Arsenic Toxicity in the Irrigation Water-Soil-Plant System: A Significant Environmental Problem

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Abstract: Environmental pollution is a major global concern. When sources of water pollution are enumerated, agriculture is, with increasing frequency, listed as a major contributor. One of the major factors determining uptake and toxicity to plants is the form of arsenic (As). Naturally occurring arsenic in groundwater of sedimentary aquifer has emerged as a global problem, and issue of major environmental concern. It is released and contaminated in agricultural soil by natural weathering, industrial production and mining. However, the same water resources are used extensively for irrigation purposes throughout the region. The two most important forms, As (V) and As (III), are taken up by completely different mechanisms. Uptake, accumulation and toxicity vary within and between plant species. In general, more As in the soil leads to higher concentrations in plants, but this depends on many factors. It is recommended to initiate an integrated program to quantify the scale of the problem in combination with the development of a water-soil-plant quality monitoring system for land degradation in agro-ecosystems. This should not only include As, but a range of physical, chemical (nutrients and contaminants) and biological parameters. Further, management options to prevent and mitigate As contamination need to be explored.
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1. Introduction

Agriculture, as the single largest user of freshwater on a global basis and as a major cause of degradation of surface and groundwater resources through erosion and chemical runoff, has cause to be concerned about the global implications of water quality. If water is polluted, it may be dangerous for plants, animals as well as for human being. The associated agrofood-processing industry is also a significant source of organic pollution in most countries. Development increases, so does the need for water. The water quality in a region largely depends on the nature and extent of the industrial, agricultural and other anthropogenic activities in the catchments (Banejad and Olyaie, 2011). Aquaculture is now recognized as a major problem in freshwater, estuarine and coastal environments, leading to eutrophication and ecosystem damage (Ongley, 1996).

Widespread use of arsenicals as pesticides has significantly contributed to the elevation of arsenic concentrations in soils (Adriano, 2001). Arsenic contamination in groundwater is a severe global environmental problem (Yavuz et al., 2010). Arsenic is a heavy metal with a name derived from the Greek word *arsenikon*, meaning potent. Arsenic is ubiquitous, found in air, water, fuels, and marine life. The daily human intake of arsenic contained in food ranges from 0.5-1 mg, with the greatest concentrations coming from fish and crustaceans. Arsenic has been used for a variety of purposes. For Arsenic and its compound are well known for its toxicity and carcinogenicity (Marcus, 2010). Individual exposes to arsenic from various sources like food, air, water, occupational settings and medicines. Contamination of arsenic in ground water is the global problem and millions of people are at a risk of arsenicosis. Contaminated ground water is the main source of exposure to inorganic arsenic to the human population. Inorganic and organic arsenic occur naturally in the environment, with inorganic forms being most abundant. Inorganic arsenic is associated with other metals in igneous and sedimentary rocks, and it also occurs in combination with many other elements, especially oxygen, chlorine, and sulfur. Organic arsenic contains carbon and hydrogen. Both inorganic and organic forms exist naturally in soils, plants, animals, and humans. Most pure, inorganic arsenic compounds are white or colorless powders with no specific smell or taste. Because it is an element, arsenic does not degrade nor can it be destroyed.

Arsenic is a crystalline metalloid that exists in several forms and oxidation states. Its

toxicity and mobility in the environment depend on both its chemical form and species (Pongratz, 1998). Total metal concentration alone is insufficient to assess its environmental impact in contaminated soils. Risk assessment of contaminants requires information on contaminant pools of differential lability and bioavailability in a soil (Wenzel et al., 2006). Soil available arsenic content is a better indicator of its phytotoxicity than total arsenic concentration (Fayiga et al., 2007). However, available arsenic concentration in soils depends on the type and strength of the extracting agent used (Jain and Ali, 2000).

