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## Uranium in well drinking water of Kabul, Afghanistan and its effective, low-cost depuration using Mg-Fe based hydrotalcite-like compounds



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#### HIGHLIGHTS

- Elevated levels of uranium were found in well drinking water in Kabul, Afghanistan.
- The uranium was shown by isotopic ratio analysis to be derived from a natural source.
- A depurative that can remove uranium from well water in Kabul was developed.
- A depurative with high performance and easy production at low cost was developed.
- The depurative may improve public health in Kabul through remediation of water.

#### A R T I C L E I N F O

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#### ABSTRACT

Toxic elements in drinking water have great effects on human health. However, there is very limited information about toxic elements in drinking water in Afghanistan. In this study, levels of 10 elements (chromium, nickel, copper, arsenic, cadmium, antimony, barium, mercury, lead and uranium) in 227 well drinking water samples in Kabul, Afghanistan were examined for the first time. Chromium (in 0.9% of the 227 samples), arsenic (7.0%) and uranium (19.4%) exceeded the values in WHO health-based guidelines for drinking-water quality. Maximum chromium, arsenic and uranium levels in the water samples were 1.3-, 10.4- and 17.2-fold higher than the values in the guidelines, respectively. We next focused on uranium, which is the most seriously polluted element among the 10 elements. Mean  $\pm$  SD (138.0  $\pm$  1.4) of the <sup>238</sup>U/<sup>235</sup>U isotopic ratio in the water samples was in the range of previously reported ratios for natural source uranium. We then examined the effect of our originally developed magnesium (Mg)-iron (Fe)-based hydrotalcite-like compounds (MF-HT) on adsorption for uranium. All of the uranium-polluted well water samples from Kabul (mean  $\pm$  SD = 190.4  $\pm$  113.9  $\mu$ g/L; n = 11) could be remediated up to  $1.2 \pm 1.7 \mu g/L$  by 1% weight of our MF-HT within 60 s at very low cost (<0.001 cents/day/family) in theory. Thus, we demonstrated not only elevated levels of some toxic elements including natural source uranium but also an effective depurative for uranium in well drinking water from Kabul. Since our depurative is effective for remediation of arsenic as shown in our previous studies, its practical use in Kabul may be encouraged.

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Abbreviations: MF-HT, magnesium (Mg)-iron (Fe)-based hydrotalcite-like compounds; ICP-MS, inductively coupled plasma mass spectrometry.

#### 1. Introduction

Deficiency of water is a problem in many countries. This problem has recently become more serious due to the disparity of rainfall caused by global warming. Utilization of well water for drinking will, therefore, become more important in the future. However, toxicity of various elements in well drinking water has significant effects on public health worldwide (Kato et al., 2013; Kumasaka et al., 2014; Ohgami et al., 2016; Yajima et al., 2015). For prevention of diseases caused by toxic elements, it is essential to first clarify pollution levels of toxic elements in well drinking water and then to develop an effective depurative.

Natural uranium is an ordinary element found in soil and rocks. Elevated uranium levels in well drinking water have been reported in North America and Europe (Arzuaga et al., 2010). Nephrotoxicity is the hallmark effect of uranium exposure (Arzuaga et al., 2010; Craft et al., 2004). Measurements of biomarkers of proximal tubule damage showed adverse renal effects in populations chronically exposed to elevated drinking-water concentrations of uranium (Arzuaga et al., 2010). In addition to nephrotoxicity, potential evidence of various toxicities of uranium including neurotoxicity, carcinogenicity and reproductive toxicity have been reported in animals and/or humans (Craft et al., 2004). Therefore, WHO recommends less than 30  $\mu$ g/L uranium in health-based guidelines for drinking water quality (WHO, 2011).

Hydrotalcite and hydrotalcite-like compounds, which have a double-layered hydroxide structure, can adsorb various elements through inorganic anion-exchange and other mechanisms (Kiso et al., 2010; Kumasaka et al., 2013, 2014; Turk et al., 2009). Therefore, use of the compounds has been proposed as a technique for removing arsenic (Kato et al., 2013; Kumasaka et al., 2013) and chromium (Xiao et al., 2011) from the water. We have also developed a patented magnesium (Mg)-iron (Fe)-based hydrotalcite-like compound  $[Mg(II)_4Fe(III)_2(OH)_{12}]^{2+}[NO_3xCO_3zH_2O]^{2-}$  (MF-HT), which was also used in this study, after testing various conditions including the ratio of Mg and Fe (Kato et al., 2013; Kumasaka et al., 2013). The patented MF-HT could adsorb arsenic, iron and barium in well drinking water in Bangladesh and Vietnam (Kato et al., 2013; Kumasaka et al., 2013). To our knowledge, however, there has been no report showing that hydrotalcite and hydrotalcite-like compounds can remove uranium in well water.

