Exposure assessment of metal intakes from drinking water relative to those from total diet in Japan
Koichi Ohno, Kohei Ishikawa, Yuki Kurosawa, Yoshihiko Matsui, Taku Matsushita and Yasumoto Magara

ABSTRACT
Daily intakes of 17 metals (boron, aluminium, chromium, manganese, nickel, copper, zinc, arsenic, selenium, molybdenum, cadmium, antimony, lead, uranium, magnesium, calcium, and iron) via drinking water and total diet were investigated in six cities in Japan. The daily metal intakes were estimated and compared with tolerable daily intake (TDI) values proposed by the WHO or Joint FAO/WHO Expert Committee on Food Additives for toxic metals and with recommended dietary allowances (RDAs) or adequate intake (AI) values proposed for essential metals by the Japanese Ministry of Health, Labour and Welfare. Among the 13 toxic metals, mean dietary intakes of 10 (except arsenic, selenium, and molybdenum) were less than 50% of TDI, suggesting that for these 10 metals the allocation of intake to drinking water in establishing guidelines or standards could possibly be increased from the normal allocation of 10–20% of TDI. For the 13 toxic metals, the contribution of drinking water to TDI was 2% or less in all six cities. Mean dietary intakes of the essential elements magnesium, calcium, and iron were less than the RDA or AI values. Drinking water did not contribute much to essential metal intake, accounting for less than 10% of RDA or AI.

Key words | dietary metal intake, drinking water quality, health risk assessment

INTRODUCTION
In the Guidelines for Drinking-water Quality of the World Health Organization (WHO), the guideline values for non-carcinogenic chemicals are derived from a certain percentage of tolerable daily intake (TDI). This percentage should be allocated on the basis of actual data on daily intakes from other sources such as food, air, and soil, but such data are not always available. As noted in the third edition of the guidelines (WHO 2004), when data on daily intake of the chemical are insufficient, the default allocation percentage to drinking water is 10% of TDI. In recent addenda to the guidelines, however, the allocation approach has changed twice. In the first addendum, the description of a specific percentage as the default allocation was changed to: “Where appropriate information is not available, values are applied that reflect the likely contribution from water for various chemicals” (WHO 2006). The values generally vary from 10 to 80%, considering the exposure from all sources (WHO 2006). In the second addendum, the use of a default allocation value was re-established and the value was changed from 10 to 20% of TDI (WHO 2008). The second addendum stated that an allocation of 20% is still protective and that 10% was found to be excessively conservative (WHO 2008). The collection of chemical intake data from other sources such as food is becoming more crucial for the review and establishment of local drinking water quality standards that reflect actual exposures to chemicals.

Although numerous studies have measured daily metal intake from total diet (Biego et al. 1998; Bordajandi et al. 2004; Maitani 2004; Santos et al. 2004; Turconi et al. 2009),
dietary patterns as well as dietary metal intake vary among countries and regions. For example, like many Asians, Japanese eat rice as a staple food and consume a lot of seafood, and this diet differs from that in other parts of the world. When reviewing or establishing local drinking water quality standards, it is important to take into account these local dietary patterns. Recently, an issue about risk assessment of essential yet toxic metals has been posed (Aggett 2008; WHO 2008). For example, copper plays an essential role as a central component of many redox active enzymes (Stern 2008), and manganese is essential for skeletal development, immune system function, and energy metabolism (Santamaria 2008). Deficiencies in such essential metals linked to enzyme activities contribute to the progression of disease (Gambling 2008). Although these elements are essential to biological processes in humans, they can also be toxic when consumed in excessive amounts (Fraga 2005). Therefore, both deficient and excessive intake of certain essential elements poses health risks.