Arsenic (As) is widely distributed in the environment, originating either from As in the soil parent material or from discharge of As onto land as a result of human activities. Consequently, people and livestock are being exposed to As via contamination of drinking water and consumption of food grown in As-contaminated soil or irrigated with As-contaminated water. Understanding how As is taken up by plants and subsequently transformed in plant tissue is therefore essential for estimating the risks posed to human and wildlife populations by As contaminated soils (Meharg and Whitaker, 2002).

Arsenic (As) is a widespread natural element, which is not a bioorganic element to plants (Stoeva et al., 2003). In terrestrial plants, both organic and inorganic As species have been found (Koch et al., 2000; Francesconi et al., 2002), with the inorganic species (Arsenate [As (V)] and arsenite [As (III)]) being the most dominant. Arsenate is the predominant As species in aerobic soils, whereas arsenite dominates under anaerobic conditions (Smith et al., 1998). Arsenic availability to plants is greatly influenced by its forms in soil. Agricultural application of arsenicals has introduced many different kinds of arsenic compounds to the soil environment. These arsenicals may influence arsenic mobility and plant uptake though they are subjected to oxidation–reduction transformation in soils.

Arsenic is a nonessential element for plants, and inorganic As species are generally highly phytotoxic. Biomass production and yields of a variety of crops are reduced significantly at elevated arsenic concentrations, with application of only 50 mg.kg^{-1} to soil significantly decreasing the yields of barley and ryegrass. Arsenic concentrations are generally low in plants (Matschullat, 2000). The limited accumulation of As by roots and its limited translocation to the shoots, is usually used by most plants such as carrot, tomato and grass. These plants contain relatively low arsenic and accumulate arsenic primarily in their root systems (Matschullat, 2000). In all plant species tested so far, it has been shown that

arsenate is taken up via the phosphate transport systems.

1.1. Toxicity of arsenicals

Arsenic is well known for its acute toxicity. For example, an ingested dose of 70-180 mg of arsenic trioxide (As_2O_3) is lethal to humans. Somewhat lower doses produce sub-acute effects in the respiratory, gastrointestinal, cardiovascular, and nervous systems (Jain and Ali, 2000). Chronic exposure to arsenic in drinking water has been linked to serious dermatological conditions, including black foot disease. Epidemiological studies have linked arsenic in drinking water with cancer of the skin, bladder, lung, liver, and kidney (Hindmarsh, 2000) and other ailments. Both As(III) and As(V) are strongly adsorbed in the human body. As(III) tends to accumulate in the tissues, whereas As(V) and organic arsenic are rapidly and almost completely eliminated via the kidneys. The MCL for arsenic in drinking water for many years was 50μg/L, but recent research has suggested that the cancer risk at 50μg/L is unacceptably high. A review of the available arsenic- and health-related data prompted the USEPA to lower the MCL to 10μg/L, the same as the World Health Organization's standard.

1.2. Arsenic and Human Health Effects

Depending on the amount ingested, arsenic can be beneficial (animal studies suggest that low levels of arsenic in the diet are essential) or adverse (high levels can be toxic). The acute lethal dose to humans can be about 2 to 20 mg/kg body weight per day (mg/kg-day). Ingesting high doses of arsenic irritates the stomach and intestines, with symptoms including nausea, vomiting, diarrhea and liver swelling. However, wide recognition of its toxicity makes arsenic poisoning today very rare. Ingesting small amounts over time produces chronic effects such as skin darkening and formation of corns, damage to peripheral nerves, cardiovascular system effects, hair and appetite loss, and mental disorders. Effects from inhaling arsenic dust include respiratory irritation, rhinitis, pharyngitis, laryngitis, and sometimes nasal perforation. Skin contact with inorganic arsenic dusts can cause dermatitis, allergic hypersensitivity, and conjunctivitis. Occupational exposure studies show a correlation between chronic arsenic exposure and lung cancer. Arsenic can also cause reproductive/developmental effects, including spontaneous abortions and reduced birth weights. Epidemiological studies indicate an association between arsenic concentrations in drinking water and increased incidences of skin, liver, kidney, lung, and bladder cancers. Studies also show an association between inhaling arsenic and lung cancer. From these

