. Gaseous metals and metal compounds also may be trapped in gas bubbles in vesiculating magma. During emplacement and cooling of tuffs, metal-rich phases crystallize in vesicles. In the Bandelier Tuff, the mineral suite is rich in sulfides, oxides, and chlorides of Fe, Pb, Bi, Cu, Ag and Re and silicates of incompatible elements (Stimac and others 1996a, b). In pumice samples from Volcán Popocatepetl, Mexico, vapor-phase Ni-rich magnetite, barite, sulfides of Fe, Cu, Zn, and Ag, and Cu-Au tellurides have been documented (Larocque and others 1998). Minerals also may crystallize from gases liberated during devitrification of matrix glass (Stimac and others 1996a, b). The fine-grained nature of minerals crystallized from a vapor phase and their location in porous tuffs makes them susceptible to mobilization by groundwater.

Stimac (1996) compiled available data on metal flux to the atmosphere resulting from various types of volcanic-emission processes; these include explosive and quiescent eruptions, passive degassing, and fluid discharges (springs and seafloor hydrothermal vents). He pointed out the large uncertainties associated with these data, and the necessity of additional research to better constrain volcanic metal fluxes. Current techniques of estimating metal fluxes involve normalizing metal concentrations in condensed gases or particulates to concentrations of S, then multiplying by the flux of SO_2 . Nriagu (1989) assumed metal/S ratios and used the measured S flux to calculate metal fluxes. This approach is not viable, as metals commonly are complexed with Cl, and there is no geochemical coherence between S and metals.

Henderson and others (1996) studied the distribution of trace metals in humus and till samples in the area of Flin Flon and Snow Lake, Manitoba, to determine the relative contribution of metals from natural and anthropogenic sources and the fate of metals associated with fall-out from a smelter. Surface humus and underlying glacial sediments were collected in a regional-scale survey and in detailed surveys near the smelter at Flin Flon. Total-metal concentrations near the smelter were enriched by as much as 250 times background. Metal concentrations decreased exponentially with distance, reaching background levels at distances averaging 40 km from the smelter. Sequential-extraction techniques indicated that metal-partitioning behaviour is specific to the element. Smelter-derived particulates were identified using SEM, and consisted of distinctive spherical particles, metal particulates, oxides and sulphates. Smelter-derived spherules were

present only in humus and not in till, and were rare outside a radius of 100 km from the smelter.

To address the need to characterize natural sources of Pb in the vicinity of the long-range atmospheric monitoring site at Dorset, Ontario, Anglin and others (1996) investigated Pb-isotopic signatures of natural surficial materials (till and lake sediments) and bedrock. The study found that Pb-isotopic signatures previously attributed to long-range atmospheric transport of metal pollution also could be derived from local natural sources (e.g., windblown dust), pointing to the need for improved methods of distinguishing sources of airborne Pb at rural and remote receptor sites.

Geosphere – biosphere interactions

Metals may enter the biosphere through the ingestion of solids (Sheppard 1998), the ingestion or absorption of liquids, and the inhalation of solids (Guthrie 1992) and gases (Fig. 1). Fyfe (1996) provided an overview of the role metals play in health, economics and the environment. Life evolved in the presence of metals in the environment, and thus organisms at all levels are adapted to metals. Indeed, many metals are essential for health. But at the same time as identifying the usefulness of metals in everyday life, it is necessary to acknowledge mistakes of the past and present, particularly in the face of the increased pressures caused by overpopulation (Fyfe 1997). Geoscientists can make a unique contribution by focussing research efforts on understanding geochemical controls on mobility and bioavailability of metals, and by improving technologies for the remediation of polluted sites.

Condoyannis and others (1996) documented elevated concentrations of Ag, As, Cd, Cu, Fe, Mn, Pb, Sb, and Zn in surface soils and plants on the floodplain of the Coeur D'Alene River, Idaho. The enrichments were as much as 186 times higher than in a control area and resulted from dispersal of mining and milling wastes from a major Ag-Pb-Zn mining district. Those authors observed increased mobility and bioavailability, related to increased pH, under wet conditions. Bioavailability also appeared to be a function of plant species. Plant-soil metal ratios decreased in the order *Potamogeton* (spp.) > *Equisetum palustre* > *Phragmites australis* > *Agrostis stolonifera*. The ability of aquatic *Potamogeton* (spp.) to take up metals otherwise considered immobile suggests that it may be a useful accumulator plant for the removal of metals from contaminated soils.

