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Introduction

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1.1 Arsenic origin, chemistry, and use

On the periodic table of elements, arsenic (number 33) is in group 15 along with nitrogen, phosphorus, antimony, and bismuth (Chapter 2). The *nucleosynthesis* of the element occurs in massive red giant stars and *supernovas* rather than from the *Big Bang*. After formation, arsenic collects in *nebulae*, which may condense into new stars and planets. In our solar system, arsenic has been detected in the atmospheres of Jupiter and Saturn, and trace amounts occur in *meteorites* and Moon rocks. On Earth, the element is largely concentrated in the *core* and in *clay-* and *sulfide-rich* portions of the *crust*. *Hydrothermal fluids* are important in transporting and concentrating arsenic in crustal rocks (Chapter 3). An appropriate understanding of the chemical properties of arsenic (Chapter 2) and its behavior in natural environments (Chapters 3 and 6) are critical in predicting risks to the environment and human health (Chapter 4), as well as selecting effective and economical technologies for treating arsenic-bearing wastes and remediating arsenic-contaminated sites (Chapter 7).

The only stable and naturally occurring *isotope* of arsenic is ^{75}As , where each atom of this isotope has 33 protons and 42 neutrons. The most common *valence states* of arsenic are -3 , 0 , $+3$ and $+5$. Arsenic and its compounds include: elemental forms, *organoarsenicals*, *arsenides*, *arsenosulfides*, *arsenites* and *arsenates*. Arsenic forms also partially substitute for sulfide, *sulfate*, and possibly *carbonate* in a variety of *minerals* (Chapter 2). In the presence of surface and near-surface *aerated* water, arsenide and arsenosulfide minerals *oxidize* to more water-soluble arsenates (Chapter 3).

For centuries, humans have utilized arsenic compounds (especially, realgar (As_4S_4), orpiment (As_2S_3), and arsenolite (As_2O_3)) in a wide variety of products, which included: pigments, medicines, alloys,

pesticides, herbicides, glassware, embalming fluids, and as a depilatory in leather manufacturing. Furthermore, the toxic properties of arsenolite and other arsenic compounds are well known and have been widely used in chemical warfare agents and to commit murder and suicide since ancient times (Chapter 5).

The term ‘*arsenic*’ probably originated from the Persian word *az-zarnikh* or other modifications of its root word, ‘*zar*’, which referred to yellow or gold orpiment (Azcue and Nriagu, 1994), 3; (Meharg, 2005), 39. In ancient Syria, the term became *zarnika*. The ancient Greeks believed that metals and other substances had masculine or feminine properties. They referred to yellow orpiment pigments as ‘*αρρενικόν*’ (*arrenikos* or *arsenikos*), which means ‘potent’ or ‘masculine’ (Azcue and Nriagu, 1994), 3; (Meharg, 2005), 38–39. From these origins, the Greek name became ‘*αρσενικόν*’ (*arsenikon*). The term was translated ‘*arsenicum*’ in Latin and ultimately evolved into ‘*arsenic*’ in French, which is also used in modern English-speaking countries. Other modern names for arsenic include: *arsénico* (Spanish), *Мышьяк* (Russian), 砷 (simplified and traditional Chinese), 砒素 (Japanese), *Arsen* (German), and *arsenico* (Italian). As European chemists began to distinguish between elements and compounds, the noun ‘*arsenic*’ was eventually restricted to refer to element 33.

1.2 Arsenic environmental impacts

In the twentieth century, arsenic was further utilized in livestock dips and feed supplements, semi-conductors, wood preservatives, and medicines. Toward the end of the century, toxicologists and other scientists began to recognize widespread arsenic poisoning in Bangladesh, West Bengal (India), and elsewhere. The arsenic came from their drinking water wells. Additional arsenic-contaminated *groundwater* and other types of arsenic contamination have been identified as serious threats in parts of Argentina, Cambodia, Chile, mainland China, Mexico, Nepal, Pakistan, Taiwan, Vietnam, and the United States (Appendix D). Perhaps, more than 100 million people may be at risk from arsenic-contaminated groundwater (Chapter 6).

