Perchlorate in sewage sludge, rice, bottled water and milk collected from different areas in China

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Abstract

As a new emerging environmental contaminant, perchlorate has prompted people to pay more attention. The presence of perchlorate in the human body can result in improper regulation of metabolism for adults. Furthermore, it also causes developmental and behavioral problems for infants and children because it can interfere with iodide uptake into the thyroid tissue. In this paper, perchlorate in sewage sludge, rice, bottled drinking water and milk was detected for investigating the perchlorate pollution status in China. The places, where the samples were collected, cover most regions of China. Therefore, the final data on perchlorate levels will give an indication of the perchlorate pollution status in China. The final determination of perchlorate was performed by ion chromatography–electrospray tandem mass spectrometry with negative mode. The concentration of perchlorate in sewage sludge, rice, bottled drinking water and milk was in the range of 0.56–379.9 μg/kg, 0.16–4.88 μg/kg, 0.037–2.013 μg/L and 0.30–9.1 μg/L, respectively. The results show that perchlorate has been widespread in China.

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Keywords: Perchlorate; IC-ESI-MS/MS; Sewage sludge; Bottled water; Rice; Milk

1. Introduction

Perchlorate is regarded as a new emerging persistent inorganic contaminant because of its specific properties, such as high water solubility, mobility and stableness (Canas et al., 2006). It mostly exists in the form of ammonium perchlorate, potassium perchlorate and sodium perchlorate, which are used as solid rocket oxidant and ignitable source in munitions and fireworks, or used in roadside flares and in air bag inflation systems (Urbansky, 1998). Naturally occurring perchlorate has been found in nitrate deposits in Chile (Orris et al., USGS Open-File Report, 2003; Environmental news, 2001). Improper treatment and accidental releases can result in contamination of environmental water and drinking water. Also, it can persist for decades due to the specific properties that mentioned above (Hogue, 2003). In recent years, perchlorate has been recognized as a new threat to the environment and drinking water. It has been listed as a new emerging contaminant in several reviews on environmental analytical methodology (Koester, 2005; Richardson and Ternes, 2005).

The toxicity of perchlorate is also receiving considerable attention in recent years. Urbansky (Urbansky, 2000) indicated that perchlorate can be taken into the thyroid and interfere with iodide uptake because it has similar size as iodide. Consequently the production of thyroid hormones was reduced and the thyroid functions were affected, which may result in the improper regulation of metabolism for adults and the developmental and behavioral problems for infants and children. In some cases, thyroid gland tumors can be caused due to the disruptions in thyroid hormone levels (http://www.nsf.org/consumer/drinking_water/perchlorate_reduction.asp?program=WaterTre). Thyroid gland tumors were spotted in rodent animals after exposure to high dose of perchlorate (Ting et al., 2006). Based on these findings, several related departments have established standard for perchlorate. In the National Academy of Science’s (NAS’s) January 2005 report (National Academy of Sciences, 2005), 0.7 μg/kg/d as a maximum.

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permissible dose for perchlorate was proposed, which correlates to a drinking water equivalent level of 24.5 μg/L if drinking water was considered as the only source of perchlorate. In February 2005, the United States Environmental Protection Agency (US EPA) also established 24.5 μg/L as an official reference dose (RfD) for perchlorate in drinking water, which is consistent with the RfD recommended by NAS (U.S. Army Center for Health Promotion and Preventive Medicine). However, there are currently no enforceable perchlorate safety standards at state or federal level, although some states have proposed their own recommended limits. Massachusetts has proposed a drinking water standard of 2 μg/L, and California is considering 6 μg/L (Rajagopalan et al., 2006). Moreover, perchlorate has been listed on the EPA’s Drinking water Contaminant Candidate List prepared under the Safe Drinking Water Act (SDWA) (Perciasepe, 1998). This suggests that perchlorate may be subject to federal regulation at some time in the future.