sets of data, the U.S. Environmental Protection Agency (EPA) has classified inorganic arsenic as a known human carcinogen. Limited information is available on the joint toxicity of arsenic with other chemicals. For neurological effects, the predicted direction of joint toxicity of arsenic and lead is greater than additive, whereas the joint toxicity of these metals is predicted to be less than additive for the kidney and hematopoietic (blood-forming) system. The joint toxicity of arsenic and cadmium on the kidney, hematopoietic system, and male reproductive system is predicted to be less than additive. Additional information on joint toxicity is provided in the companion chemical mixtures fact sheet (Figure 1).
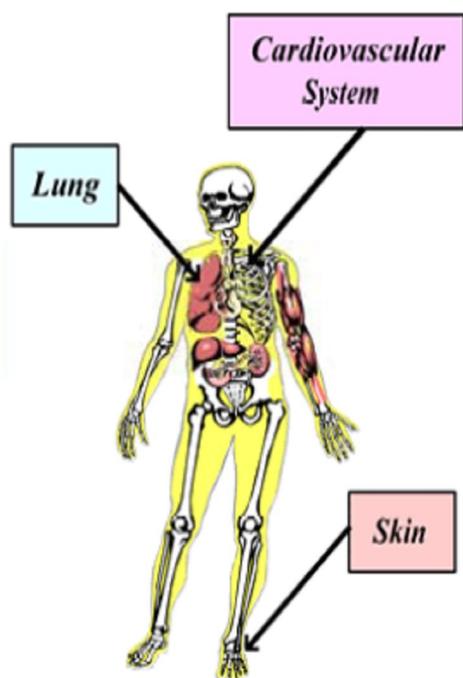

Fig 1. Primary organs affected when arsenic is inhaled or ingested

1.3. Arsenic contaminated irrigation water: the risks

To date, only limited attention has been paid to the risks of using contaminated groundwater for irrigation. Irrigation water with high levels of As may result in land degradation in terms of crop production (loss of yield) and food safety (food chain contamination) (Brammer, 2005; Duxbury et al., 2003). Long-term use of As-contaminated irrigation water could result in As accumulation in the soil. If absorbed by the crops, this may add substantially to the dietary As intake, thus posing additional human health risks. Over time, As accumulation in the soil

could reach soil concentrations toxic to crops, thus reducing yields (Figure 2).
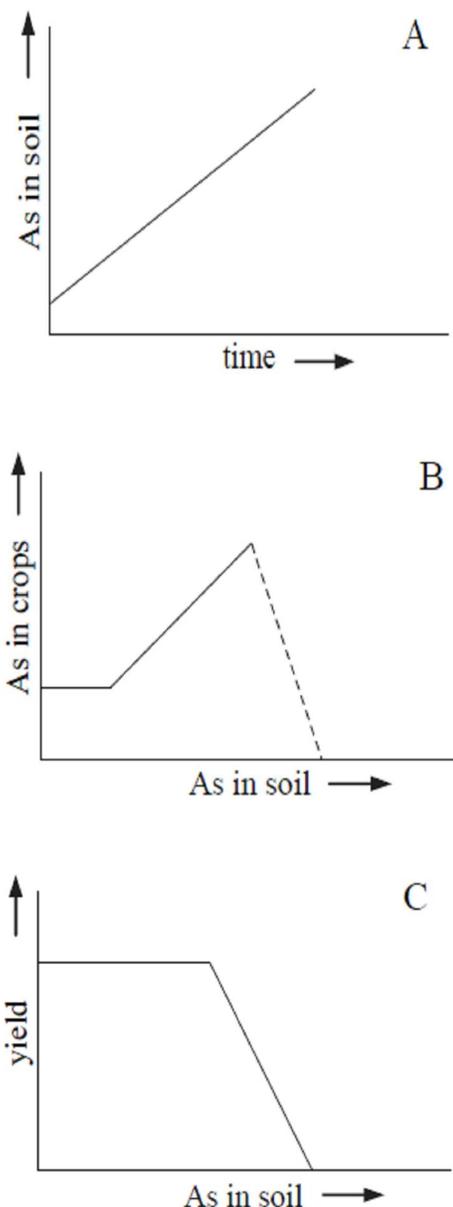

Fig 2. The possible risks of using As-contaminated irrigation water over time.