Rather than always resulting from industrial spills, the improper disposal of arsenic-bearing wastes, or the excessive application of *arsenical* pesticides, arsenic contamination in groundwater often originates from the mobilization of natural deposits in *rocks*, *sediments*, and *soils*, and sometimes from *geothermal* water (Chapters 3 and 6). The *oxidation* of arsenides and arsenosulfides in mining wastes or natural rock formations can release arsenic into groundwaters and surface waters. Another important source of arsenic in groundwater is the *reductive dissolution* of arsenic-bearing iron and other (oxy)(hydr)oxides in *anaerobic* soils, sediments, and rocks (Chapters 3 and 6). Under oxidizing conditions in surface waters and near-surface groundwaters, arsenic is removed from solution by *sorbing* onto or *coprecipitating* with iron and other (oxy)(hydr)oxides. In many areas, organic industrial wastes, as well as soluble organic matter from livestock manure, septic tanks, and landfills infiltrate into the subsurface. These organic compounds are *reductants*, which increase microbial activity and promote reducing conditions in the subsurface. In the absence of abundant sulfide, the reducing conditions convert Fe(III) into water-soluble Fe(II), which dissolves the iron (oxy)(hydr)oxides and releases their arsenic into associated groundwater (Behr and Beane, 2002; Burgess and Pinto, 2005; Stollenwerk and Colman, 2003). The microbial oxidation of organic matter also produces bicarbonate and other carbonate species, which would raise the alkalinity of groundwater and desorb arsenic from mineral surfaces (Burgess and Pinto, 2005; Appelo *et al.*, 2002; García-Sánchez, Moyano and Mayorga, 2005; Anawar, Akai and Sakugawa, 2004). In particular, any orpiment and realgar in *igneous* and *metamorphic* bedrocks would tend to dissolve in the presence of bicarbonate (Kim, Nriagu and Haack, 2000).

In response to the widespread arsenic contamination (Appendix D), many governments have instituted regulations on the disposal of arsenic-bearing wastes and arsenic emissions from ore smelters and

coal-combustion power plants. The World Health Organization (WHO) also recommended an arsenic limit of $10 \mu\text{g L}^{-1}$ (micrograms per liter) for drinking water, and many developed nations have adopted this recommendation as a regulatory standard (Appendix E). Furthermore, several nations have established programs (such as *Superfund* in the United States) to *remediate* arsenic-contaminated areas. In 1999, the *National Priority List* for the Superfund Program of the US Environmental Protection Agency (US EPA) identified 1209 sites in the United States that had serious environmental and human health risks ((US Environmental Protection Agency US EPA, 2002a), 2; Appendix E). After lead, arsenic was the most common inorganic contaminant (568 sites or 47 % of the total; (US EPA, 2002a), 2).

1.3 Arsenic toxicity

Exposure to arsenic can result in a variety of health problems in humans, including various forms of cancer (e.g. skin, lung, and bladder), cardiovascular and peripheral vascular disease, and diabetes (Chapter 4). Overall, both inorganic and organic As(III) forms tend to be more toxic to humans than the As(V) forms. Humans may be exposed to arsenic through inhalation, dermal absorption, and ingestion of food, water, and soil. Inhalation exposure can result from industrial emissions, cigarette smoking, and *flue gas* from coal-combustion power plants and ore smelters. In air, arsenic primarily *sorbs* onto particulate matter. Once arsenic-bearing gases or particles enter the airway and deposit on lung surfaces, the arsenic is absorbed further into the body. Inhalation of arsenic depends on the size of the particles and absorption depends on the solubility of the chemical form of the arsenic (Chapter 4). When compared with ingestion, the risks associated with the dermal absorption of inorganic arsenic are generally low. Like ingestion, any dermal effects would depend on the source of the arsenic (e.g. water, soil, chromated copper arsenate (CCA)-preserved wood). As discussed in Chapter 4, controlled ingestion studies in humans indicate that both As(III) and As(V) are well absorbed from the gastrointestinal tract. Between 45 and 75 % of the dose of various As(III) and As(V) forms are excreted in urine within a few days, which suggests that gastrointestinal absorption is both relatively rapid and extensive (Tam, Charbonneau and Bryce, 1979; Yamauchi and Yamamura, 1979; Buchet, Lauwerys and Roels, 1981a; Buchet, Lauwerys and Roels, 1981b; Lee, 1999).

