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Heavy Metal Transport and Fate in the Environmental Compartments

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ABSTRACT

This review article will address transports, fates, and behaviors of heavy metals in different environmental compartments (air-water-soil-biota), especially their processes of sinks and remobilization between water and soil, due to addressing as the importantly environmental parts for entering, immobilizing, and remobilizing of heavy metals. The interaction of air-water-soil-biota would tell about the dynamic environment. Heavy metals can be changed upon their characteristics and the properties of the environment where they are concerned. Some of heavy metals i.e. Hg, Cd and Pb would be described for their fates and behaviors, as well as effects not only in human but also in ecosystem. Moreover, heavy metals' consideration as the processes of sinks and remobilization would represent for clearer understanding of behaviors when they are adsorbed onto organic matters or sediments and released to the water. Lastly, these understandings would help to plan and predict the transportation, contamination, adsorption and accumulation of interested chemicals in the environment.

Keywords: Heavy metal, Transport, Fate, Environmental compartments

INTRODUCTION

Generally, the environment is always dynamic. When some physical changes or incidents created from the nature or human have been occurred on the earth, they would influence the changes of atmosphere, watershed, land and living thing. For example, industrial areas have been polluting various kinds of chemicals including heavy metals; they may pollute the atmosphere, watershed and land. These chemicals will contaminate, circulate, and then accumulate in various environmental compartments (air-water-soil-biota), probably called contaminant pathways. They are, additionally, represented as atmosphere-hydrosphere-lithosphere-biosphere interconnected compartments (Walker *et al.*, 2006).

Presently, various polluted chemicals especially heavy metals have been mostly created by human activities of both point and non-point sources e.g. industry, agriculture, transportation, community (Walker *et al.*, 2006). They have been radically and rapidly contaminated and accumulated, as well as dispersed and moved among the compartments. Consequently, it is necessary to study their transports accompanying with fates and behaviors in air-water-soil-biota; including especially to investigate environmentally important heavy metals i.e. mercury (Hg), cadmium

(Cd) and lead (Pb), and heavy metals' processes of sinks and remobilization between soil and water compartments. These ideas would be useful and fruitful for further risk assessment by detecting ambient environmental qualities, analyzing and recognizing problems, monitoring and evaluating occurred problems, as well as determining and ensuring command and control procedures (Cheevaporn, 2004).

INTERACTION OF AIR-WATER-SOIL-BIOTA

For making clearer pictures in the chemicals' transport, fate and behavior, it is obviously important to understand the relationship of air-water-soil-biota, which is called compartments' interaction.

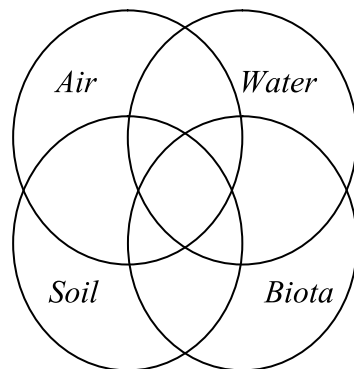


Figure 1 Compartments' interaction (Gulliver, 2007)

Figure 1 shows the relationship of air-water-soil-biota, contact areas among each other as subset. These components have their own properties i.e. air (e.g. temperature, wind speed, light intensity, etc.), water (e.g. depth, flow rate, temperature, pH, etc.), soil (e.g. organic, inorganic, etc.) and biota (e.g. flora, fauna, size, etc.). Then, it is crucial to understand in each area, expressing different characteristics. These vastly pathways will bring any substances including chemicals into the dynamic environment or ecosystem. Since a given chemical that has its own physicochemical properties e.g. concentration, polarity (water solubility), vapor pressure (volatilization), Henry's law constant (H) (gas solubility), octanol-water partition coefficient (K_{ow}) (hydrophobicity index), organic carbon normalized distribution coefficient (K_{oc}) (organic soil sorbent capability), sorption, diffusivity, etc. (Corsi, 1998) is polluted to different compartment, it will act or behave in different ways. The concentration or characteristic of this chemical could be increased, decreased or remained the same upon its fate. This is because the transportation or movement depends on properties of the chemicals themselves and properties of the environmental compartments (Walker *et al.*, 2006).

