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.

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 then focused on uranium, which is the most seriously polluted element among the 10 elements. Mean ± SD (138.0 ± 1.4) of the 238U/235U 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 ± SD = 190.4 ± 113.9 μg/L; n = 11) could be remediated up to 1.2 ± 1.7 μg/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.
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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 deputative.

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 (Kato et al., 2004). Therefore, WHO recommends less than 30 μg/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 \( [\text{Mg}(\text{II})_4\text{Fe}(\text{III})_2(\text{OH})_{12}]^4\text{NO}_3^\text{CO}_3\cdot2\text{H}_2\text{O} ]^{2-} \) (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 health 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 μg/L and 0.1 μg/L, respectively. Mean ± SD of the \(^{238}\text{U}/^{235}\text{U}\) 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., 2006).

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 μm 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; Sprynskyj 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. \( \text{Na}_2\text{CO}_3 \) is cheap and was used as eluant for hydrotalcite in a previous study (Lazaridis et al., 2004). Moreover, \( \text{Na}_2\text{CO}_3 \) seems to be safe for humans because it is used in medicines (Yuan et al., 2006) and foods (Palou et al., 2001). After considering the possibility of practical use in the future, an elution experiment using \( \text{Na}_2\text{CO}_3 \) was performed by the method described below. After the adsorption experiment using the MF-HT for 500 μg/L uranium-containing solutions, the MF-HT used was reuspended in ultrapure water with 10% \( \text{Na}_2\text{CO}_3 \) and the desorbed uranium levels from the MF-HT in the solutions were measured. It was also sequentially examined whether the \( \text{Na}_2\text{CO}_3 \)-treated MF-HT could maintain it’s ability to adsorb uranium.
2.3. Statistical analysis

Because there was no normal distribution for the levels of uranium, titanium and silica, the Mann-Whitney U test and Spearman’s rank correlation test were used for our univariate analyses according to the method previously described (Kato et al., 2004, 2011; Ohgami et al., 2016). A difference with \( P < 0.05 \) was judged as significant.

3. Results

3.1. Levels of 10 elements in well drinking water from Kabul, Afghanistan

Levels of 10 toxic elements [chromium (Cr), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd), antimony (Sb), barium (Ba), mercury (Hg), lead (Pb) and uranium (U)], which are shown in the World Health Organization (WHO) health-based guidelines for drinking-water quality, in well drinking water samples from Kabul, Afghanistan (\( n = 227 \)), and percent of wells with levels exceeding levels in WHO guidelines (unsafe wells) are shown.

### Table 1

Concentrations of 10 elements in well drinking water from Kabul, Afghanistan.

<table>
<thead>
<tr>
<th>Elements</th>
<th>Guidelines ((\mu g/L))</th>
<th>Mean ((\mu g/L))</th>
<th>SD</th>
<th>MAX</th>
<th>MIN</th>
<th>Unsafe wells ((%))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr</td>
<td>50</td>
<td>6.2</td>
<td>12.2</td>
<td>66.0</td>
<td>0.0</td>
<td>0.9</td>
</tr>
<tr>
<td>Ni</td>
<td>70</td>
<td>4.1</td>
<td>6.3</td>
<td>54.2</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Cu</td>
<td>2000</td>
<td>6.5</td>
<td>20.4</td>
<td>173.8</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>As</td>
<td>10</td>
<td>3.9</td>
<td>8.5</td>
<td>104.6</td>
<td>0.0</td>
<td>7.0</td>
</tr>
<tr>
<td>Cd</td>
<td>3</td>
<td>0.0</td>
<td>0.2</td>
<td>0.2</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Sb</td>
<td>20</td>
<td>0.1</td>
<td>0.2</td>
<td>1.6</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Ba</td>
<td>700</td>
<td>79.5</td>
<td>51.5</td>
<td>236.8</td>
<td>6.5</td>
<td>0.0</td>
</tr>
<tr>
<td>Hg</td>
<td>6</td>
<td>0.0</td>
<td>0.0</td>
<td>0.4</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Pb</td>
<td>10</td>
<td>0.2</td>
<td>0.6</td>
<td>4.7</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>U</td>
<td>30</td>
<td>25.1</td>
<td>52.3</td>
<td>515.1</td>
<td>0.5</td>
<td>19.4</td>
</tr>
</tbody>
</table>

Names of 10 elements (Elements), values in WHO health-based drinking water guidelines for elements (Guidelines), mean, standard deviation (SD), maximum (MAX) and minimum (MIN) values for the elements in well drinking water samples from Kabul, Afghanistan (\( n = 227 \)), and percent of wells with levels exceeding levels in WHO guidelines (unsafe wells) are shown.

