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Arsenic contamination of the soil–wheat system irrigated with high arsenic groundwater in the Hetao Basin, Inner Mongolia, China

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HIGHLIGHTS

- Arsenic-contaminated groundwater led to As accumulation in irrigated soils.
- High As groundwater irrigation increased As bioavailability in topsoils.
- Wheat grain As was positively correlated with bioavailable As forms in topsoils.
- Elevated As in wheat grains was related to high As groundwater irrigation.
- Less problematic water resources were suggested for wheat irrigation.

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ABSTRACT

As one of the most important crop in the world, wheat (*Triticum aestivum* L.) was irrigated with low As water and high As water. However, little is known about As cycling in the soil–wheat–water system. Two wheat fields (site G and site Y), irrigated with high dissolved As (178 μg L^{-1}) groundwater and low dissolved As (8.2 μg L^{-1}) surface water, respectively, were systematically sampled in the Hetao Basin, including irrigation water, soils and plants. The annual As (including dissolved As and suspended As) input per m^2 was estimated at 140 and 36.7 mg in site G and site Y, respectively. Topsoils of site G contained relatively higher As content (average 18.8 mg kg^{-1}) than those of site Y (13.8 mg kg^{-1}). Arsenic content of wheat grains in site G is systematically higher than in the site Y, which were positively correlated with non-specifically sorbed-As and amorphous Fe/Al oxide-bound As in topsoils. Arsenic-contaminated groundwater led to As accumulation in irrigated soils and the increase in As bioavailability, and subsequently resulted in the increase in As content of wheat grain. It suggested that less problematic water resources should be used for wheat irrigation in order to avoid As accumulation in the soil–plant system.

1. Introduction

Arsenic (As) is a ubiquitous trace metalloid and found in virtually all environmental media (Fitz and Wenzel, 2002). Arsenic concentrations in natural waters range from <0.5 μg L^{-1} to >5000 μg L^{-1} (Mandal and Suzuky, 2002). High As concentrations in groundwater have been found in Argentina, Bangladesh, Chile, China, Hungary, Mexico, West Bengal (India), and Vietnam (Smith et al., 1998; Guo et al., 2014). Up to around 30–35 million people in Bangladesh are estimated to be exposed to As in drinking water at concentrations above 50 μg L^{-1}, and six million in West Bengal (Chakrabarti et al., 2002; Mandal and Suzuky, 2002). Skin disorders, including hyper/hypopigmentation changes and keratosis, are the most common external manifestation, although skin cancer has also been identified (Smith et al., 1998). Today, more and more people are aware of the As problem and avoid using highly As-contaminated groundwater as drinking water sources. Chronic inorganic As exposure from dietary sources receives more and more concern (Williams et al., 2007). It is apparent that ingestion of drinking water is not the only elevated source of As to the diet. Long-term use of As-contaminated water for irrigation has resulted in elevated As levels in agricultural soils and therefore food chains (Meharg and Rahman, 2003; Roychowdhury et al., 2005; Williams et al., 2005; Dahal et al., 2008).

It is generally accepted that soil–plant transfer of As is one of the principal pathways for human exposure to As. Epidemiological studies

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further demonstrated that there was a significant correlation between As concentrations in human hairs and those in local rice (*Oryza sativa* L.) and soils (Lin et al., 2001). In Bangladesh, irrigation with As-contaminated groundwater may lead to elevated As concentrations in rice grain and straw, which led to severe losses in yield (Huq et al., 2006; Meharg and Rahman, 2003; Duxbury et al., 2003). Irrigated with high As water, crops showed various As concentrations in plant tissues. Sunflower and maize exhibited decreasing trends in As content from roots to leaves and stems to seeds, in which roots exhibited the highest contents (Neidhardt et al., 2012). The impact of As accumulation in soil on yield and the transfer of As to the food chain depends on its bioavailability in soils.

Recently, it is demonstrated that total content of As in the soils does not necessarily represent its biological availability and potential toxicity (Newman and Jagoe, 1994; Anawar et al., 2006), which are much more important for assessing possible environmental impacts. Sequential extraction procedure has been extensively used to understand physicochemical mobility of metal and metalloid elements and evaluate their potential bioavailability (Hlavay et al., 2004; Bacon and Davidson, 2008). The input of irrigated water As into soils would affect As forms and its potential bioavailability. It is essential to evaluate the effect of high-As water irrigation on soil As forms and its relation to As transfer into crops.

In Inner Mongolia, China, As poisoning from drinking groundwater has been reported since 1990 (Gao, 1990). It has been found that high As groundwater with As concentrations greater than 50 μg L^{-1}, the Chinese drinking water guideline value in rural areas, occurred in 627 villages of Inner Mongolia (Sun et al., 1995). The Hetao Basin is one of the most severely affected areas in Inner Mongolia (Guo et al., 2014), where prevalence rate of chronic As poisoning was around 15.5% (Jin et al., 2003). Although drinking water resources have been changed to low-As tap water for more than 10 years, symptoms of As chronic poisoning in most patients have not been alleviated. Other pathways obviously exist for As from environment into human bodies. Although farmlands in the flat plain are mostly irrigated by the Yellow river-diverted water with low As concentration, groundwater has been used for irrigation near the Langshan mountains, where the diverted water is not available due to the higher altitude (Jia et al., 2014). In this region, more than 10,000 wells are used for irrigation. Recently, Jia et al. (2014) observed that around 70% of investigated wells had As concentrations greater than 10 μg L^{-1}. However, the impact of irrigation with groundwater containing high concentration of As in As bioavailability in soils and As accumulation in crops is less known. Neidhardt et al. (2012) found that As was slightly enriched in the maize field topsoils irrigated with high As groundwater, and roots of sunflower and maize had high As contents. Furthermore, it is unclear how irrigation with groundwater high in As affects As accumulation in the topsoils of wheat field and in the grains of wheat, which is the second largest food crop in China (Hanson et al., 1982) and the main food in the basin.