Generally, air and water are the main pathways of chemicals' entry and diffusion into the environment. Solid particles would disperse and carry to a far away area, such as the distribution of lead into the North Sea (meaning that it was generated from land, transported by air, and absorbed in water and biota) of England coast that the sources came from a lot of places e.g. industries, hazardous waste disposals, lead-suspended air conditions, etc. (Cheevaporn, 2004). Not only air and

water, but also soil and biota are the contaminant pathways of chemicals. For example, DDT was originally synthesized by Othmar Zeidler, German Scientist, in 1874 and has been used since 1942 for agricultural purposes as pesticide (Edwards, 2002). After post harvesting, this chemical was normally washed by rain from soil to water or carried by wind from soil to air (may be dropped in a far away water area). Additionally, it has been also absorbed and accumulated in biota (flora and fauna on land and in watershed) called bioaccumulation that is increasingly accumulated in each trophic level along food chains as biomagnifications (Boonsaner, 2006).

HEAVY METAL CONFRONTING CONSIDERATION

As mentioned above, anthropogenic activities are the main sources of pollutants or chemicals. Especially, mining of metal ores has increased the occurrence of heavy metal contamination at the Earth's surface. Opencast mining activities, specifically, have a serious environmental impact in air, water and soil, even in biota (Rodriguez *et al.*, 2008). Heavy metals are released naturally to soils and surface waters by physical and chemical weathering, and volcanic explosion, but they are rapidly elevated their inputs from anthropogenic sources e.g. industry, agriculture, fossil fuel combustion, and waste disposal; for example, cadmium (Cd) input by fertilizers increases its concentrations in agricultural soils and is associated with increasing cadmium levels in crops (Jones *et al.*, 1992).

Table 1 Anthropogenic enrichment factors (AEF) for total global annual emissions of mercury, cadmium, lead, zinc, and manganese in the 1980s (all values 10⁶ kg/year)

Metal	Anthropogenic sources (A)	Natural sources	Total (T)	AEF (A/T) (100%)
Mercury (Hg)	100	50	150	66
Cadmium (Cd)	8	1	9	89
Lead (Pb)	300	10	310	97
Zinc (Zn)	130	50	180	72
Manganese (Mn)	40	300	340	12

(Source: Walker *et al.*, 2006)

The extent to which anthropogenic activities contribute to global cycle of metals can be described by the anthropogenic enrichment factor (AEF), which is the proportional percentage of metal emissions between anthropogenic sources and natural sources, as shown in table 1. It is clear that anthropogenic activities are responsible for the majority of the global movement of mercury (Hg), cadmium (Cd), lead (Pb), and zinc (Zn) but are relatively unimportant in the cycling of manganese (Mn).

After entering the environment, the heavy metals are transported and may be transformed into other chemicals. Transport can occur within a compartment or between compartments. Transformation processes in the environment involve photo-degradation (e.g. ultraviolet) or chemical degradation (e.g. hydrolysis) or biodegradation (e.g. bacterial decomposition). Heavy metals may also be transformed within organisms and this is called biotransformation (Boonsaner,

2006). The examples of heavy metals conducted are mercury, cadmium, and lead. They will be considered and described as the examples of environmentally important and influential elements as follows:

MERCURY (Hg)

Extensionally known for the Minamata disease, it was caused by the release of methyl mercury (CH_3Hg^+) from the industrial wastewater of the Chisso Corporation's chemical factory into Minamata Bay and Shiranui Sea. This highly toxic heavy metal was accumulated in shellfish and fish, eaten by the local people, living in these areas. Consequently, living organisms i.e. cat, dog, pig, and human deaths continued over 30 years (Ministry of Environment, 2002). Mercury exists in the inorganic form as free mercury (Hg^0), mercury ion in salts and complexes (Hg^{2+}); or as organic mercury compounds, such as phenyl mercuric salts, which have been used as fungicides and herbicides, and alkyl-mercury compounds including methyl mercury, produced by humans, sediment anaerobic microorganisms, and possibly by fish (Boonsaner, 2006).