### Table 2

Uranium absorption in the indicated percent of weight of MF-HT.

<table>
<thead>
<tr>
<th>Percent of MF-HT (wt/wt)</th>
<th>Uranium concentration mean ± SD ((\mu g/L)) (( n = 3 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.010%</td>
<td>51.43 ± 0.76</td>
</tr>
<tr>
<td>0.013%</td>
<td>41.91 ± 0.41</td>
</tr>
<tr>
<td>0.015%</td>
<td>29.21 ± 0.09</td>
</tr>
<tr>
<td>0.017%</td>
<td>6.36 ± 1.25</td>
</tr>
<tr>
<td>0.025%</td>
<td>2.70 ± 1.74</td>
</tr>
<tr>
<td>0.100%</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>0.250%</td>
<td>0.09 ± 0.06</td>
</tr>
<tr>
<td>0.500%</td>
<td>0.03 ± 0.01</td>
</tr>
<tr>
<td>1.000%</td>
<td>0.04 ± 0.00</td>
</tr>
</tbody>
</table>

The indicated percentages of weight of MF-HT (percent of MF-HT = weight of MF-HT per weight of solution) were suspended in solutions containing 500 \(\mu g/L\) uranium. Then the solutions in which MF-HT was suspended were shaken at 300 rpm for 1 h. Uranium concentrations in the supernatants after centrifugation of the solutions are shown.

### Table 3

Uranium absorption time in 1% of weight of MF-HT.

<table>
<thead>
<tr>
<th>Time (sec)</th>
<th>Uranium concentration mean ± SD ((\mu g/L)) (( n = 3 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>15</td>
<td>0.05 ± 0.02</td>
</tr>
<tr>
<td>30</td>
<td>0.02 ± 0.01</td>
</tr>
<tr>
<td>60</td>
<td>0.03 ± 0.01</td>
</tr>
</tbody>
</table>

After suspending 1% of weight of MF-HT in solutions containing 500 \(\mu g/L\) uranium, the solutions were shaken at 300 rpm for the indicated time. Uranium concentrations in the supernatants after centrifugation are shown.

3.2. Levels of 10 elements in well drinking water from Kabul, Afghanistan

Levels of 10 toxic elements [chromium (Cr), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd), antimony (Sb), barium (Ba), mercury (Hg), lead (Pb) and uranium (U)], which are shown in the World Health Organization (WHO) health-based guidelines for drinking-water quality, in well drinking water from Kabul in Afghanistan are shown in Table 1. Although arsenic at a concentration exceeding that in the guidelines was found in 7.0% of the water samples, the concentrations were less than 50 \(\mu g/L\) in all of the water samples except for in one sample in which the concentration exceeded 100 \(\mu g/L\). The mean arsenic concentration in the well drinking water samples from Kabul (3.9 \(\mu g/L\)) was much lower than that in our previous study in Bangladesh (207.0 \(\mu g/L\)) (Kumasaka et al., 2013) and about the same as that in our previous study in Malaysia (5.7 \(\mu g/L\)) (Kato et al., 2010), though arsenic pollution in well water in northern Afghanistan has been previously reported (Anmi et al., 2008). Chromium and uranium at levels exceeding those in the WHO health-based guidelines were also found in 0.9% and 19.4% of the water samples, respectively (Table 1). Maximum concentrations of chromium and uranium in well water samples from Kabul were 1.3- and 17.2-fold higher, respectively, than the values in the fourth edition of WHO health-based guidelines. There was only one sample of well water containing levels of arsenic (14.0 \(\mu g/L\)) and uranium (208.9 \(\mu g/L\)) that exceeded the levels in the WHO health-based guidelines. There was no well water sample in which the levels of chromium and uranium both exceeded those in the WHO health-based guidelines. These results...
indicate that areas with chromium-polluted, arsenic-polluted and uranium-polluted well water are different in Kabul. Since a method for depuration of arsenic from well drinking water was investigated in our previous studies (Kato et al., 2013; Kumasaka et al., 2013; Yajima et al., 2015), we focused on analysis of uranium pollution, the most serious among the 10 toxic elements, in the well drinking water from Kabul.

