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Contribution of tap water to chlorate and perchlorate intake: A market basket study ☆

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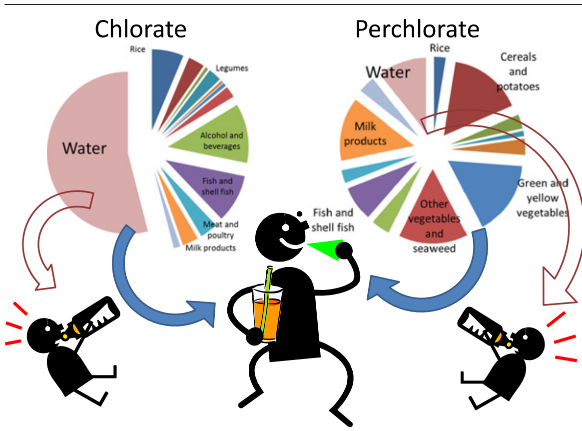
HIGHLIGHTS

- Tap water contribution to total chlorate intake was first revealed.
- Cooking with tap water significantly influenced total chlorate intake.
- Chlorate intake from water is important especially when rice is major food.
- Total perchlorate intake was higher than the previous U.S. study due to vegetables.
- Total chlorate and perchlorate intake from baby formulas and water were high.

GRAPHICAL ABSTRACT



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ABSTRACT

The contributions of water to total levels of chlorate and perchlorate intake were determined using food and water samples from a market basket study from 10 locations in Japan between 2008 and 2009. Foods were categorized into 13 groups and analyzed along with tap water. The average total chlorate intake was 333 (min. 193–max. 486) μg/day for samples cooked with tap water. The contribution of tap water to total chlorate intake was as high as 47%–58%, although total chlorate intake was less than 32% of the tolerable daily intake, 1500 μg/day for body weight of 50 kg. For perchlorate, daily intake from water was 0.7 (0.1–4.4) μg/day, which is not high compared to the average total intake of 14 (2.5–84) μg/day, while the reference dose (RfD) is 35 μg/day and the provisional maximum tolerable daily intake (PMTDI) is 500 μg/day for body weight of 50 kg. The highest intake of perchlorate was 84 μg/day, where concentrations in foods were high, but not in water. The contribution of water to total perchlorate intake ranged from 0.5% to 22%, while the ratio of highest daily intake to RfD was 240% and that to PMTDI was 17%. Eight baby formulas were also tested — total chlorate and perchlorate intakes were 147 (42–332) μg/day and 1.11 (0.05–4.5) μg/day, respectively, for an ingestion volume of 1 L/day if prepared with tap water.

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1. Introduction

Chlorate and perchlorate are known as micropollutants in water. Chlorate is an impurity in sodium hypochlorite used as a disinfectant, and was added to the Japanese drinking water quality standard in November 2007 (in effect from April 2008) (MHLW, 2007). One of the known health effects of chlorate is oxidative damage to red blood cells, and the tolerable daily intake (TDI) was set to 30 μg/kg/day (MHLW, 2003). Chlorate is mainly thought to be present in water and its intake via the atmosphere seems unlikely (WHO, 2011). Therefore, 80% of TDI was allocated to drinking water, and the standard value of 600 μg/L was derived and established (MHLW, 2007). At present, 80% of TDI is also allocated to water in the guidelines issued by World Health Organization (WHO); due to the differences in standard body weight, the value in the WHO guideline is 700 μg/L (WHO, 2011).

Perchlorate is utilized for various purposes, including as a propellant for rocket fuel, explosives, fireworks, and airbags, and is also included in sodium hypochlorite (Kosaka et al., 2011). Perchlorate is known to suppress the uptake of iodine in the thyroid gland. In February 2005, the U.S. National Academy of Sciences (NAS) announced a perchlorate reference dose (RfD) of 0.7 μg/kg/day (NRC, 2005). In the same year, the U.S. Environmental Protection Agency (USEPA) announced a drinking water equivalent level (DWEL) of 24.5 μg/L (EPA, 2009) (when 100% contribution rate is assumed as 2 L of water a day consumed by an adult of body weight 70 kg). In 2008, the USEPA issued an Interim Health Advisory Level (HAL) of perchlorate of 15 μg/L (EPA, 2008). The USEPA has decided to regulate perchlorate under the Safe Drinking Water Act (SDWA) and is now developing a proposed national primary drinking water regulation for perchlorate and anticipates publication of the proposed rules for public review and comment in 2013 (EPA, 2012). JECFA (2011) established a the provisional maximum tolerable daily intake (PMTDI) of 0.01 mg/kg based on human data and including an uncertainty factor of 10 considering potentially vulnerable subgroups, such as pregnant women, fetuses, newborns and young infants.

