



Concentrations and potential health risks of strontium in drinking water from Xi'an, Northwest China

Hui Zhang^{a,*}, Xue Zhou^a, Luobin Wang^a, Wendong Wang^b, Jinlan Xu^a

^a School of Environmental and Municipal Engineering, Xi'an University of Architecture and Technology, Xi'an 710055, China

^b School of Human Settlements and Civil Engineering, Xi'an Jiaotong University, Xi'an 710049, China

ARTICLE INFO

Keywords:

Drinking water
Monte Carlo simulation
Strontium
Health risk

ABSTRACT

Information about the concentrations of strontium (Sr(II)) in drinking water in China and the corresponding health risks to Chinese residents is lacking. This study investigated Sr(II) in drinking water through a monthly sampling campaign in twelve locations in Xi'an, Northwest China. A health risk assessment for different age groups and exposure pathways were carried out by Monte Carlo simulation. The results show Sr(II) concentrations of 0.06–1.69 mg/L in all drinking water samples, which exceeded the minimum reporting level (MRL) of 0.3 µg/L. Also, one sample exceeded the health reference level (HRL) of 1500 µg/L. Higher Sr(II) levels were recorded in groundwater supply zones and springs, and more potential changes in Sr(II) occurred in distribution pipes transporting groundwater. The non-carcinogenic risk associated with Sr(II) exposure via drinking water was less than 1, indicating no significant health risk to the residents of Xi'an. As the first attempt to provide information on the health risks of Sr(II) in drinking water in China, findings from this study can be useful for the development of potential strategies for risk control and management.

1. Introduction

Strontium (Sr(II)) is an alkaline earth metal with high mobility and reactivity. It has four stable isotopes, ⁸⁸Sr, ⁸⁶Sr, ⁸⁷Sr, ⁸⁴Sr, and thirty-one unstable isotopes. Sr(II) can be found in surface water and groundwater as a result of the dissolution of its naturally occurring compounds (celestite (SrSO₄) and strontianite (SrCO₃)). Since the conventional coagulation/filtration treatment process is not effective in Sr(II) removal, Sr(II) enters into drinking water and is transported to customers' taps by distribution systems (O'Donnell et al., 2016).

Sr(II) is an important mineral in human bones and teeth and is not currently regulated in drinking water (Shin et al., 2017). However, recent studies show that ingestion of Sr(II) may pose a potential threat to human health due to its role in abnormal skeletal developments and bone calcification (Langley et al., 2009). Because of the potential hazards to human health, from 2013, the United States Environmental Protection Agency (USEPA) requires that any sample of drinking water with Sr(II) over 0.3 µg/L be reported (USEPA, 2015). Moreover, in October 2014, the USEPA announced a preliminary regulatory determination for strontium in drinking water and set the health reference level (HRL) for Sr(II) at 1500 µg/L (USEPA, 2014). However, until now, the strontium regulation has not been announced by the USEPA because further studies are needed for effective removal methods and the health

risks under unregulated situations (USEPA, 2015).

As the fifteenth most abundant element in the earth's crust, Sr(II) may be widely distributed in groundwater and some surface water near coastal or mountainous regions (Chaalal et al., 2015). According to the monitoring data from 2013 to 2015 in the United States, 5.60% of 4413 public water systems had Sr(II) concentrations over the HRL (1500 µg/L), and all but one system had Sr(II) levels exceeding the minimum reporting level (MRL) (0.3 µg/L) (USEPA, 2015). The average concentration of Sr(II) in United States drinking water is reported to be approximately 1.10 mg/L (Watts and Howe, 2010). Several publications also report on Sr(II) concentrations in drinking water in other parts of the world. El-Sayed and Salem (2015) performed a hydrochemical assessment of surface Nile water and groundwater in Southwest Cairo, and recorded strontium concentrations ranging from 0.630 to 1.213 mg/L with a mean of 0.867 mg/L. Furthermore, Hinwood et al. (2015) reported strontium concentrations of 0.001–1.350 mg/L in Western Australian drinking water, with a median value of 0.13 mg/L.