1.4 Arsenic treatment and remediation

1.4.1 Introduction

Many methods for treating arsenic in water, gases, and solids are utilized in both *waste management* and remediation. Waste management refers to the proper handling, treatment, transportation, and disposal of wastes or other byproducts from mining, utility, agricultural, municipal, industrial, or other operations so that they are not environmental or human health threats. While waste management attempts to prevent environmental contamination, *remediation* deals with sites that have already been contaminated. Remediation refers to the restoration of a site through the treatment of its contaminated soils, sediments, *aquifers*, air, water, previously discarded wastes, and/or other materials so that they no longer pose a threat to the environment or human health.

Inorganic As(V) and As(III) are the dominant forms of arsenic in most natural waters, wastewaters, contaminated soils and sediments, and solid wastes (Chapter 7). The vast majority (approximately 89–98.6 %) of atmospheric arsenic is associated with particulates rather than existing as vapors. Both gaseous and particulate arsenic are inhalation hazards and may also contaminate surface soils, sediments, and waters

near their points of origin (Chapter 4; (Shih and Lin, 2003; Chein, Hsu and Aggarwal, 2006; Hedberg, Gidhagen and Johansson, 2005; Martley, Gulson and Pfeifer, 2004)).

1.4.2 Treatment and remediation of water

Considering the millions of people threatened by arsenic-contaminated groundwater, low-cost technologies are desperately needed to effectively treat arsenic in water, especially in developing nations. Artificial sorption and coprecipitation with iron (oxy)(hydr)oxides are some of the more effective and popular technologies for removing inorganic As(V) from water (Chapter 7). Coprecipitation is accomplished by adding salts (such as Fe(III) chlorides or sulfates) to the water to precipitate iron (oxy)(hydr)oxides. Sorbents are commonly placed in treatment columns. Iron (oxy)(hydr)oxides usually have to be imbedded in support materials to maintain the *permeability* of the columns. Popular iron (oxy)(hydr)oxide sorbents include: goethite (α -FeOOH), akaganéite β -FeO(OH), and *ferrihydrites* (variable compositions). Considering that many wastewaters and the vast majority of natural waters have pH values below 9 (Krauskopf and Bird, 1995), 225), any dissolved inorganic As(III) in aqueous solutions primarily exists as unreactive H_3AsO_3^0 (Chapter 2). Most water treatment technologies require that H_3AsO_3^0 be oxidized to As(V) *oxyanions* (i.e. H_2AsO_4^- and HAsO_4^{2-}) before treatment (Chapters 2 and 7). However, zerovalent iron (Fe(0)) is an example of a sorbent that can effectively remove inorganic As(III) from water without preoxidation steps. Zerovalent iron may also be installed in *permeable reactive barriers* (PRBs) to remove arsenic from groundwater. Chapter 7 discusses many other treatment and remediation methods for arsenic-contaminated water.

1.4.3 Treatment and remediation of solid wastes, soils, and sediments

As further discussed in Chapter 7, a variety of technologies are available for treating arsenic-bearing wastes or remediating arsenic in soils and sediments. Many, but not all, of the technologies can be utilized in either waste treatment or remediation. Overall, the main goal of waste treatment and remediation technologies is to either encapsulate the arsenic in an inert matrix that resists *leaching* in natural environments (e.g. *in situ* vitrification and *solidification/stabilization* followed by landfilling) or attempt to isolate the arsenic into a smaller and more manageable volume for disposal (e.g. *pyrometallurgical treatment*).

Waste management and remediation technologies for arsenic in solid materials often utilize heat or electric currents. In pyrometallurgical treatment, incinerators or furnaces volatilize and capture arsenic from soils, sediments, or solid wastes. *Vitrification*, including *in situ* vitrification, refers to the melting of soils, sediments, and solid wastes to primarily incinerate organic contaminants and encapsulate arsenic and other inorganic species into melts. The melts then cool into impermeable and chemically resistant glass. The US EPA considers vitrification to be the *BDAT* (*best demonstrated available technology*) for treating arsenic in soils (US Environmental Protection Agency US EPA, 1999), C.1. Electrokinetic methods refer to *in situ* and, in some cases, *ex situ* technologies that remove contaminants from wet soils, sediments, or other solid materials by passing electric currents through them. Unlike *in situ* vitrification, the currents in electrokinetic methods are too low to melt the materials. Instead, the electric currents cause ions (including As(V) oxyanions) and charged particles in aqueous solutions within contaminated solid materials to migrate toward electrodes, where they may be collected or otherwise treated (Mulligan, Yong and Gibbs, 2001), 193, 199–200.