Aide and Cwick (1998) examined soil-forming processes that influence nutrient status and metal bioavailability in a boreal forest ecosystem near Thompson, Manitoba (as part of NASA's BOREAS project). Selective sequential extraction indicated that nutrients and metals in the cation-exchange fraction and/or sequestered by the organic matter pool are bioavailable fractions, whereas the amorphous and crystalline Mn- and Fe-oxide fractions buffer the more active pools against changing soil conditions. Important soil-forming processes include: decalcification of the surface horizon; eluviation-illuviation; pedoturba-

tion; melanization and humification; limited oxide formation (confined to the upper tier of soil); biocycling (e.g., sphagnum decomposition); acidification (in particular, leaching of organic acids from Jack Pine); and mobilization of nutrients that support vegetation.

Geophagy refers to the purposeful or inadvertent ingestion of soil by animals or humans, and may constitute a significant pathway for metal ingestion. However, quantification of metal ingestion is difficult as it is necessary to measure the amount of soil ingested, the total metal concentration of the soil particles ingested, and the bioavailability of the soil-bound metals in the gut. Geophagous adults may consume as much as 50 g/day of soil. Accidental ingestion by adults is on the order of 60 mg/day. Children, who are more susceptible to the potential toxic effects of metal consumption, also tend to ingest more soil. Experiments indicated that, relative to total soil concentrations, metals are enriched in the fine soil particles that adhere to human hands because it is the metal-rich clay particles that cling to skin. Results of experiments on animals indicated that the bioavailability of Cs increases fourfold when Cs-spiked feed is ingested without added soil than when ingested with added soil, as the soil components provide a competitive sink for Cs. In contrast, there is no difference in bioavailability of Pb from feed with or without the addition of soil (Sheppard 1998).

Geosphere – hydrosphere – biosphere interactions

Interactions between environmental reservoirs are, in some cases, mediated by biological activity. In these cases, metals do not actually enter the biosphere; however, the presence of organisms catalyses other reactions. Donahoe and Chongxuan Liu (1998) reported that Fe-oxhydroxide adsorption/desorption processes have greater influence than sediment organic matter on the mobility of trace metals in a biologically zoned, riverine freshwater wetland in central Alabama. There, soil-weathering reactions are the predominant source of trace metals. Aluminum cycling is a function of weathering and clay-mineral formation. In contrast, cycling of Pb, As, B, Cd, Sr, and Ba involves their adsorption onto surfaces of Fe-oxhydroxide phases, with subsequent release of the metals due to bacterially-mediated reductive dissolution of ferrihydrite. Both Fe-reducing and sulphate-reducing bacteria are important. Released metals may move upward due to diffusion or downward with the hydraulic gradient.

Rasmussen and others (1998) studied trace metal concentrations in lake sediments on the Canadian Shield near Huntsville, Ontario. They documented large variations in Hg concentrations between lakes, and strong correlations between Hg content of lake sediments and smallmouth bass tissue. Organic matter was important in the release, transport and redistribution of metals including Hg, and shapes of vertical concentration profiles were influenced mainly by diagenetic processes. The extent to which anthropogenic processes may have affected the lake environments is difficult to assess; however, geological distri-

bution of metals and natural processes have played an important role in the current distribution of Hg and other metals.

Hydrosphere – biosphere interactions

Metals derived from human pollution enter the hydrosphere (Fig. 2) and may eventually return to reside in other hosts in the biosphere. In companion posters, MacFarlane and others (1996) and Shwetz and others (1996) presented trace-element distribution and speciation data for waters, bottom sediments, and *Dreissena polymorpha* (zebra mussels) from the heavily industrialized St. Clair River, Lake St. Clair and western Lake Erie. Total trace-metal concentrations indicated point sources of metal loading in the Lake Huron-Lake Erie corridor. Sequential extractions indicated that Cu is associated with the organic fraction, and all other metals are associated with the reducible fraction. Al-Aasm and others (1998) examined the stable-isotope composition of, and trace-metal distribution in, sediments and zebra mussels from western Lake Erie. Zebra mussels incorporate trace elements into their shells, and thus may be useful biomonitors of metal pollution. The metal concentrations in the shells increased in growth increments corresponding to warmer water temperatures (as determined from stable-isotope signatures).