However, in China, the level of Sr(II) in public drinking water is still unknown, and the corresponding health risks have not been well investigated to date. Since Sr(II) is not included in the Standards for Drinking Water Quality in China, its measurement is ignored in most water supply systems. Consequently, it is of great significance to investigate the concentrations and health risks of Sr(II) in drinking water

* Corresponding author.

E-mail address: zhanghui@xauat.edu.cn (H. Zhang).

<https://doi.org/10.1016/j.ecoenv.2018.08.017>

Received 7 January 2018; Received in revised form 1 August 2018; Accepted 2 August 2018

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in China.

Health risk assessment (HRA) is the most important step before taking regulatory actions on chemicals in drinking water (Pan et al., 2014; Yousefi et al., 2018). HRA estimates the probability of adverse human health effects by using a method based on deterministic values for input data (Pang et al., 2017; Zhai et al., 2017). HRA is commonly composed of five steps, including problem formulation, hazard and dose-response assessment, exposure assessment, risk characterization and uncertainty analysis (Siddique et al., 2015). The methods used in HRA include the deterministic (non-probabilistic) approaches and probabilistic approaches. The former can implement a single-point risk estimation by calculating a hazard quotient (HQ) value (the ratio of an exposure concentration to an adverse effect concentration) based on the average scenarios or the maximum regulatory values (Yousefi et al., 2018). By comparing the estimated HQ values with a reference value of one, the risks caused by the target chemical can be determined. Although the deterministic approaches are simple and easy to use, the results may be unrealistic due to the high variations in input parameters (Lonati and Zanoni, 2012; Hosseini Koupaie and Eskicioglu, 2015). The probabilistic approaches, such as Monte Carlo simulation, can provide a more realistic risk assessment of chemicals. In Monte Carlo simulation, the exposure and effect values are shown with cumulative probability distributions, which are obtained by repeated calculations with random parameter values (Peng et al., 2016). The probabilistic approaches have been applied extensively to examine the risks of various chemicals in drinking water (Wu et al., 2011; Ding et al., 2015; Zhang et al., 2016).

As mentioned above, the levels of Sr(II) in drinking water in China have yet to be documented, and its effects on human health have not been evaluated. Due to the absorption of corrosion products and hydraulic and/or chemical disturbances, the potential concentrations of Sr(II) may vary in drinking water distribution systems (DWDSs) (Gerke et al., 2014; Sun et al., 2017). However, the Sr(II) distributions have not been well investigated in China. Therefore, this study aimed to investigate the levels and variations of Sr(II) in drinking water in Xi'an in Northwest China and to estimate the health risks to residents through the consumption of drinking water based on the measured concentrations of Sr(II). The factors that most affected the outcomes were identified to provide specific guidance for the risk management as well.

2. Materials and methods

2.1. Study area and sampling strategy

Xi'an, the capital of Shaanxi Province, is the largest city in Northwest China. The urban area is 9983 km² and the population is 8.71 million (Zhang et al., 2017a). The total water demand 1100,000 m³ is supplied by six water treatment plants (WTPs) with groundwater and surface water as the feed sources. The daily production of the four groundwater WTPs (No. 2, No. 3, No. 4 and No. 5 WTPs) is 130,000–200,000 m³, while that of the other two surface water WTPs (Qujiang and Nanjiao WTPs) is 710,000–900,000 m³. Groundwater is untreated prior to chlorination in contact tanks, and the dose of chlorine is in the range of 0.5–0.8 mg/L. Surface water is treated by chemical flocculation, settling, and sand filtration (prior to entering the chlorine contact tanks). Polymeric aluminum ferric chloride (PAFC) is used as the coagulant at a dose of 20–30 mg/L. Deep bed filtration at 8–10 m/h is adopted for solid-liquid separation, and the dose of chlorine is 0.5–0.8 mg/L. The produced water from both sources is transported through a single DWDS with a total length of 1493 km. The DWDS is principally made of gray cast iron (66%), steel (11%), ductile iron (10%), unplasticised polyvinyl chloride (UPVC) (6%), concrete (4%), and polyethylene (PE) (3%).