In accordance with the ever-increasing concern regarding perchlorate in the United States, more and more reports have been published on the occurrence of perchlorate in different environmental matrices such as groundwater, milk, biological sample, et al (Krynitsky et al., 2004; Snyder et al., 2005; Valentin-Blasini et al., 2005; United States Government Accountability Office (GAO), 2005; CDHS, 2005; Martine-lango et al., 2006; Stetson et al., 2006; Barron et al., 2006; Dyke et al., 2006). For example, the federal and state agencies investigated the status of perchlorate pollution in groundwater, surface water, soil and public drinking water in 35 states in US and found the presence of perchlorate at almost 400 sites. The highest perchlorate concentration was found in California and Texas (United States Government Accountability Office (GAO), 2005). In November 2005, California Department of Health Services (CDHS) found that perchlorate has existed in 395 wells in 96 water systems and the maximum concentration was 820 μg/L (CDHS, 2005). Snyder (Snyder et al., 2005) et al determined perchlorate in 11 different natural water bodies and 21 brand bottled drinking water, and found that there was perchlorate pollution in most samples.

With the intense investigations on the occurrence of perchlorate in US, it was clear that perchlorate can be considered as a widespread environmental contaminant. However, there are few data available in China and other countries reflecting their national perchlorate pollution status. As there are lots of fireworks produced and set off in most regions in China, there could be perchlorate pollution in China. Liu (Liu et al., 2002; Liu et al., 2003) et al detected perchlorate in water in several water plants. Nevertheless, there is no extensive research on the occurrence of perchlorate in China. In order to provide a preliminary assessment on the presence of perchlorate in China, we collected samples of sewage sludge, rice, milk and bottled water from different parts of China. These samples were subsequently analyzed for perchlorate by IC-ESI-MS/MS. The IC-ESI-MS/MS method is considered to be unambiguous and high sensitive method that has been used for determination of perchlorate in lettuce, cantaloupe, bottled water, and milk with limit of quantitation of 1.0 μg/kg, 2.0 μg/kg, 0.5 μg/L, and 3.0 μg/L respectively (Krynitsky et al., 2006; Krynitsky et al., 2004).

There are several reasons why we choose the four matrices that mentioned above. Firstly, to our knowledge there are no published reports for the detection of perchlorate in sewage sludge. In addition, it is a significant investigative object for monitoring contaminants in environment because the sewage sludge can accumulate many elements and compounds. The US EPA (DTSC, 2004) indicated that most standard physical and chemical water or wastewater treatment processes are not generally applicable to remove or destroy the perchlorate ion. Thus, there is a possibility that perchlorate may also accumulate in sewage sludge. Furthermore, in China some sewage sludge is often used as fertilizer because it contains many nutritious compounds for crops. If high concentration of perchlorate in sewage sludge, then there is a risk that perchlorate could be absorbed by humans after a series of transfers. Therefore, the determination of perchlorate in sewage sludge provides an excellent method for evaluating both the prevalence and magnitude of exposure. Secondly, rice is one of the main crops and is the basic food for more than 60% of the population in China. Around 20% of the crop farming areas in China is used for planting rice and 50% of farmers are engaged in planting rice (Zhai and Cheng, 2003; overview of rice production). Perchlorate could be taken up and accumulated in rice from soil, ground water and so on. Thus, the determination of perchlorate in rice is important for assessing the health effect on the general population and would also illustrate the perchlorate pollution status in China. As bottled drinking water and milk are both important commodities, they were therefore also included as investigative objects. The extensive data collected in this paper can help to reflect the current status of perchlorate pollution in China.

2. Experiment section

2.1. Sample collection

A total of 31 sewage samples were collected from 26 cities in China between February and June in 2005. These cities cover most regions of China, and their locations are shown in Fig. 1. The samples were stored in a freezer at −20 °C until analysis.