Mercury is transported to ecosystem via surface runoff and through the atmosphere that it is tightly bound to both organic and inorganic particles. Sediments with high sulfur content will strongly bind mercury. Organic acids such as fulvic and humic acid are usually associated with mercury that is not bound to particles (Fairbrother *et al.*, 2001). Methyl mercury released in surface water can undergo photodecomposition to inorganic mercury; however, it can also be bioaccumulated by plankton, algae, fish, and bird throughout human. For instance, in fish, the rate of absorption of methyl mercury is faster than inorganic mercury; therefore, a net result of methyl mercury is high in the muscle tissue (Kendall *et al.*, 2002).

CADMIUM (Cd)

Cadmium has long been recognized as a toxic element. Its importance as an environmental contaminant was demonstrated in the outbreak of Itai-itai disease caused by smelter wastes that contaminated rice paddies in Fuku, Japan (Ljung, 2001). Cadmium deposits are found as sulfides with zinc, copper, and lead deposits, and cadmium is recovered as a by-product of smelting processes for those metals. A major environmental source of cadmium is vapor emissions accompanying with dust that contaminate surrounding soil and water through fallout during smelting. (Boonsaner, 2006) Natural soil concentrations of cadmium are less than 1 ppm and average about 0.4 ppm. Sewage sludge is often contaminated with cadmium, which then concentrates in plants grown on contaminated soils. Cadmium enters agricultural soils from impeding the usage and disposal of domestic sewage sludge, and as a contaminant of phosphate fertilizers. (Jones *et al.*, 1992)

Cadmium concentrations in fresh waters are usually less than 1 ppb, while in seawater range from 0.05 to 0.2 ppb and average about 0.15 ppb (Forstner and Wittmann, 1983). Higher concentrations of cadmium in surface water are usually due to agricultural plants using fertilizers, plating operations, batteries, plastics manufacture, or from sewage effluent. Furthermore, drinking water in soft-water areas can conserve as a source of cadmium through corrosion of plumbing. (Fairbrother *et al.*, 2001)

LEAD (Pb)

The use of lead, its mining and its production process date back several centuries. Changing usage patterns rather than increased consumption determine present environmental inputs from human use of lead. Batteries, gasoline additives, and paint pigments are major uses, but combustion of lead additive gasoline is the major source of environmental pollution by lead, especially organic lead e.g. tetraethyl lead and tetramethyl lead (Fairbrother *et al.*, 2001). As lead additive size diameter in gasoline is less than 1 μ ; it can transport to far distance more than 30 km. Although these lead additives were presently cancelled, they are still found and detected in dust particles (Boonsaner, 2006). Thus, lead is primarily an atmospheric pollutant enters soils and waters as fallout.

Lead enters aquatic systems from runoff or as fallout of insoluble precipitates and is found in sediments. Typical fresh-water concentrations are between 1 and 10 μ g/l while natural lead concentrations in soil range from 2 to 200 ppm and average 10 to 15 ppm. Deep ocean waters, below 1,000 m, contain lead at 0.02 to 0.04 μ g/kg concentrations; in contrast, surface waters contain from 0.2 to 0.35 μ g/kg (National Academy of Science, 1972). Besides, drinking water concentrations of lead may be greatly increased in soft-water area through corrosion of lead-lined piping and connections. However, average drinking-water intake is considerably less than food sources (Fairbrother *et al.*, 2001). Inorganic lead is normally absorbed in digestion and respiratory systems of organism, while organic lead e.g. alkyl lead is mostly absorbed by skin. But, the toxicity testing has declared that the lead intake by oral is more six-times toxic than absorbed by skin. (Boonsaner, 2006)