Note: A: input of As via irrigation water can lead to accumulation of As in the soil over time. B: depending on bioavailability, uptake and transport within the plants, higher soil concentrations may be reflected in higher concentrations in crops. The dotted line indicates that at a certain level the plant growth becomes severely inhibited and As

concentrations in the plants are then no longer relevant. C: with an increase in soil concentration, yields are expected to stay more or less constant until a threshold level is reached, after which yield will decline.

Reliable and representative data are therefore needed to assess and manage the risks of As-contaminated irrigation water.

2. Arsenic in agriculture: current knowledge
2.1.1. Soil Chemistry

Low levels of As are naturally present in the soil (Matschullat, 2000). The background levels are around 5 mg/kg worldwide with substantial variation depending on the origin of the soil (Mandal and Suzuki, 2002). The behavior of As is distinctly different under flooded (anaerobic) and non-flooded (aerobic) soil conditions, with flooded conditions being likely the most hazardous in terms of uptake by plants and toxicity, as will be explained in this chapter. Taking into consideration that rice is the staple crop in Asia, that its cultivation largely takes place under flooded conditions, and that its high demand for irrigation water, often from groundwater resources, understanding the behavior of As under flooded soil conditions is of particular importance.

2.1.2. Arsenics speciation in the soil

As exists in the environment in various organic and inorganic forms (species). The most important inorganic species are arsenate (AsV) and arsenite (AsIII). Monomethylarsenic acid (MMA) and dimethylarsenic acid (DMA) are the most common organic species in the soil, but their natural presence is low compared to inorganic As (Abedin et al., 2002; Fitz and Wenzel, 2002). Speciation of inorganic As in the soil is largely controlled by reduction and oxidation processes (redox). Under aerobic (oxidizing) conditions AsV predominates, whereas AsIII predominates under anaerobic (reducing) conditions (Fitz and Wenzel, 2002; Takahashi et al., 2004). For example, in an experimental paddy field 30 percent of the As was present as AsIII under non-flooded conditions and up to 70 percent was present as AsIII under flooded conditions (Takahashi et al., 2004). Under more reducing conditions, AsIII became by far the predominant species and the solubility of As increased sharply. Microbial activity can influence As speciation via various mechanisms such as redox reactions with Fe and As and via (de) methylation of As species (Fitz and Wenzel, 2002; Mahimairaja et al., 2005).

2. 2. Plants
2.2.1. Uptake

AsIII and AsV are taken up by different mechanisms. AsV is taken up via the high affinity phosphate uptake system (Meharg, 2004). PO_4 additions have therefore been suggested to reduce uptake because of competition between PO_4 and AsV for uptake. For rice grown in pots with soil and irrigated with AsV contaminated water, no effect of PO_4 on As accumulation in rice plants was observed (Abedin et al., 2002). Abedin et al. (2002) suggested that the plants were effectively exposed to AsIII and not to AsV because of the reducing soil conditions. An alternative explanation is that PO_4 competes with AsV both for both sorption at Fe-plaque and for uptake, minimizing the overall effect of PO_4 (Chen et al., 2005). As summarized in various papers, the addition of PO_4 to As-contaminated soils to minimize As uptake is controversial under non-flooded condition (Abedin et al., 2002; Fitz and Wenzel, 2002). AsIII is actively taken up by so-called water channels (aquaporins) in the roots (Meharg and Jardine, 2003). Laboratory experiments have shown that Boro (dry season) rice cultivars take up less AsIII and AsV than Aman (rainy season) rice cultivars. This may be related to physiological or morphological differences between the root systems (Abedin et al., 2002). However, this does not imply that Boro rice will accumulate less As than Aman rice under field conditions, because Boro rice is irrigated with As-rich groundwater whereas Aman rice is rain fed. The uptake mechanism of organic As is largely unclear (Meharg, 2004). It seems that monomethylarsenic acid (MMA) and dimethylarsenic acid (DMA) are taken up by rice plants but that the rate of uptake is much lower compared to inorganic As (Abedin et al., 2002). To date, it has not been possible to predict As uptake by plants from the soil. Most papers only include total As concentrations in the soil and the As concentration in the irrigation water. It has been suggested that total As can be regarded as potentially bioavailable in paddy fields, because most of it is bound to FeOOH. Good correlations between total As in soil and plants are however not always found (Jahiruddin et al., 2005; Miah et al., 2005).

