

Temporal variation of arsenic and nitrate content in groundwater of the Duero River Basin (Spain)



P. Mayorga^a, A. Moyano^a, Hossain M. Anawar^{b,*}, A. García-Sánchez^c

^aDepartamento de Producción Vegetal, Universidad de Valladolid, Soria, Spain

^bSchool of Earth and Environment, The University of Western Australia, Crawley WA 6009, Australia

^cDepartment of Environmental Geochemistry, IRNASA-CSIC, Salamanca, Spain

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ABSTRACT

High concentrations of arsenic (As) in groundwater of the Duero River Basin have created a public health concern in some provinces of Spain. However, the mechanism of As mobilization and influence of surface water–groundwater interaction, nutrients and different geochemical reactions on As removal have not yet been clearly reported. Therefore, this study investigated the possible mechanisms of As release, and temporal variations of As with respect to nitrate content in groundwater. Hydrogeochemical characteristics of groundwater sampled along three years in a region of central Spain showed high As contents exceeding EU guideline value of 10 µg/l. Significant positive correlations were found between As and bicarbonate concentrations in water samples. These results suggest a possible mechanism of As mobilization from sediments to groundwater as follows: bicarbonate ions can displace HAsO_4^{2-} adsorbed on surface of aquifer Fe oxyhydroxide, other minerals and sediments. In addition, the high pH values of the groundwater might also favor the As desorption processes. The results showed that As concentrations in water samples decreased along with the increase in nitrate concentrations across the whole period of study (2001, 2003 and 2007). The negative and significant correlations between As and nitrate contents in water reflect the temporal variation of As concentration due to the effect of surface water–groundwater interaction and nutrient input into groundwater. The increased use of nitrogen fertilizers and pig manure in agriculture practices increased the nitrate content in groundwater during the period of this study, which could have favored the precipitation of Fe oxyhydroxides and As adsorption.

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1. Introduction

High concentration of As in groundwater threatens human health and constitutes a high-priority public health problem worldwide, especially in several countries such as Argentina, Bangladesh, Chile, China, Hungary, India, Mexico, Taiwan, Vietnam and USA (Nriagu, 1994; Welch et al., 2000; BGS-DPHE, 2001; Smedley and Kinniburgh, 2002; Welch and Stollenwerk, 2002). Mechanisms of As mobilization to groundwater have been proposed by different researchers in different types of aquifers. Arsenic release from iron oxides appears to be the most common cause of the elevated As concentration in groundwater (Welch et al., 2000). Smedley and Kinniburgh (2002) reported that the reducing nature of groundwater in Bangladesh would reduce As(V) to As(III) and causes the possible desorption of As, since As(III) is less strongly adsorbed by hydrous ferric oxides (HFO) than As(V). Nickson

et al. (1998, 2000), McArthur et al. (2001, 2004), Kneebone et al. (2002) and Anawar et al. (2003) have reported that reductive dissolution of As-bearing iron oxyhydroxide, and not just As(V) reduction, can be considered the most probable mechanism responsible to mobilize high concentrations of As in groundwater. Using model calculation, Appelo et al. (2002) showed that dissolved carbonate might displace arsenate from the sorption sites of the aquifer minerals causing mobilization of As to groundwater. Bicarbonate ions can extract As from aquifer sediment samples in both oxic and anoxic conditions (Anawar et al., 2004). The results of Harvey et al. (2002) showed that organic carbon may quickly mobilize As in groundwater of Bangladesh, whereas the addition of oxidant compounds, e.g. nitrate, may lower As concentration. Most recently, Seddique et al. (2008) suggested that biotite is a primary source of As in aquifer sediments, where chemical weathering of biotite might be the primary formation mechanism, and prevailing reducing conditions contribute to the expansion of As-enriched groundwater in Bangladesh.

Arsenic adsorption on and desorption from the surfaces of aquifer minerals, especially by HFO, can have an influence on the mobility, reactivity and toxicity of As in groundwater (Smedley

* Corresponding author. Address: School of Earth and Environment, The University of Western Australia, 35 Stirling Highway, Crawley WA 6009, Australia. Tel.: +61 8 6488 1714; fax: +61 08 6488 1050.

E-mail address: anawar4@hotmail.com (H.M. Anawar).

and Kinniburgh, 2002). The strong retention of As by HFO is probably caused by the formation of inner-sphere mononuclear or binuclear monodentate- bidentate complexes (Fendorf et al., 1997). Both the surface complexation by HFO and redox processes involving iron ions have a strong effect on As mobility in groundwater (Cummings et al., 1999). The microbial degradation of organic matter requires oxidant consumption like dissolved oxygen (O_2), nitrate (NO_3^-), etc. Therefore, oxidant or reductant pollutants in groundwater, e.g. nitrate released from the agricultural practices, such as fertilization and widespread use of pig manures could affect As mobility in groundwater aquifers. Nitrate is consumed after O_2 , but before Fe(III), because nitrate is situated after O_2 , and before Fe(III) or HFO in the redox scale (McBride, 1994). Benz et al. (1998) suggested that the oxidation of Fe(II) at circumneutral pH can be achieved by chemoheterotrophic nitrate reducing bacteria. More recently, some studies (Weber et al., 2001; Rhine et al., 2007; Sun et al., 2008) have identified the bacteria as the factor of oxidation of both Fe(II) and As(III) by NO_3^- , together with the CO_2 fixation. Therefore, in aquatic ecosystems, nitrate contamination has a strong influence on the cycle of As, given the oxidation