HEAVY METAL ASSESSMENT IN THE ENVIRONMENTAL COMPARTMENTS

After considering of the heavy metals, they clearly convey their fates and behaviors in different compartments. Heavy metals, as mentioned above, have originally been derived from geological weathering, and polluted by industrial processing, fossil fuels' burning and waste disposal leaching that they are the main sources of heavy metals' contamination in the environmental compartments. Normally, the properties of heavy metals have a characteristically lustrous appearance, are a good electricity conductor, and generally enter chemical reactions as positive ions (Walker *et al.*, 2006). They are generally less water soluble; the tendency is able to adsorb onto particulate matter or bind with organic groups, called organometallic compounds, thereby forming lipophilic compounds and ions. Therefore, their distributions are found within animals and plants, as well as in sediments.

The fates and behaviors of heavy metals entering the compartments have been changing and transferring among conditions where they are. The greater part of the dissolved heavy metals transported by natural water system is rapidly adsorbed onto particulate matter under their physicochemical properties. However, heavy metals immobilized in bottom sediments do not necessarily stay in that condition, but may be released as a result of chemical changes into water or living organism (Forstner and Wittmann, 1983).

According to Forstner and Wittmann (1983) and Cheevaporn (2004), their fates and behaviors are controlled by processes of sinks and processes of remobilization, addressing as entering compartments. This review article would emphasize on the fates and behaviors of heavy metals when transferring between solid (e.g. soil, sediment, suspended solid) and aqueous (e.g. water, solution) phases. The processes of sinks consist of adsorption and co-precipitation, precipitation, and incorporation in biological activity, and the processes of remobilization comprise elevated salt concentration, change in redox condition, lowering of pH, increasing use of organic complexing agents, and biochemical process. These processes would be described as the followings.

PROCESSES OF SINKS

There are several processes creating metal enrichments in aquatic solid substances as sinks. Forstner and Wittmann (1983) and Cheevaporn (2004) suggested the three groups of heavy metal associations in aquatic solid substances as the follows.

Adsorption and co-precipitation

Heavy metals (Me^+) chemically react as positive ions. The mechanism of their adsorption is based on the sorptive properties of negatively charged particles e.g. silicon hydroxides ($SiOH^-$) and aluminum hydroxides ($AlOH^-$) groups in clay minerals, ferrous hydroxides ($FeOH^-$) and manganese hydroxides ($MnOH^-$) groups in hydrous iron (Fe) and manganese (Mn) oxides, carboxyl and phenolic OH- groups in organic substances. The balancing negative charges is a selective process which accounts for preferential adsorption of specific positive charges and the release of equivalent charges associated with other species. Additionally, all fine-grained materials with a large surface area per volume are capable of accumulating heavy metals ions at solid-liquid interface as a result of intermolecular forces (Forstner and Wittmann, 1983).

Normally, the hydrous aluminum, iron, and manganese, particularly the redox-sensitive Fe- and Mn-hydroxides and oxides under oxidizing conditions, constitutively act as significant sinks of heavy metals in water systems. These hydroxides and oxides e.g. ferric oxides ($Fe_2O_3^-$) and ferric hydroxide ($Fe(OH)_3$), representing as Fe^{3+} , readily sorb or co-precipitate cations and anions. And, under reducing conditions the sorbed heavy metals are readily mobilized; accumulations of hydrous Fe/Mn oxides can, therefore, act as a major source of dissolved metals (Fe^{2+}) in natural waters. The latter mechanism is particularly effective in the presence of higher concentration of dissolved sediments and organic matters (Jenne, 1976).

Precipitation

Every metal has its own equilibrium constant of solubility product (K_{sp}), which is used to describe a saturated solution – relating ionic compounds (soluble salts) of relatively low solubility – in which a dynamic equilibrium exists between solid substance and its aqueous ions (Purdue University, 2008).

If the solubility product of a particular compound is exceeded, precipitation of the compound will occur until the product of the ionic concentrations is not over the K_{sp} value. Therefore, the precipitation will be occurred as a result of exceeding the solubility product that it is considered from the multiplied result of ion products comparing with its K_{sp} value (Ratchamongkol Technology University, 2008). If the multiplied result of ion products exceeds the K_{sp} value it is over the saturated solubility that the precipitation will be occurred.