2.2.2. Translocation and accumulation

With the exception of hyper accumulators such as certain ferns, the translocation of inorganic As from the roots to the above ground parts is limited. Organic As is more readily translocated but the uptake is much lower compared to inorganic As (Carbonell *et al.*, 1998). In pot experiments with rice plants exposed to As added via AsV in irrigation water, plant parts were ranked according to the As concentrations as follows: root > straw > husk > grain. Concentrations in all plant parts increased with

the exposure concentration (Abedin *et al.*, 2002). This is a common observation for other plants as well (Bleeker *et al.*, 2003; Carbonell *et al.*, 1998; Sneller *et al.*, 1999b).

3. Soil culture irrigated with As-contaminated water

        Abedin et al. (2002) exposed rice cultivar to AsV and studied growth and As uptake. The first observed adverse effect was a reduced root biomass at 0.2 mg/l. Other effects including reduction of plant height, spiklet weight, number of spiklets and grain yield started at 2 mg/l. In an almost similar experimental setup, a reduced root biomss, grain number and grain weight (g/pot; 26 percent reduction) was found at 1 mg/l (Abedin et al., 2002). Comparing the two studies suggests that the lowest As concentrations associated with toxic effects deviated substantially despite the similar setup. The main reason is probably the difference in the lowest As concentrations used in the irrigation water, namely 0.2 mg/l in Abedin et al. (2002) and 1.0 mg/l in Abedin et al. (2002). In both studies, first effects occurred already at those levels. This indicates that the range of exposure concentrations did not include a concentration so low that it did not cause any effect. It seems that for this particular experimental setup, the lowest concentration causing adverse effects is equal to or below 0.2 mg/l. Smith et al. (1998), cited in Abedin et al. (2002), reported that rice, bean, oats can suffer from phytotoxicity at a soil concentration of 20 mg/kg, whereas for maize and radish this is 100 mg/kg. According to Sheppard (1992), also cited in Abedin et al. (2002), soil type is the most important variable for toxicity of inorganic As to plants, with soil texture one of the most important factors. Inorganic As was five times more toxic in a sandy soil (40 mg/kg) than in a clayey soil (200 mg/kg). Yan-Chu (1994), also cited in Abedin et al. (2002) found a rice yield reduction of ten percent at 13 and 23 mg/kg soil. In sandy soil with 47–52 mg/kg, rice growth was reduced by up to 50 percent and completely inhibited at 109–157 mg/kg soil. Islam et al. (2004) carried out a similar experiment with the same soil and rice cultivars as Jahiruddin et al. (2004) with the difference being that AsV was added via irrigation water during Boro rice cultivation in the Islam et al. experiment. During the Aman cultivation As-free irrigation water was used, resembling the field situation. With an increase in As concentration in the irrigation water, first an increase in grain yield was observed, both for Boro rice and Aman rice. After that, yields declined (Figure 3). As concentrations in grains steadily increased with As levels in irrigation water (Figure 3). Within the tested range of As concentrations in irrigation water, the