In 2011, the Water Supply Division, Ministry of Health, Labour and Welfare, Japanese government, posed an evaluation value of 25 μg/L for perchlorate, assuming that the allocation of TDI to perchlorate in water is 10%, while it has been included in "the items for further study" as supplementary items to the Japanese Drinking Water Standard (MHLW, 2011).

Previously, we measured the concentrations of chlorate and perchlorate in bottled water and commercial beverages (Asami et al., 2009a), and also reported that the concentration of chlorate in some tap water exceeded 600 μg/L before the standard for chlorate was in effect (Asami et al., 2008) The highest chlorate concentration, 2900 μg/L was found due to high dose of sodium hypochlorite in 2006. Chlorate was present in degradation byproducts of sodium hypochlorite added in water purification process as an oxidative chemical and/or disinfectants (Kosaka et al., 2007). We also investigated the current state of chlorate and perchlorate in the Tone River basin in Japan, a main water source for Tokyo. The results indicated that perchlorate was present in environment water, drinking water, and water supplies in a wide region in Tone River basin due to environmental contamination mainly attributable to industrial effluents from few specific sources including a chlorate and perchlorate manufacturer and a facility conducting electrolysis processes. Takatsuki et al. (2009) made a survey on perchlorate contamination in leafy vegetables sold in Tokyo. Among 82 leafy vegetable samples tested, perchlorate was detected in the range of 0.3 ng/g and 29.7 ng/g in 79 samples. It was found at relatively high concentration in spinach and leafy green vegetables. Although it is not clear why they have higher concentration, it may have been possibly affected from water used for cultivation, as one of the sources of perchlorate.

In setting the guideline value for chlorate, the allocation ratio to water is 80% as one of disinfection byproducts, which is much higher than the default assumption of 10%–20% for other items, since chlorate was not considered to be exposed from other sources than water (WHO, 2011). To authors' knowledge, there is no data on total chlorate intake, therefore, intake survey through food is important, especially when considering tap water.

In 2008, the U.S. Food and Drug Administration reported the results of a perchlorate intake survey in the USA (Murray et al., 2008). However, the intake in Japan has yet to be investigated where cooking (boiling) and eating of rice at home is common, which seems a little different from that in the USA and European countries and which might be affected by constituents in tap water. The contribution of tap water is important to consider the allocation ratio to water based on the diet and, to our knowledge, there have been no previous surveys of chlorate intake.

In addition, the diet of infants differs from that of adults. The WHO guideline specifies the amount of drinking water for children weighing 10 kg as 1 L per day, and that for infants weighing 5 kg as 0.75 L per day (WHO, 2011). The concentration of perchlorate in milk was investigated in both Japan and the USA (Dyke et al., 2007). A relatively high concentration of perchlorate was detected in Japan. As infants rely on breast milk or baby formula for their daily nutritional intake, data regarding the amounts of chlorate and perchlorate in breast milk and formula are required to allow estimation of their intake levels by infants; however, no such data have been reported in Japan.

In the present study, we analyzed samples for the Total Diet Study (TDS) in cooperation with the National Institute of Health Sciences, Japan, and local health science institutes, estimated the amounts of chlorate and perchlorate intake, and compared the levels of their intake derived from food and tap water. In addition, a purchase survey was conducted on baby formula.

2. Materials and methods

2.1. Reagents and solutions

All solutions were prepared from ultrapure water obtained using a Gradient A10 water purification system (Millipore, Bedford, MA). Calibration standards were prepared by diluting 1000 mg/L certified standard solutions of chlorate (Kanto Chemical, Tokyo, Japan) and perchlorate (GFS Chemicals, Columbus, OH) in ultrapure water.

2.2. Test samples

From 2008 to 2009, foods were purchased and collected using the market basket method at 10 different locations across Japan, and were prepared at each location for use as test samples. Locations were selected to cover throughout Japan considering geographical and cultural differences as in the National Nutrition Survey (MHLW, 2005). At each sampling location, about 150 kinds of food were purchased from local grocery stores according to the methods used by the National Nutrition Survey (MHLW, 2005). Local custom and preference were taken into account when food items were selected, so the composition of the composite food samples was a little different to each other. Based on the amount of food intake according to the survey, and using the same method as described previously in detail (Ohno et al., 2010), a wide variety of foods were divided into 13 different groups (Table 1). After appropriate pre-cooking (normally boiling or baking), the foods were mixed and homogenized to make composite samples. Tap water was used for the pre-cooking at six locations, and pure water was used at other four locations. Representative intake levels for each food group in this study are shown in Fig. 1. The 14th group consisted of tap water from municipal drinking water treatment plants that used surface water as the source. The amount of water intake was not included in the above-mentioned national survey, so the level is represented as 2 L based on normal assumptions