Provision of high-quality well drinking water is generally important for improving public heath conditions. At present, however, there has been very limited information about levels of elements in well drinking water in Afghanistan, though there was one report showing high levels of arsenic in well water from northern Afghanistan (Amini et al., 2008). In this study, we performed fieldwork research to examine levels of 10 elements in well drinking water from Kabul in Afghanistan. We then proposed how to overcome the pollution of toxic elements in the well drinking water.

#### 2. Materials and Methods

## 2.1. Analysis of 10 elements and isotopic ratio of uranium in well drinking water

Well water is generally used as drinking water in Kabul. Since drinking water has a large effect on human health, we investigated well water in Kabul. Sampling in this study was carried out by the method previously described (Kato et al., 2010, 2013). Sampling was performed in districts 1, 3, 4, 6, 7, 8, 9, 10, 15 and 16, which are recognized by the name in Kabul, Afghanistan. Well water samples were collected in polyethylene bottles. Each bottle was filled with sampled water after rinsing out the inside of bottle with sampled

well water. Tightly capped bottles were sent to Japan by airplane after keeping them at room temperature for a few weeks in Afghanistan. Samples were kept at 4 °C and measurements of total levels of elements were completed within 2 weeks after arrival in Nagoya University. This study was approved by the Ethical Committee of Nagoya University (approval no. 2013-0070) and the Ethical Committee in Chubu University (approval no. 250007) in Japan and the Ministry of Public Health in Afghanistan. Since some owners of wells on private land did not want to have detailed information about pollution of their wells published, we performed sampling after reaching a conditional agreement that information to identify the polluted wells will not be published. The levels of 10 elements in a total of 227 water samples (Table 1) were examined by using inductively coupled plasma mass spectrometry (ICP-MS) (7500cx, Agilent Technologies Inc, CA) according to the method previously shown (Yajima et al., 2012). The limits of detection for uranium and the other 9 elements in ICP-MS were 0.01  $\mu$ g/L and 0.1  $\mu$ g/L, respectively. Mean  $\pm$  SD of the <sup>238</sup>U/<sup>235</sup>U isotopic ratio in water samples (n = 49) including highly uranium-polluted samples was measured by ICP-MS according to the method previously shown (Ma et al., 2000).

#### 2.2. Adsorption and elution experiments using the MF-HT

The MF-HT was synthesized following the method previously described and was used after confirmation of the solid structure by an X-ray diffractometer (Rigaku RINT2000) (Kumasaka et al., 2013). Particles of  $<250 \,\mu$ m in diameter were used in this study. Since the pH range was between 6.0 and 8.0 in well drinking water samples from Kabul, adsorption and desorption experiments using solutions containing uranium (Seishin Trading Co., Ltd.) were performed after adjusting pH in the solutions to 7.0 at room temperature. As Kabul is the premises for practical use, an adsorption experiment using the MF-HT for well drinking water from Kabul was, however, carried out at room temperature without regulating pH.

Adsorption experiments for the solutions containing uranium (Figs. 1 and 2 and Tables 2 and 3) and well drinking water from Kabul, Afghanistan (Fig. 3) were performed. Since 35 of 46 adsorption experiments were performed by the batch method in previous studies (Fan et al., 2012; Sprynskyy et al., 2011; Zou et al., 2009), the method was used to analyze solutions containing uranium and well water. After the indicated percents of weight of the MF-HT had been added to the solutions containing uranium and well water, the solutions were shaken at 300 rpm for the indicated times. The total period after starting incubation of the HF-HT is the indicated time plus 65 s because 65 s is needed until the shaker (Bioshaker BR-21UM, TAITEC Co., Koshigaya, Japan) used in the experiments reaches a constant velocity of 300 rpm. After centrifugation of the suspensions, uranium concentrations in the supernatants were measured by ICP-MS. As shown in our previous reports (Kato et al., 2013; Kumasaka et al., 2013), the equilibrium data using the MF-HT were evaluated by Langmuir isotherms.

Uranium adsorbed by MF-HT was eluted in the alkaline solutions in our experiments. Na<sub>2</sub>CO<sub>3</sub> is cheap and was used as eluant for hydrotalcite in a previous study (Lazaridis et al., 2004). Moreover, Na<sub>2</sub>CO<sub>3</sub> seems to be safe for humans because it is used in medicines (Yuan et al., 2000) and foods (Palou et al., 2001). After considering the possibility of practical use in the future, an elution experiment using Na<sub>2</sub>CO<sub>3</sub> was performed by the method described below. After the adsorption experiment using the MF-HT for 500  $\mu$ g/L uranium-containing solutions, the MF-HT used was resuspended in ultrapure water with 10% Na<sub>2</sub>CO<sub>3</sub> and the desorbed uranium levels from the MF-HT in the solutions were measured. It was also sequentially examined whether the Na<sub>2</sub>CO<sub>3</sub>-treated MF-HT could maintain it's ability to adsorb uranium.

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