observed toxic effects and As accumulation in grains reported by Islam et al. (2004) were far less compared to the observations within the range of soil concentrations used by Jahiruddin et al. (2004). At first, the patterns seem to differ, but a closer look reveals that it is most likely that the range of concentrations used by Islam et al. (2004b) was narrower than that used by Jahiruddin et al. (2004). Comparing the two sets of results for 0–10 mg/kg As in soil shows a similar pattern. In spite of this, it is not known what the true exposure concentrations were and the results cannot be extrapolated to the field. The reports of both sets of authors had the same short comings regarding chemical analysis and the overall description of the methodology.
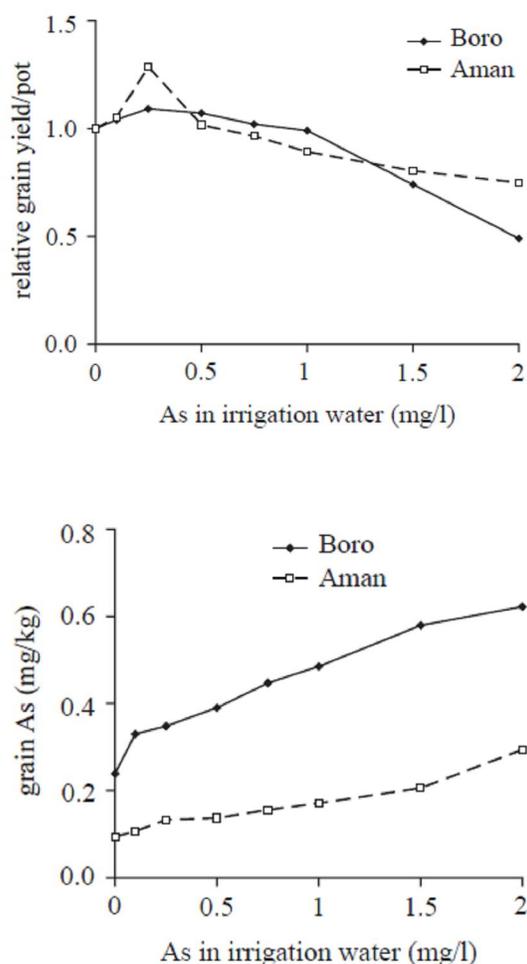

Figure 3. The effect of As on grain yield and on As concentrations in grains of Boro and Aman rice cultivars consecutively grown in the same pots.

Note: Pots were irrigated with contaminated water only during the Boro cultivation. As-free irrigation water was used during the following Aman cultivation.

In conclusion, none of the existing toxicity data can be regarded as representative of the field situation and extrapolations are not yet possible. A better understanding of As in the soil in relation to uptake and toxicity is therefore urgently needed. Ideally, soil parameters should be identified that correlate with uptake and toxicity. The development of a methodology for toxicity experiments that give results representative of field conditions has to be emphasized. With the elevated As levels found in various paddy fields because of long–term irrigation with contaminated water, it may be possible to study phytotoxicity at the field level. Results from such studies would by definition be representative of the field situation, but a thorough understanding of the critical parameters involved would still be necessary in order to extrapolate the data to locations with other environmental conditions (Heikens, 2006).

4. Discussions

There are indications that soil concentrations are increasing over time because of irrigation with As-contaminated water. Data are, however, insufficient in terms of quantity and quality. It is thus still unclear under what specific conditions and over what period of time As is accumulating in the soil. The risk of As-contaminated irrigation water to crop production has received little attention until now. To evaluate current and future soil concentrations, representative toxicity data for crops are needed, both for flooded and non-flooded soil conditions. Thus, field studies to test if As is one of the factors limiting growth in the field should be emphasized. Further, it should become clear what soil parameters correlate with uptake and toxicity and, based on that information, a toxicity database for different rice cultivars and other crops could be developed to set standards for As in flooded and non-flooded soils.

This review has attempted to summarize the incidents of arsenic contamination in the irrigation water-soil-plant system. It poses a significant risk to public health. Therefore, the first priority to remediate the crisis should be early identification of the affected sources, and the next hurdle is to provide arsenic-safe water to the affected masses. It is necessary to seal the highly contaminated tube wells to protect the non-contaminated aquifers.

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