A total of 65 rice samples were collected from 26 provinces in China between February and August in 2005. No samples were collected from Qinghai, Shanxi and Xizang Province where the rice was not planted. The samples were stored in a freezer at-20 °C until analysis.

All bottled drinking water and milk were purchased from large-scale supermarkets in Beijing between February and March in 2006. The bottled drinking water samples covered almost all the brands that we can obtain and the milk samples included all the popular brands that sell well in China. The samples were stored in a freezer at 0–4 °C until analysis.

2.2. Materials

All solutions were prepared with deionized water that was further purified by EASYpure LF system (Barnstead, USA) with a specific resistance of 18.3 MΩ cm−1. Acetonitrile with chromatographic grade was purchased from Merck Co. Perchlorate was obtained by dilution of concentrated stock solution prepared by dissolving solid reagent (Analytical grade, Beijing Chemical Plant, China) in deionized water. The stock solutions were stored in a refrigerator at 4 °C. Standard solutions were prepared daily by serial dilution of the stock solution prior to use.
2.3. Samples preparation

All samples went through a series of pretreatment processes before analysis, except for bottled drinking water, which needed no further cleanup. The pretreatment processes for sewage sludge, rice and milk are as follows.

Sewage sludge samples were freeze-dried and homogenized by sieving through a stainless steel 75-mesh (0.5 mm) sieve. 2.0 g samples were weighed into a 50 mL conical flask added with 20 mL water and then extracted in mechanical shaker at a speed of 200 rpm for 3 h at room temperature. Then, the mixture was centrifuged for 25 min at 6000 rpm and the supernatant solution was taken out. At last, the 15 ml of supernatant solution went through 0.22 $\mu$m Nylon membrane, OnGuard H and OnGuard RP successively, which were used for the removal of solid substance, metallic ion and hydrophobic compounds respectively.

All rice samples were air-dried and ground into powder and then homogenized with a 40-mesh sieve. 5.0 g of rice powder was weighed into a 50 mL conical flask added with 20 mL water and then extracted in a shaker at 200 rpm for 2 h at room temperature. Then, the mixture was centrifuged for another 25 min at 6000 rpm. After that, the supernatant solution was filtered through 0.22 $\mu$m Nylon membrane and pre-activated OnGuard RP, successively.

The pretreatment procedure for milk was based on a literature written by Aribi (Aribi et al., 2006). 5 mL of milk sample was put into a 50 mL conical flask and added with 5 mL of deionized water and 20 mL of acetonitrile that was used for the removing of protein. After shaking by hand for 2 min the mixture was transferred into a centrifuge tube and centrifuged at 6000 rpm for 20 min at room temperature. Supernatant was filtered through a 0.22 $\mu$m pore size nylon membrane before injection.

2.4. Instrumental analysis

The quantification of perchlorate was performed on an ion chromatography (Dionex) coupled with an electrospray tandem mass spectrometer (ESI-MS-MS) (ABI US). The method (IC-ESI-MS/MS), which we adopted, is based on the one reported by Aribi (Aribi et al., 2006). The ESI-MS/MS was operated in MRM mode and negative electrospay ionization. The transition 100.8/84.9, 98.8/66.9 and 100.8/68.9 were monitored for confirmation the target analyte and 98.8/82.9 for quantifying. Samples were injected into the sample loop using an AS50 autosampler (Dionex). Quantification was carried out with the external calibration standard method. Calibration curves constructed for perchlorate were in the range of 50.0–50,000 ng/L for sludge, rice and milk and 10.0–10,000.0 ng/L for bottled drinking water. A quadratic calibration equation ($R^2=0.99$) was obtained and used for quantification of perchlorate. The method detection limits (MDL) of perchlorate for sludge, rice, milk, and bottled drinking water were 0.1 $\mu$g/kg, 0.04 $\mu$g/kg, 10.0 ng/L and 2.0 ng/L, respectively.