The objectives of this study are to (1) investigate differences in soil As and As bioavailability in topsoils between wheat fields irrigated with Yellow river-diverted water and with high As groundwater, (2) evaluate the impact of soil As and As forms on As contents in wheat grains, and (3) assess the association of irrigation with high As groundwater with wheat quality in the Hetao Basin (HB).

2. Materials and methods

2.1. Area description

The study area lies in the northwestern margin of the Hetao Basin (HB) between the Langshan Mountain Ranges and the Yellow River (Supporting Information S1). The local climate is characterized by low average annual precipitation of 120–220 mm and high evaporation reaching 2000–2500 mm (Guo et al., 2008; Kottek et al., 2006). Precipitation mainly concentrates in July and August, accounting for 56.3% of the total rainfall. The average annual temperature is 7.7 °C, with the highest temperature in July (23.8 °C) and the lowest temperature in January (−10.5 °C).

The Hetao Basin is known to be one of the Chinese oldest crop-producing areas, where artificial irrigation systems are widely distributed to divert water from the Yellow river into the basin for agricultural irrigation (Guo et al., 2011). As the major food for the local residents, wheat is widely cultivated in the basin. There are two irrigation methods for wheat planting. One is the Yellow River-diverted water irrigation in the flat plain. The other is groundwater irrigation near the alluvial fans. The Yellow River-diverted water normally has low As concentration (Guo et al., 2011), while groundwater for irrigation near alluvial fans mostly has high As concentration (Jia et al., 2014). Two sites were selected for sampling wheat and soils in July 2012 (Supporting Information S1). One site (106°57′39″E, 41°00′02″N) is located near Fengchan village, which has been irrigated with high As groundwater for around eight years (Site G). This site is 5.0 km apart from the Langshan Mountain Ranges. The other site (106°57′04″E, 40°55′13″N) is located near Hongqi village, which has been irrigated with the Yellow River-diverted water for more than 10 years (Site Y). Each site has an area of 140 m^2 (15 m × 8 m). Both sites were used for growing wheat.

2.2. Irrigation water sampling and analysis

Groundwater for irrigation was sampled from deep wells near the site G after pumping (around 60 min) until the flowing water showed a stabilized temperature, pH, EC, and ORP. Parameters including water temperature, total dissolved solids (TDS), pH, ORP, and electrical conductivity (Ec), were measured at the time of groundwater sampling by using a multiparameter portable meter (HI9828, HANNA). Concentrations of S^{2-} and Fe(II) were measured using a portable spectrophotometer (HACH, DR2800) with methylene blue and 1, 10 phenanthroline methods, respectively. Alkalinity was measured using a Model 16900 digital titrator (HACH) using bromocresol green-methyl red indicator. In addition, the Yellow River-diverted water was collected from the irrigation channels. All samples were filtered through 0.45 μm cellulose acetate membrane filters (Sartorius) in the field. Water samples for major and trace element analysis were collected in 100 mL HNO_3-washed polyethylene bottles, followed by addition of 6 M ultrapure HNO_3 to pH <2. Samples for analysis of As species were preserved with 0.25 M EDTA (McCleskey et al., 2004). Samples for anion analysis were not acidified. All samples were subsequently stored cold until analysis. To assess particulate As in water samples, particle-loading filters were also preserved in cold until analysis.

Concentrations of major cations and trace elements were determined by ICP-AES (iCAP 6300, Thermo) and ICP-MS (7500ce, Aglient), respectively. The analytical precision of ICP-AES and ICP-MS was 0.5%. The detection limit for As was 0.01 μg L^{-1}. Unacidified aliquots were analyzed for F^-, Cl^-, NO_3^-, and SO_4^{2-} concentrations by Ion Chromatography with a Dionex DX-120, with the analytical precision less than 3.0%. Arsenic species in groundwater samples were analyzed by HPLC-ICP-MS, with the relative standard deviation (RDS) <±5% and the analytical precision of 2.0% (Guo et al., 2011). Detection limits of As(III) and As(V) were 1 μg L^{-1}. The particle-loading filters were dried at 105 °C, digested by HNO_3–H_2O_2–HF-acid in the laboratory, and then measured with ICP-MS.

2.3. Soil sampling and analysis

From each site, one soil pits of 1 m^3 close to the water inlet was excavated in order to assess the vertical soil geochemistry at harvest time (Fig. 1). Profile samples were taken one from depth intervals of 0–1 cm, 1–2 cm, 2–5 cm, 5–10 cm and in steps of 10 cm up to 1 m in depth. For examination of the lateral As distribution in topsoils, samples were systematically taken at depths of 0–2 cm (Fig. 1). At each wheat site, the sampling grid covered the complete field (15 × 8 m) with a