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Heavy metal(loid)s and organic contaminants in groundwater in the Pearl River Delta that has undergone three decades of urbanization and industrialization: Distributions, sources, and driving forces

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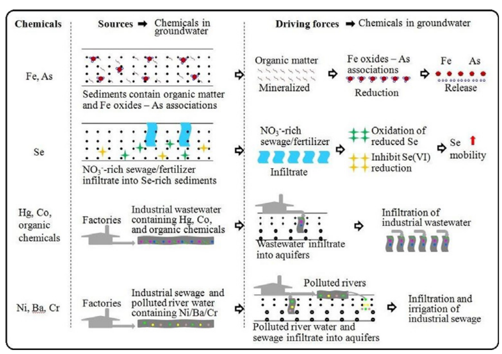
HIGHLIGHTS

- Undrinkable groundwater related to heavy metal(loid)s was mainly due to Fe and As.
- 18 organic chemicals were detected in groundwater but below allowable limits of China.
- Groundwater Fe/As was mainly driven by reduction reaction in Fe/As rich sediments.
- Groundwater Se was mainly driven by the infiltration of NO_3^- into sediments.
- Groundwater Ni/Ba/Cr/Hg/Co and organic chemicals mainly from industrialization

GRAPHICAL ABSTRACT



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ABSTRACT

Urbanization and industrialization have increased groundwater resource demands, and may drive the change of heavy metal(loid)s and organic chemicals in groundwater in the Pearl River Delta (PRD), southern China. Thus, a comprehensive understanding of the distributions, sources, and driving forces of heavy metal(loid)s and organic chemicals in groundwater in the PRD is vital for water resource management in this region. In this study, eight heavy metal(loid)s and fifty-five organic chemicals in groundwater across the PRD were investigated. The results show that undrinkable groundwater related to heavy metal(loid)s was mainly due to high concentrations of Fe (19.3%) and As (6.8%). Eighteen organic contaminants were detected in groundwater in the PRD, where the most frequently detected organic contaminant was naphthalene, and its detection rate was 2.51%. In 5.3% of all groundwater samples, one or more organic contaminants were found. All detected organic contaminants, except ones without allowable limits, in groundwater were at concentrations below allowable limits of China. The mean concentrations of heavy metal(loid)s in granular aquifers were higher than those in fissured and karst aquifers, especially for Fe and As. Except Se, the mean concentrations of other heavy metal(loid)s and the frequency of detection of organic contaminants in groundwater in urbanized and peri-urban areas were higher than those in non-urbanized areas, especially for Hg, Co, and organic contaminants. Fe, As, and Se in groundwater mainly originated from the release of Fe/As/Se rich sediments. The former two were driven by reduction reactions, while the latter was driven by oxidation resulting from the infiltration of NO_3^-. In contrast, other five heavy metal(loid)s and organic contaminants in groundwater mainly originated from the anthropogenic sources, such as the

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infiltration of industrial sewage. It is evident that urbanization and industrialization are two powerful driving forces for heavy metal(loid)s and organic contaminants in groundwater in the PRD.

1. Introduction

The wholesale transformation of agricultural and natural ecosystems to more intensive uses of urbanization and industrialization is among the biggest anthropogenic impacts on earth (Vitousek et al., 1997). In the past few decades, urbanization and industrialization are two of the most widespread environmental changes in China. For instance, the Pearl River Delta (PRD) in the Guangdong Province is one of the fastest developing regions in China. It has undergone a rapid urbanization and industrialization since the late 1970s. Most of the land-use change in the PRD is converted from agricultural land to urban areas, and large-scale investments in industrial development play the major role in urban land conversion (Seto et al., 2002). In addition, the deterioration of surface water quality in this area occurred in the past three decades due to urbanization and industrialization (Cheung et al., 2003; Huang et al., 2013; Ouyang et al., 2006). As a result, groundwater becomes an alternative water resource for drinking, irrigation, and industrial purposes in the PRD. However, the contamination of heavy metal(loid)s and organic chemicals in groundwater often occurs in a rapidly urbanized and industrialized area (Huang et al., 2012; Huang et al., 2013; Leung and Jiao, 2006). Moreover, groundwater with high concentrations of heavy metal(loid)s and/or organic chemicals is harmful to human health and crop growth when it is used for drinking and irrigation (Agency for Toxic Substances and Disease Registry (ATSDR), 2015). Therefore, to improve the groundwater resource management in this area, a comprehensive understanding of the distributions and sources of heavy metal(loid)s and organic chemicals in groundwater in the PRD is necessary.