### 3.2. The isotopic ratio of uranium in well drinking water

Natural uranium is composed of $^{234}$U, $^{235}$U and $^{238}$U (Durakovic, 2005) isotopes. We further analyzed the $^{238}$U/$^{235}$U isotopic ratio to examine the source of uranium in randomly selected well water samples ($n = 49$) from Kabul. Mean ± SD (138.0 ± 1.4) of the ratio was within the range (95% CI: 132.1 < ratio < 144.1) of previously reported naturally occurring uranium (Durakovic, 2005).

### 3.3. Adsorption models for uranium adsorption in MF-HT

After developing the MF-HT by the method reported previously (Kato et al., 2013; Kumasaka et al., 2013), we next examined adsorption kinetics and isotherms. Plural adsorption models including the Freundlich adsorption isotherm were examined, and Langmuir isotherms were used as the most suitable method for investigating the equilibrium adsorption results of uranium in the MF-HT. Our equilibrium data (Fig. 1A) and the Langmuir isotherm for uranium adsorption by MF-HT ($R^2 > 0.95$; Fig. 1B) provide evidence of uranium adsorption by the MF-HT.

### 3.4. The amount of uranium adsorption in the MF-HT

As shown in Table 2, 0.015% weight of the MF-HT depurated solutions containing 500 μg/L uranium to less than 30 μg/L, which corresponds to the value in WHO health-based guidelines. These results suggest that approximately 6.7 L of maximally uranium-polluted well water in Kabul (500 μg/L) can be depurated by 1 g MF-HT.

### 3.5. Adsorption time for uranium in the MF-HT

We further examined the adsorption time mediated by MF-HT. We used uranium solution which concentration was almost the same as the maximal uranium-polluted well water in Kabul (500 μg/L). When treated with 1% weight of the MF-HT, the uranium level in the solution was decreased from 500 μg/L to 0.05 μg/L within 15 s, and the reduced levels were also comparable after 15, 30 and 60 s (Table 3). It should be noted that 65 s, which is the time taken for the shaker to reach a constant velocity of 300 rpm, in addition to the indicated time is required for MF-HT-mediated adsorption as stated in the Materials and Methods section.

### 3.6. Reusability of the MF-HT as a depurative for uranium

We then examined the reusability of the MF-HT. After adsorbing uranium by the MF-HT (lanes 1 and 2 in Fig. 2A), uranium could be eluted from the uranium-adsorbed MF-HT by treatment with 10% sodium carbonate (lane 3 in Fig. 2A). The uranium-eluted MF-HT could again adsorb uranium (lanes 1 and 2 in Fig. 2B), indicating that the uranium adsorption ability of the MF-HT was maintained after treatment with 10% sodium carbonate. These results suggest reusability of the MF-HT as a depurative for uranium.

### 3.7. Adsorption of uranium in well water from Kabul by the MF-HT

We finally examined whether the MF-HT could actually deurate uranium from well drinking water obtained in Kabul, Afghanistan. Adsorption experiments were carried out using polluted well water samples ($n = 11$) containing more than 100 μg/L of uranium. Concentrations of uranium (mean ± SD: 217 ± 112.08 μg/L) in the well water samples treated with 1% weight of the MF-HT for 1 min were reduced to 1.24 ± 1.67 μg/L (Fig. 3). The results indicated that the MF-HT is useful as a depurative for uranium-polluted well water in Kabul.

### 4. Discussion

Previous studies showed that mean or median uranium levels in drinking water in exposed areas were about 7 μg/L in Sweden,
28 µg/L in Finland, 15–20 µg/L in Saskatchewan, Canada, 39 µg/L in Quebec, Canada and >100 µg/L in Nova Scotia, Canada (Arzuaga et al., 2010). Uranium levels in drinking water in non-exposed areas were less than 1 µg/L (Arzuaga et al., 2010; Kato et al., 2010). Previous studies also showed various toxicities of uranium (Arzuaga et al., 2010; Craft et al., 2004). The results indicate that a mean uranium level of 25.1 µg/L in well drinking water is a potential health risk for residents in Kabul because most residents regardless of age and sex use well water as daily drinking water.