3. Results and discussion

3.1. Perchlorate in sewage sludge

Total 31 sewage sludge samples were collected from different sewage plants that are located in different areas of China. The result showed that perchlorate was detected at different concentrations in all samples (as shown in Table 1). Fig. 2 showed the concentration distribution of perchlorate in sewage sludge and Fig. 4 showed the distribution frequency of perchlorate concentration in sewage sludge with the sorting bins arranged in a logarithmic scale. As shown in Fig. 4, the solid line indicates a Gaussian distribution and the limited number of data available could loosely fit in a lognormal distribution, which means that the datum were authentic. We can find out the concentration distribution of perchlorate easily from the figure.

The concentration of perchlorate in sewage sludge was in the range of 0.56–379.9 $\mu$g/kg, with the average concentration at 21.7 $\mu$g/kg. This indicates that the perchlorate contamination is widespread in China. Among these, the concentration of perchlorate in the sample in the Suzhou sewage treatment plant was found to be the highest, which was 379.9 $\mu$g/kg. Detailed information on the sewage sludge plant was collected, including the treatment capacity,
population served by the plant, sources of the sewage (domestic, industrial, or combined), and type of sludge treatment and so on. It was found that 58% of the wastewater was collected from electronic industry and 42% from domestic sewage in the Suzhou sewage treatment plant. As indicated in the “perchlorate treatment technology update” (EPA 542-R-05-015, 2005) edit by USEPA, perchlorate has a wide variety of uses in areas ranging from electronics manufacturing to pharmaceuticals. Therefore, it was easy to interpret the high concentration of perchlorate in the Suzhou sewage sludge. It is noticeable that although more than 90% of wastewater in Urumchi, Xining and Beijing1 sewage treatment plants comes from domestic sewage, high concentration of perchlorate was also spotted in the three samples. This indicates that perchlorate exists in our daily life and we have been widely exposed to perchlorate.

As for the different sewage treatment plants in the same city, the concentration of perchlorate was also quite different. For example, the maximum concentration of perchlorate detected in the samples from Xiamen was 9.08 μg/kg and the minimum was 0.56 μg/kg. Different source of wastewater resulted in the different perchlorate content. And the similar concentrations of perchlorate were obtained in the two samples from Guangzhou. Much higher concentration of perchlorate (37 and 72 μg/kg) was spotted in Beijing sludge samples besides Suzhou.

<table>
<thead>
<tr>
<th>Sampling site</th>
<th>Perchlorate (μg/kg)</th>
<th>Sampling site</th>
<th>Perchlorate (μg/kg)</th>
<th>Sampling site</th>
<th>Perchlorate (μg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Baoding</td>
<td>17.9±1.4</td>
<td>Hangzhou</td>
<td>2.34±0.33</td>
<td>Xiamen1</td>
<td>3.41±0.33</td>
</tr>
<tr>
<td>Beijing1</td>
<td>37.0±2.6</td>
<td>Hefei</td>
<td>6.26±0.09</td>
<td>Xiamen2</td>
<td>6.85±0.08</td>
</tr>
<tr>
<td>Beijing2</td>
<td>72.7±0.7</td>
<td>Lanzhou</td>
<td>4.68±0.14</td>
<td>Xiamen 3</td>
<td>0.56±0.01</td>
</tr>
<tr>
<td>Changsha</td>
<td>2.58±0.06</td>
<td>Linyi</td>
<td>1.84±0.03</td>
<td>Xiamen 4</td>
<td>9.08±.22</td>
</tr>
<tr>
<td>Chongqing</td>
<td>4.06±0.21</td>
<td>Pingdingshan</td>
<td>4.02±0.12</td>
<td>Xi’an</td>
<td>7.57±0.16</td>
</tr>
<tr>
<td>Datong</td>
<td>1.65±0.02</td>
<td>Qingdao</td>
<td>8.97±0.31</td>
<td>Xi’ning</td>
<td>16.3±0.3</td>
</tr>
<tr>
<td>Guangxi</td>
<td>2.77±0.06</td>
<td>Shanghai</td>
<td>1.70±0.03</td>
<td>Yinchuan</td>
<td>3.58±0.14</td>
</tr>
<tr>
<td>Guangzhou1</td>
<td>8.67±0.29</td>
<td>Shenyang</td>
<td>3.19±0.11</td>
<td>Zaozhuang</td>
<td>7.30±0.22</td>
</tr>
<tr>
<td>Guangzhou2</td>
<td>5.91±0.01</td>
<td>Suzhou</td>
<td>379.9±14.7</td>
<td>Zhengzhou</td>
<td>4.95±0.07</td>
</tr>
<tr>
<td>Haerbin</td>
<td>7.21±0.09</td>
<td>Tianjin</td>
<td>8.21±0.30</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Haikou</td>
<td>2.41±0.23</td>
<td>Urumchi</td>
<td>30.5±0.8</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2. The concentration distribution of perchlorate in sewage sludge.
3.2. Perchlorate in rice