Solidification/stabilization refers to reducing the mobility of a contaminant in sediments, soils, other solids, or even liquid wastes by mixing them with Portland cement, lime (CaO), cement kiln dust, clays, slags, *polymers*, water treatment *sludges*, iron-rich gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), coal *flyash*, and/or other *binders* (Mulligan, Yong and Gibbs, 2001), 193; (Leist, Casey and Caridi, 2000), 132; (US Environmental Protection Agency US EPA, 2002b), 4.1; (Mendonça *et al.*, 2006). Arsenic is immobilized by both physical

and chemical processes. The binder physically encapsulates the contaminant in an inert matrix that resists leaching (*solidification*). The binder may also create chemical bonds with the arsenic (*stabilization*), such as calcium from the binders reacting with As(V) to form calcium arsenates. To maximize the solidification/stabilization of arsenic-bearing wastes, any As(III) is usually preoxidized (Jing, Liu and Meng, 2005), 1242.

During much of the twentieth century, CCA preservatives were widely used in many nations to protect outdoor wood from microorganisms, fungi, wood-feeding insects, and marine borers (Chapters 5 and 7). *Leaching tests* indicate that arsenic from the wood could contaminate surrounding soils, sediments, and water. The risks of arsenic toxicity from exposure to CCA-treated wood are uncertain. There are reports of children experiencing arsenic poisoning from playing in soils near CCA-treated wood (Nriagu, 2002), 20. However, other studies indicate that arsenic exposure from the wood and associated soils is negligible (Pouschat and Zagury, 2006; Nico *et al.*, 2006). Although the wood is no longer commercially available in many nations, the life expectancy of CCA-treated wood is at least 30 years in terrestrial environments and about 15 years in salt water (Christensen *et al.*, 2004), 228; (Hingston *et al.*, 2001), 54. Thus, the issues dealing with the handling and disposal of CCA-treated wood will persist for decades.

Several nations (e.g. Denmark) or regional and local governments (e.g. Minnesota in the United States) have restricted the landfilling and/or incineration of CCA-treated wood. In some circumstances, CCA-treated wood may be mixed with cement and utilized in construction (Gong, Kamdem and Harchandran, 2004). CCA-treated wood may also be detoxified with acidic, organic, or other extracting solutions, provided that the use and disposal of the solutions are cost effective (Helsen and Van den Bulck, 2004), 281; (Kakitani, Hata and Katsumata, 2007). Although CCA preservatives are designed to protect wood from fungi and other organisms, several researchers are developing strains that are resistant to CCA so that they could be used to extract arsenic and detoxify wood wastes.

1.4.4 Treatment of flue gases

About 60 % of anthropogenic arsenic emissions to the global atmosphere originate from flue gases emitted by copper ore smelters and coal-combustion facilities (Matschullat *et al.*, 2000), 301. The main method for reducing arsenic emissions to the atmosphere involves capturing As₄O₆ vapors by injecting sorbents into flue gases before they are released into the atmosphere. Potentially effective sorbents for arsenic in flue gases include: hydrated lime (Ca(OH)₂, portlandite), lime, calcium carbonate (CaCO₃), *limestone*, and *flyash* (Helsen and Van den Bulck, 2004), 287, 289; (Jadhav and Fan, 2001; Taerakul, Sun and Golightly, 2006; Gupta *et al.*, 2007); Chapter 5). The injection of hydrated lime is especially effective and probably removes volatile As₄O₆ through the formation of calcium arsenates (Ca₃(AsO₄)₂ · nH₂O, where n > 0), at least over a temperature range of 600–1000 °C (Mahuli, Agnihotri and Chauk, 1997); Chapter 5).

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