Atmosphere – hydrosphere – biosphere

In the hydrosphere, the fate of metals derived from atmospheric deposition commonly may be determined by biological activity, as in the case of methylation of Hg (Matilainen 1995). In their study of boreal forest catchments containing different types of wetlands, Kelly and others (1996) reported that all catchments were sinks for total Hg. Upland areas retained methyl Hg, whereas wetlands were net sources of methyl Hg. Variations in internal hydrology of wetlands and yearly differences in water yield resulted in differences in source strength for methyl Hg among wetland types. Thus, modelling of methyl-Hg production in any catchment requires a knowledge of the percentage and type of wetland area and annual water yield.

Kettles and Bell (1996) observed significant vertical variations in Pb-isotopic ratios in peat core samples collected from a bog near Detour Lake, northeastern Ontario. The $^{206}\text{Pb}/^{204}\text{Pb}$ ratios were highest in the peat which accumulated during the Hypsithermal, an episode of post-glacial warming. The authors concluded that Pb-isotopic fingerprinting of peat, in combination with an understanding of the history and context of bog development and careful attention to related parameters (e.g., bulk density, total Pb abundance, loss-on-ignition), has considerable potential for discriminating between metal sources in the landscape.

Characterization of individual reservoirs: techniques and observations

Accurate characterization of metal-residence sites in individual environmental reservoirs may be necessary before

it is possible to understand the flux of metals between spheres. Plouffe (1998) reported on Hg research in central British Columbia, where cinnabar occurrences in bedrock are responsible for elevated Hg concentrations in surficial sediments. He emphasized that criteria must be established for recognizing Hg derived from natural sources as a prerequisite for effective remedial action. A combination of criteria such as mineral form, regional geochemical patterns, metal associations and depth of sample collection were used to assign a natural origin for the observed metal distributions.

Klassen (1998) discussed the overall significance of regional and vertical variations in metal concentrations in glacial deposits of Canada. In many places, natural concentrations exceed safe-limit criteria established in some jurisdictions. The author related the observed compositional variations to variations in provenance, based on determinations of ice-flow directions and the history of glacial erosion and transport. In central Newfoundland, As concentrations in till range from 10 ppm to over 1000 ppm at the regional scale, depending on the bedrock source. Significant variations of metal concentrations also may occur as a function of depth at a given location. For example, in a sequence of glacial deposits overlying massive-sulphide mineralization at Buchans, the lowermost till contains >1000 ppm Pb and Zn, whereas metal concentrations in the overlying sediments are an order of magnitude lower. Klassen (1998) concluded that a knowledge of natural metal concentrations in soils and soil-parent material is a prerequisite for evaluating the impact of human activity, and for establishing appropriate clean-up criteria.

Characterization requires the use, and sometimes development, of appropriate analytical techniques. Hall and others (1996) described the application of selective-leach procedures to studies of base-metal dispersion around several mines in Manitoba, and to the identification of surface geochemical signatures at sites of underground nuclear explosions. The authors noted that while phase-selective leaches have been used for some time, the sensitivity of ICP-MS now allows geochemists to take full advantage of the former techniques for environmental research. Analysis of some elements remain problematic: Hg, for example, tends to be readsorbed back onto the substrate during leaching. To minimize this effect, the authors recommended changing from a batch to a flow-through mode of operation.

Conclusions

Much work remains to be done both in characterizing metal-residence sites in individual reservoirs and in understanding processes and pathways responsible for the movement of metals between environmental reservoirs. Toward this end, a second symposium on Trace Metals in the Environment has been organized for the GAC-MAC meeting in 1998 in Quebec City, Canada. The sym-

posium will focus on early diagenetic processes in modern sediments, their effect on vertical metal concentration profiles, and their influence on the reconstruction of anthropogenic input histories.

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