In this paper a total of 65 rice samples were analyzed. The results are shown in Table 2. Fig. 3 showed the concentration distribution of perchlorate in rice in a map. The concentration of perchlorate in rice was in the range of 0.16–4.88 μg/kg. The average concentration was 0.83 μg/kg. There was perchlorate detected with concentrations more than 1.0 μg/kg in 17 of all the 65 samples. Similar to the sewage sludge sample, the data on rice could also loosely fit in a log-normal distribution and reasonably reflect perchlorate pollution status of some areas in China. The distribution frequency of perchlorate concentration in rice, with the sorting bins arranged in a logarithmic scale, is shown in Fig. 4b.

Although no distinct pattern on the distribution of perchlorate in rice samples can be deduced, the result still showed that there was a prevalent perchlorate exposure in rice in China. The highest concentration (4.88 μg/kg) perchlorate was found in sample of Hebei. It is reported that perchlorate has been detected in food crops, plant species and fruits such as tobacco, lettuce, wheat, alfalfa, soybean, tomato, cucumber, lemon, grapefruit, orange and cantaloupe (Sanchez et al., 2006; Krynitsky et al., 2006; Sanchez et al., 2005a,b; Jackson et al., 2005; Yu et al., 2004; Sundberg et al., 2003; Ellington et al., 2001). Previous studies (Smith et al., 2004; Jackson et al., 2005) have also showed that perchlorate-tainted irrigation water, soil and fertilizer were the potential source of perchlorate that detected in food and crops. Significant content of perchlorate was detected in tobacco and some leafy vegetables as well as fruiting and seed crops that were irrigated with some perchlorate-contaminated water (Sanchez et al., 2006; Sanchez et al., 2005a,b). The trunks, roots and branches of a lemon tree had the concentration of perchlorate below or close to minimum reporting level, while the fruits and leaves contained perchlorate with the content as high as 128 and 1835 g/kg (dry weight) respectively (Sanchez et al., 2006) after irrigation with water that contained an average of 6 μg/L perchlorate for 5 years. It was showed that leaves and fruits were the significant portions of plants and trees to accumulate perchlorate and the accumulation factor in leaves was much higher than in fruits. It was supposed that accumulation occurred when water transpires through the leaves and the accumulation was the

![Fig. 3. The concentration distribution of perchlorate in rice.](image-url)
highest in outer leaves (Sanchez et al., 2005). So rice may also have the ability to accumulate perchlorate from water and soil. Therefore, even trace levels of perchlorate in source waters and soils will result in high concentration of perchlorate in crops, and ultimately be absorbed by human beings. The reason of different concentrations of perchlorate in rice samples detected may be the trait of fluidity and variable nature of the irrigation water.