Water samples were collected monthly from outflows of the six WTPs and twelve locations (S1–S12) (shown in Fig. 1) in six administrative zones of Xi'an during the period 5 Jan 2015–4 Dec 2016. The zoning (groundwater and surface water dominated supply) based on

hydraulic analysis is illustrated in Fig. 1 (Zhang et al., 2017a). In Regions 5 and 6, considerable groundwater is provided for the drinking water production. However, in the other regions, drinking water is mainly obtained from Qujiang and Nanjiao WTPs, which is produced from surface water. As described in Fig. 1, the possible origins of S1–S12 were: surface water from Qujiang and Nanjiao WTPs for S1–S5 and S9–S11, groundwater from No. 3–No. 5 WTPs for S6 and S7, groundwater from No. 2 WTP for S12, and blended water from No. 3–No. 5, Qujiang and Nanjiao WTPs for S8.

Samples were collected between 9 A.M. and 5 P.M. from the faucet of selected washrooms near the street. Before sampling, the faucet was turned on and allowed to run for about five minutes to obtain water from the public distribution system and to avoid sampling stagnant water. The samples were collected in cleaned polyethylene bottles. All samples were stored at 4 °C and transported to the laboratory for analysis.

2.2. Sample analysis

All reagents, except hydrochloric acid (guarantee reagent grade), were analytical grade and purchased from Xi'an Chemical Reagents Co., China. All solutions were prepared using deionized water with an electrical conductivity of 18.2 MΩ cm from a Milli-Q system (Millipore, Milford, MA, USA). Standard solution of Sr(II) (1000 mg/L) were purchased from the National Center for Reference Materials (Beijing, China). The Sr(II) working solutions were obtained by stepwise dilution from standard solution just before use.

The collected water samples were filtered through 0.45 μm filters and then acidified to pH < 2 with the addition of ultrapure nitric acid (0.15% v/v). The content of Sr(II) in the treated samples was measured using an inductively coupled plasma atomic emission spectrometer (ICP-AES) with a Thermo Elemental (Franklin, MA) model 6500 Duo ICP-AES. The optimized ICP-AES conditions were as follows: forward power, 1100 W; argon flow of coolant, auxiliary and nebulizer, 18 L/min, 0.8 L/min, and 1.0 L/min, respectively. The wavelength used for Sr(II) determination was 407.771 nm. The measurements were carried out according to the standard methods of water and wastewater examinations (Rice and Bridgewater, 2012). The limit of detection (LOD) of Sr(II) by this method was 0.001 mg/L. All samples were analyzed in triplicate, and the average values were used for data analysis.

2.3. Quality assurance/quality control

All analyses were carried out at the China Urban Water Quality Monitoring Laboratories in Xi'an, which is a standardized laboratory in China. For quality control (QC), each analytical batch contained a procedural blank, fortified samples or certified reference materials (CRMs), and blind duplicates. The CRM GBW(E) 080242 from the National Research Center for Certified Reference Materials (Beijing, China) were employed, and the recoveries ranged between 88% and 110%. Moreover, the analytical value of the CRM was within the 95% confidence interval of the certified value, and the relative deviation of duplicates was within 10%.

2.4. Exposure and health risk assessment

Humans are potentially exposed to Sr(II) from drinking water through oral ingestion, dermal absorption, and inhalation (USEPA, 1989). However, due to the lack of toxicological data for Sr(II) such as inhalation reference dose and transfer efficiency from water to air, inhalation is not included in this study. The population exposure to Sr(II) was determined by estimated daily doses (EDDs), which were calculated for two exposure route as follows (Zimoch and Lobos, 2015):

$$EDD_{\text{oral}} = \frac{C_w \times IR_w \times EF \times ED}{BW \times AT} \quad (1)$$

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