To date, a couple of studies have already confirmed that heavy metal (loid)s contamination occurred not only in surface water but also in groundwater in some areas within the PRD due to the urbanization and industrialization (Huang et al., 2013; Huang et al., 2014a). Moreover, a previous study has also found the natural source of some metalloids, such as arsenic (As), with high concentrations in a granular aquifer within the PRD (Wang et al., 2012). In addition, the effects of expansion of construction land on some aspects of hydrogeochemical conditions in the PRD have also been well documented in a previous study (Huang et al., 2018). However, a regional scale survey on the distributions, sources, and driving forces of heavy metal(loid)s and organic contaminants in groundwater in the PRD has received little attention.

This study aimed to investigate the distributions of heavy metal (loid)s and organic contaminants in groundwater in the PRD at a regional scale, and to discuss sources and driving forces for heavy metal (loid)s and organic contaminants in groundwater. Among them, the impacts of urbanization and industrialization on the distributions of heavy metal(loid)s and organic contaminants in groundwater in the PRD have been highlighted. The results will benefit groundwater resource management for a sustainable development in the rapidly urbanized areas in China and other countries.

2. Materials and methods

2.1. Study area

2.1.1. Geographical, geological and hydrogeological conditions

The PRD was formed as a result of the Tibetan Plateau uplift during the Tertiary and Quaternary Periods and is located in southern China (Fig. 1). It has a total area of 41,698 km^2 with a population of over 57 million in 2016. The East River, West River, and North River merge in the south of the area and form the Pearl River, which finally discharges into the South China Sea. The central and southern parts of the PRD are mostly covered by Quaternary sediments. Quaternary sediments consist of two terrestrial sequences and two marine sequences interbedded with each other (Zong et al., 2009). The younger marine sequence has an elevation above −20 m, and the older marine sequence is located between −15 m and −40 m (Fig. 1). The younger terrestrial sequence can be sandy fluvial deposits or clayey silt weathered from the in situ materials during the last glacial period to become a local intermediate aquifer. The older terrestrial sequence is dominated by sand and gravel and has become the basal aquifer (Jiao et al., 2010). Groundwater in Quaternary sediments is recharged by vertical infiltration from precipitation and agricultural irrigation, and the lateral flow of bedrock groundwater in hilly regions (Sun et al., 2009). In addition, seawater intrusion of the groundwater occurs over a large area of this region. The bedrocks include shale, sandstone, limestone, dolomite, granite, and gneiss, ranging in age from Cambrian to Tertiary, and crop out in the hilly areas in the PRD. Aquifers in these areas are mainly associated with fractures and with springs discharging up to hundreds cubic meters per day. Karst aquifers account for <10% of the total area and are around the PRD plain, while the groundwater yield can be as high as thousands of cubic meters per day. The general direction of the regional groundwater flow in the aquifers of the PRD is northwest and northeast toward the coast (GHST, 1981).

2.1.2. Expansion of construction land and relevant human activities characteristics

The PRD promoted rural urbanization and industrialization during the past three decades (Ye et al., 2013). From 1988 to 2006, the construction land in the PRD increased to 6816 km^2 in 2006, more than two times that in 1988 (Fig. S1). The increased construction lands during the above period were characterized by numerous factories, a high proportion of non-local population, and a lack of sewer systems, especially in newly formed peri-urban areas (Huang et al., 2013). In contrast, only a small number of factories were built on the urbanized areas formed before 1988 (Sun et al., 2009; Huang et al., 2018). As a result, factories near rivers (or streams) directly discharged wastewater into rivers (or streams) illegally in these newly formed urbanized areas, and sewage infiltration into groundwater commonly occurred in these newly formed peri-urban areas (Sun et al., 2009).

2.2. Sample collection

Around 400 groundwater samples were collected from three types of aquifers (257 wells in granular aquifers, 132 wells in fissured aquifers, and 10 wells in karst aquifers), and the sampling densities were 30–50 samples and 5–10 samples for every thousand square kilometers in plain and hilly regions, respectively. Two 250-mL polyethylene bottles were used to store groundwater for the analysis of inorganic chemicals. One bottle was acidified with nitric acid to a pH of <2 to determine trace elements. The other bottle used for the major ions analysis was unacidified. Another 1-L brown glass bottle for the analysis of organic chemicals was within a stainless steel sampler and collected groundwater (below water table 50 cm) in situ with no headspace. All samples were stored at 4 °C until laboratory procedures could be performed.