3.3. Perchlorate in milk

It is universally known that milk is important for human health, especially for infants and children because the thyroid hormone is essential for the development of the brain (Kirk et al., 2003). Dasgupta (Kirk et al., 2003, 2005) detected perchlorate in 47 dairy milk samples collected from 11 states and 36 human breast milk samples collected from 18 states in USA. The study showed that detectable perchlorate was present in all the dairy milk samples and in 35 of 36 human breast milk samples. The maximum values of perchlorate in dairy and breast milk were 11 and 92 μg/L with average levels of 2.0 μg/L and 10.5 μg/L, respectively (Kirk et al., 2005). Dasgupta (Dyke et al., 2007) also analyzed the perchlorate in dairy milk samples from 48 different locations in Japan (mean 9.4±2.7 μg/L, n=54). In this paper, we analyzed perchlorate in 17 milk samples, which were collected from large-scale supermarkets in Beijing and covering all the well-known and marketable brands. Total twelve brands of milk including 8 sorts of pure milk and 9 kinds of sour milk were investigated. The result is shown in Table 3. The highest and lowest concentration detected in pure milks was 7.62 (Brand 2#-1) and 0.69 μg/L (Brand 12#) and the results in sour milks were 9.1 (Brand 1#-2) and 0.3 μg/L (Brand 7#). There were significantly differences in the same brand with different flavor except for brand 2#. The reason for that may be that different treatment for milk was used. The average levels in pure and sour milks were 3.98 and 4.24 μg/L respectively, and the total mean value was higher than the average level (2.0 μg/L) of perchlorate detected in dairy milk samples by Dasgupta (Kirk et al., 2005). Compared with the data (the average value is 5.76 μg/L) given by the US Food and Drug Administration (US FDA), the average concentration in this paper was lower and detectable levels of perchlorate were spotted in all the 17 milk samples.

Fig. 4. Frequency distribution of perchlorate content of (a) sewage sludge and (b) rice samples. The bins successively span perchlorate concentration differentials of 1, 2, 4, 8 μg/L for (a) and 0.1, 0.2, 0.4, 0.8 μg/L for (b), in a logarithmically increasing order.

<table>
<thead>
<tr>
<th>Milk</th>
<th>Sample no.</th>
<th>Perchlorate (μg/L)</th>
<th>Sample no.</th>
<th>Perchlorate (μg/L)</th>
<th>Sample no.</th>
<th>Perchlorate (μg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brand 1#-1</td>
<td>5.30±0.30</td>
<td>Brand 4#-1</td>
<td>2.98±0.11</td>
<td>Brand 8#-2</td>
<td>0.34±0.02</td>
<td></td>
</tr>
<tr>
<td>Brand 1#-2</td>
<td>9.1±2.9</td>
<td>Brand 4#-2</td>
<td>6.56±0.24</td>
<td>Brand 9#</td>
<td>0.85±0.07</td>
<td></td>
</tr>
<tr>
<td>Brand 2#-1</td>
<td>7.62±0.21</td>
<td>Brand 5#</td>
<td>0.90±0.06</td>
<td>Brand 10#</td>
<td>1.39±0.03</td>
<td></td>
</tr>
<tr>
<td>Brand 2#-2</td>
<td>7.9±0.3</td>
<td>Brand 6#</td>
<td>6.38±0.04</td>
<td>Brand 11#</td>
<td>1.75±0.09</td>
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<tr>
<td>Brand 3#-1</td>
<td>7.60±0.19</td>
<td>Brand 7#</td>
<td>0.30±0.01</td>
<td>Brand 12#</td>
<td>0.69±0.08</td>
<td></td>
</tr>
<tr>
<td>Brand 3#-2</td>
<td>5.75±0.21</td>
<td>Brand 8#-1</td>
<td>4.52±0.78</td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 4

Perchlorate in Bottled Water (n=3)