The mean uranium concentration in well drinking water from Kabul in our study was more than 9-fold higher than that (2.72 µg/L) in well drinking water from Fallujah, Iraq, where depleted uranium bombs were used (Alaani et al., 2011). Since depleted uranium bombs have been used in Kabul, we examined the source of elevated uranium in the water. The $^{238}\text{U}/^{235}\text{U}$ isotopic ratio in the water samples (138.0 ± 1.4) in this study was in the range of previously reported ratios of natural source uranium (132.1–144.1; 95% CI) (Durakovic, 2005). We further examined the correlations among uranium, titanium and silica in well water samples containing high levels (more than 30 µg/L) of uranium (Kato et al. unpublished observation, n = 41) because a previous study showed that a depleted uranium bomb contains titanium and silica (Trueman et al., 2004). Spearman’s rank correlation coefficients between uranium and titanium and between uranium and silica were 0.004 and −0.227, respectively, in this study. Taken together, the results suggest a natural origin as a mechanism of the increased level of the uranium in well water in Kabul.

In this study, uranium adsorption through the MF-HT was verified by an adsorption experiment using Langmuir isotherms and an elution experiment using sodium carbonate and the MF-HT. The mechanism of uranium removal through the MF-HT might depend on adsorption of the surface complexation, which is similar to goethite (a hydrous ferric oxide), a well-known sorptive material (Durakovic, 2005; Lazaridis, 2003). In particular, adsorption due to highly increased surface basic sites may work in the MF-HT. The MF-HT could remove uranium from water within 60 s. Maximum adsorption capacity for uranium in the MF-HT for the solution containing 500 µg/L uranium was calculated to be 3133 mg/g in theory from the results shown in Table 2. The range of adsorption capacities for uranium in 12 adsorbents was 0.6–1526 mg/g in previous studies (Zou et al., 2009), and thus the adsorption capacity for uranium in the MF-HT is higher than those of the 12 previously reported adsorbents (Zou et al., 2009).

The cost of the hydrotalcite developed by using magnesium and aluminum was shown to be 0.7 cents per kg in a previous report (Gillman, 2006), while the MF-HT was developed by using magnesium and iron in this study. If the costs of aluminum and iron, both of which are raw materials for the hydrotalcite and the MF-HT, respectively, are equivalent and 20 L of maximally uranium-polluted well water in Kabul (500 µg/L) could be adsorbed by 3.2 mg of the MF-HT in theory, the MF-HT could deplete uranium from 20 L of well drinking water for consumption by a family per day at a cost of <0.001 cents per day. Reusability of the MF-HT is also suggested from our results for adsorption and elution of uranium from the MF-HT (Fig. 2). Furthermore, the MF-HT can be produced quickly using inexpensive instruments. It was shown that the MF-HT could adsorb both trivalent arsenic and pentavalent arsenic from well drinking water with high efficiency (43 mg for trivalent arsenic by 1 g of MF-HT) in our previous studies (Kato et al., 2013; Kumasaka et al., 2013). The MF-HT could be a sustainable depuration system for well drinking water in Kabul due to its high performance, low cost and easy production method, though further experiments will be needed to yield practical applications of the MF-HT.

5. Conclusions

In this study, we demonstrated for the first time chromium, arsenic and uranium at levels exceeding those in the WHO health-based guidelines among the 10 toxic elements in well drinking water in Kabul, Afghanistan. We then focused on uranium pollution in the water because the level and frequency of uranium pollution in the water samples were the highest among the 10 elements. After showing that the $^{238}\text{U}/^{235}\text{U}$ isotopic ratio in the water samples was within the range of natural source uranium, we finally demonstrated that the MF-HT could be an effective tool to remediate uranium pollution in well drinking water in Kabul.

Acknowledgements

This study was supported in part by Grants-in-Aid for Scientific Research (A) (15H01743 and 15H02588) and (B) (24390157 and 24406002), Grant-in-Aid for Challenging Exploratory Research (26670525), Grant-in-Aid for Scientific Research on Innovative Areas (24108001) and Grant-in-Aid for Research Activity Start-up (15H06274) from the Ministry of Education, Culture, Sports, Science, and Technology, the Mitsubishi & Co., Ltd. Environment Fund (R13-0014), Foundation from Center for Advanced Medical and Clinical Research of Nagoya University Hospital, The Mitsubishi Foundation (27310), Ichihara International Scholarship Foundation (196) and KENKO-KAGAKU Zaidan (Health Sciences Foundation).