Incorporation in biological activity

Metals are non-biodegradable and cannot break down into less harmful components. Detoxification by organisms consists of hiding active metals ions within a protein e.g. metallothionein (binding covalently to sulfur) or depositing them in insoluble form in intracellular granules for long-term storage or excretion in the feces. Essential metals e.g. calcium, phosphorus, potassium, sodium, chlorine, sulfur, and etc. are used for ionic balance and as integral parts of amino acids and nucleic acids. Non-essential metals such as mercury, lead, and cadmium, in addition to being toxic above certain or threshold level, may also affect organisms by inducing deficiencies of essential metals through competition at active sites in biologically important molecules (Walker *et al.*, 2006).

These metals involve and in-corporate with biological activities as importantly; for example, zinc is an essential element of at least 150 enzymes (Walker *et al.*, 2006), and iron is a part of hemoglobin, the oxygen-carrying pigment in red blood cells, as well as unimportantly; for instance, mercury and lead are bioaccumulated in living organisms (e.g. diatoms and phytoplanktons).

PROCESSES OF REMOBILIZATION

There are several processes of heavy metals' remobilization from suspended matters and sediments that they are potentially hazardous not only to aquatic living organisms but also to humans and other organisms. The remobilization is mainly caused by the following items occurring from chemical changes in water (Forstner and Wittmann, 1983).

Elevated salt concentration

A desorption of heavy metals from solid materials is occurred due to competing of increased salt contents. Especially in river-estuary boundaries or estuarine environment, the contents of alkali or alkali earth metal groups (representing as cation) e.g. sodium, magnesium, calcium, etc. are enriched. These metals' cations will compete with the adsorbed metal ions onto solid particles or sediments; then, they are released to water. According to Forstner and Wittmann (1983), these adsorbed metal ions would be remobilized by desorption or dissolution.

Change in redox condition

Extensively known, eutrophication caused by an increase in nutritious substances e.g. nitrogen and phosphorus compounds from fertilizers, detergents, and feces, has created oxygen depletion of water courses. Organic substances, mostly of

algae, require oxygen for biodegradation (Walker *et al.*, 2006). Hence, the sediment acts as a sink for oxygen which is supplied through the sediment surfaces at a rate governed by three factors (Mortimer, 1971; Forstner and Wittmann, 1983): 1) a biological oxygen demand respiration and metabolic activities in the sediment; 2) a chemical oxygen demand arising from the fact that inorganic elements such as Fe^{2+} released to the sediment from decomposing biological matter accumulation in reduced form; and 3) diffusion which regulates transportation.

Major components which affect the rate of transported metals are organic matter, iron, and manganese, which are available for redox processes defining as oxidation and reduction reactions. Redox potential (Eh) is expressed as volts and defines the tendency of an environment to receive or supply electrons that the oxidizing environment would have a high Eh value, normally higher than 600 mv (Fitzpatrick *et al.*, 2008). In the reducing environment at a sediment depth of approximately 20 cm the Eh value decreases sharply. At the same time, a strong enrichment of iron and manganese in the sediments can be observed between 15 and 10 cm depth (Forstner and Wittmann, 1983). Within this condition, heavy metals are readily remobilized in through the water compartment.

Lowering of pH

In general, a lowering of pH affects an increase in solubility; thus, in neutral solutions the solubility is increased by orders of magnitude, while pH 4 complete dissolution is largely achieved. For example, iron, manganese, and cadmium sulfides are readily soluble in hydrochloric acid, whereas copper, lead, and mercury sulfides are only soluble in oxidizing acids, the most effective being nitric acid. Moreover, generated hydronium ions (H^+) will compete with metal ions to adsorb with sorptive particles e.g. carbonate; then, the metal ions are released.