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Perchlorate (μg/L)</th>
<th>Sample no.</th>
<th>Perchlorate (μg/L)</th>
<th>Sample no.</th>
<th>Perchlorate (μg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>&lt;0.002</td>
<td>1</td>
<td>0.037±0.002</td>
<td>12</td>
<td>0.088±0.003</td>
</tr>
<tr>
<td>2</td>
<td>&lt;0.002</td>
<td>2</td>
<td>&lt;0.002</td>
<td>13</td>
<td>0.046±0.002</td>
</tr>
<tr>
<td>3</td>
<td>&lt;0.002</td>
<td>3</td>
<td>&lt;0.002</td>
<td>14</td>
<td>&lt;0.002</td>
</tr>
<tr>
<td>4</td>
<td>&lt;0.002</td>
<td>4</td>
<td>0.061±0.004</td>
<td>15</td>
<td>0.140±0.004</td>
</tr>
<tr>
<td>5</td>
<td>&lt;0.002</td>
<td>5</td>
<td>&lt;0.002</td>
<td>16</td>
<td>0.117±0.003</td>
</tr>
<tr>
<td>6</td>
<td>&lt;0.002</td>
<td>6</td>
<td>&lt;0.002</td>
<td>17</td>
<td>&lt;0.002</td>
</tr>
<tr>
<td>7</td>
<td>&lt;0.002</td>
<td>7</td>
<td>0.213±0.013</td>
<td>18</td>
<td>0.077±0.010</td>
</tr>
<tr>
<td>8</td>
<td>0.037±0.007</td>
<td>19</td>
<td>0.088±0.002</td>
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</tr>
<tr>
<td>9</td>
<td>0.332±0.017</td>
<td>20</td>
<td>&lt;0.002</td>
<td></td>
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</tr>
<tr>
<td>10</td>
<td>0.049±0.008</td>
<td>21</td>
<td>&lt;0.002</td>
<td></td>
<td></td>
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<tr>
<td>11</td>
<td>0.247±0.013</td>
<td>22</td>
<td>2.013±0.015</td>
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</tbody>
</table>
The detectable perchlorate has been spotted in lettuce, cantaloupe and some leafy vegetables. A large variety of forage has been proven to accumulate perchlorate that originates from irrigation water (Jackson et al., 2005; Dyke et al., 2007). As young peoples are the main consumer of milk and the main negative effect of perchlorate is the insufficient mental development to children, the discovery of perchlorate in milk should be considered seriously.

3.4. Perchlorate in bottled drinking water

In this paper the perchlorate in bottled drinking water was also analyzed to assess the perchlorate pollution in China. A total of 29 bottled drinking water samples were investigated, in which seven were purified water samples. The result showed that no perchlorate was detected in 15 samples, including all seven purified water samples. In the other 14 samples, low concentrations of perchlorate were detected. The average and highest concentration was 0.25 μg/L and 2.01 μg/L (No.22), respectively. The concentration of perchlorate in all samples but one was lower 0.50 μg/L. On the whole, low perchlorate pollution was found in bottled water, especially for the purified water. An investigation conducted by US FDA in 2004 regarding the occurrence of perchlorate in the US (Exploratory data on perchlorate in food, 2004), found that there was no perchlorate detected in most bottled water. The concentration in bottled water was lower than underground water and surface water in many areas in USA. Both the result from our study and the US FDA study showed that bottled drinking water has not been extensively polluted by perchlorate.

We hypothesize that there are two reasons for the low or no detectable concentration of perchlorate in bottled water. Firstly, the main sources of bottled water are often nature reserves, where there are few industry and pollution sources. Secondly, as indicated in the hygienic standard of bottled purified drinking water of China (GB17324, 1998), distillation or ion exchange methods are usually used for the treatment for the bottled water. So perchlorate could be removed from the water because it has high affinity with the ion exchange stationary phase. That no perchlorate was detected in all purified water samples could also confirm this point. Consequently, there is no or low concentration of perchlorate in bottled drinking water. As shown in Table 4, only one sample contained a perchlorate concentration higher than 0.5 μg/L. In general, the concentration in all samples was lower than RID in drinking water regulated by EPA.

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