of Fe(II) to produce As-sorbing particulate like HFO, and to generate also more oxidized form of As, that is As(V), which is more reactive than As(III) (Senn and Hemond, 2002). On the other hand, the inhibition of Fe(III) reduction by denitrification through biotic and abiotic processes, leads to As co-precipitation with or adsorption on HFO in the aquifer sediments. Thus, the increase of nitrate content in groundwater can inhibit the As release from sediments and propitiate the precipitation of Fe(III) oxyhydroxides, which promote a mechanism for the immobilization of As through precipitation and adsorption decreasing the groundwater As content. This explanation can be substantiated taking into account a test result done at Bangladesh, where dissolved As levels declined when nitrate was injected into the aquifer, being probably a result of As adsorption mechanism (Harvey et al., 2002).

In Spain, As was detected in groundwater of the Tertiary Tajo River Basin in the South Iberian Meseta up to a concentration of $91 \mu\text{g/l}$ (Aragónés Sanz et al., 2001; Hernández García and Fernández Ruiz, 2002) and in the Tertiary Duero River Basin of the North Iberian Meseta up to $600 \mu\text{g/l}$ with an average content of $40 \mu\text{g/l}$ (García-Sánchez and Álvarez-Ayuso, 2003; Sahun et al., 2004;

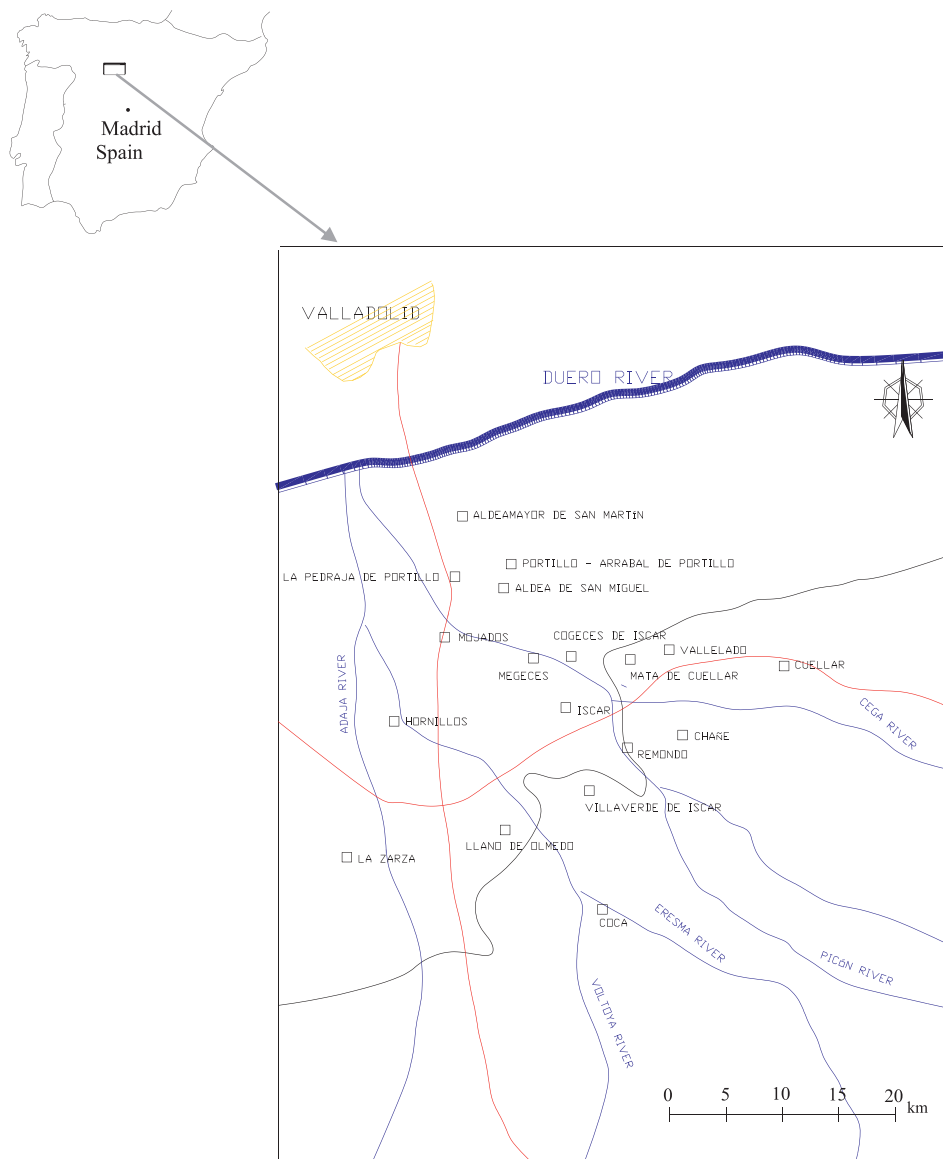


Fig. 1. Sketch of study area and sampling points.

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