Increasing use of organic complexing agents

In natural waters, there are a lot of organic complexing agents, still increased in many rivers due to sewage treatment effluents; these agents can affect the metal remobilization from aquatic sediments. For example, nitrilotriacetic acid (NTA) is a possible alternative to the polyphosphate in detergents. NTA has been applied to the extent of a complexation of heavy metals in aquatic sediments by means of creating high soluble organometallic complexes. Therefore, NTA complexing agent promotes the release of metals from sediments (Gregor, 1972).

Biochemical process

When heavy metals adsorb onto particulate organic matters or sediments, they can be remobilized into water by microbial process as biodegradation; for instance, organic matter is destructed to molecular weight components that are more capable of complexing other metal ions. (Forstner and Wittmann, 1983; Cheevaporn, 2004) Then, the adsorbed metals are released.

DISCUSSION OF HEAVY METAL CONSIDERATION IN ENVIRONMENTAL STUDY

This section would help to consider and analyze the heavy metals in which compartments should be studied due to their fates and behaviors. Since they can, of course, express the tendency to settle or stay upon their physicochemical properties and environmental compartments' properties, the highest amount compartment found should be conducted to research or study. Because, this high accumulated compartment indicates the heavy metal exposure risk of surrounding organisms, even the heavy metal bioaccumulation inside the organisms.

Cheevaporn (2004) and Gulliver (2007) said that the forecast of toxicant transport (including fate and behavior understanding) in the environment can expect what organism would be mostly affected from that considered toxicant. Therefore, the toxicant transport route found in the highest concentration is called "critical pathway"; moreover, the organism group including human, living in the highest toxicant contaminated area, is called "critical group". In the case of environmental monitoring, if the high risk organism living in the contaminated area has the toxicant concentration level lower than the accepted standard, it can be concluded that the other surrounding organisms also have lower concentration when comparing with the standard.

Especially, the main critical pathways of heavy metals as considered from their physicochemical properties, processes of sinks and processes of remobilization in general surface waters are soil compartment (e.g. aquatic suspended matters and sediments) and biota compartment (e.g. planktons and fish), as well as water compartment upon the changed water body conditions crated naturally or anthropogenically. For risk managing of the heavy metals' contamination in the environmental compartments, it is necessary to detect compartment site and ambient environmental qualities for screening and recognizing probably toxicant problems; for instance, the Minamata disease was proved apparently from mercury contamination and the Itai-itai disease was caused from lead contamination. These heavy metals' contamination would be found in soil compartments because they represent positives ions; and mercury in the form of organometallic, e.g. methyl mercury (CH_3Hg^+), are accumulated in aquatic organisms.

After that, the monitoring and evaluating of environmental qualities must be investigated; the contaminated compartment site is occurred from human activity (e.g. mining, industry) or naturally found in this area, and do the found concentrations exceed the environmental standards or criteria. Additionally, the determining and ensuring command and control procedures would help to monitor that are determined control procedures appropriate for heavy metals protection, or have occurred problems from pollutant been academically prevented and also treated. Furthermore, an important thing is using a chemical transport model, in mathematical terms, to understand the transport and distribution of chemicals including heavy metals between different compartments of the environment. It sometimes can also be used to predict their transportation and distribution from fates and behaviors, as well as apply to in situ monitoring environmental qualities.

CONCLUSION

Heavy metals are non-biodegradable pollutants, several of them have transported and widespread in the environmental compartments (air-water-soil-biota) through mostly anthropogenic activities (e.g. mining, industry, agriculture, etc.). In aerially contaminated soils, they tend to persist for many years in the surface layers of soil. In aquatic systems, heavy metals may become blocked as sinks in bottom sediments, where they may remain for many years. However, because of their remobilizing process if the pH falls, heavy metal solubility increases, and then they become more released. Moreover, they may be accumulated in a wide range of organisms because of their tendencies to follow the same biochemical pathways as the essential element, and because of their fates when adsorbing with organic matter or transforming to the easiest form for bioaccumulation.

These heavy metals' studies would yield the profitability in monitoring the heavy metals contaminated in the environment. The fates and behaviors would tell in which areas should be investigated because of their high concentrations. They also help to assess the risk of exposure of surrounding organisms.

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