In the present study, we investigated daily metal intake (DMI) of 17 metals (toxic, essential yet toxic, and essential metals) from total diet and drinking water in six cities in Japan. We estimated the contribution to TDI of toxic elements as proposed by the WHO or Joint FAO/WHO Expert Committee on Food Additives (JECFA). We also estimated the contribution to recommended dietary allowances (RDAs) proposed by the Japanese Ministry of Health, Labour and Welfare (JMHLW). The RDA represents the dietary intake level that is sufficient to meet the nutrient requirements for 97–98% of a Japanese population of a certain age and gender (JMHLW 2005). We focused on the contribution of metal intake via drinking water to TDI and/or RDA. For toxic metals, the margins of TDI that could be allocated to drinking water for establishment or review of drinking water quality standards are discussed. The contribution of drinking water to daily essential metal intake is also discussed.

METHODS

Sample collection

Samples were collected by the market-basket method from six cities across Japan. At each sampling location, about 150 kinds of food were purchased from grocery stores according to the methods used by the National Nutrition Survey (JMHLW 2004). Food items that are generally cooked before consumption were cooked by the usual methods such as boiling or baking. The items were cooked without addition of any oil or seasoning, because these additives belonged to different categories. Then, all food items were categorized into 15 groups in accordance with the classification of the National Nutrition Survey (Figure 1). Finally, food samples of each group were homogenized to make a composite sample. Drinking water samples were collected from the tap where the preparation of the food was performed. In all six cities, drinking water was processed in municipal drinking water treatment plants that use surface water as the source. The drinking water samples were assigned to the 14th group. The food composite samples were stored at −30°C in a deep freezer and were defrosted just before analysis. Drinking water samples were stored at 4°C in a refrigerator.

Analytical methods

The composite food samples were digested in a microwave digestion system (ETHOS TC, Milestone S.r.l., Bergamo, Italy) by the following procedure. A portion of 0.5–1.0 g (wet weight) was weighed into a PTFE vessel, and 4 mL of nitric acid and 1 mL of hydrogen peroxide (Ultra Pure Grade; Kanto Chemical Co., Inc., Tokyo, Japan) were added. The basic program of the microwave digester was as follows: increase the temperature from room temperature to 100°C in 1 minute, followed by 200°C in 2 minutes, 300°C in 3 minutes, 400°C in 4 minutes, 500°C in 5 minutes, and 600°C in 6 minutes. The samples were allowed to cool to room temperature before analysis.
to 210°C over 30 min, maintain that temperature for 15 min, and then cool to room temperature over 10 min; the maximum power was 1,000 W (Dolan & Caper 2002). Digested solution was brought up to 50 mL with Milli-Q water (Milli-Q Advantage, Millipore, Billerica, MA, USA). The concentrations of 14 metals (boron, aluminium, chromium, manganese, nickel, copper, zinc, arsenic, selenium, molybdenum, cadmium, antimony, lead, and uranium) in drinking water and in the digested solutions of the food composite samples were determined using an inductively coupled plasma-mass spectrometer (ICP-MS; HP-4500, Agilent Technologies, Inc., Palo Alto, CA, USA). The instrumental parameters were as follows: RF power, 1,200 W; RF matching, 1.8 V; sample skimmer cone in Ni; plasma flow rate, 16 L/min; auxiliary flow rate, 1.1 L/min; nebulizer flow rate, 1.2 L/min. Gallium (m/z = 69) and yttrium (m/z = 89) were used as the internal standards.

The concentrations of magnesium, calcium, and iron in drinking water and in the digested solutions were determined by ICP-atomic emission spectrometry (ICPS-7510, Shimadzu Corp., Kyoto, Japan).

**Validation of measurement method**

To check the validity of the analysis, seven standard reference material (rice flour [SRM1568a], spinach leaves [SRM1570a], bovine muscle powder [SRM8414], oyster tissue [SRM1566b], nonfat milk powder [SRM1549], wheat flour [SRM1549a], and “typical diet” [SRM1548a]) were purchased from the National Institute of Standards and Technology (Gaithersburg, MD, USA). These standard reference materials were analyzed in the same manner as the food composite samples in this study and compared with the certified values of the standard reference materials.

The standard reference material–certified or reference values and our analyzed values were compared by the recovery ratio and z-score (Dolan & Caper 2002). For almost all metals, the recovery ratio was between 75 and 125% and/or the absolute value of the z-score was less than 2.5, which showed good agreement between the certified or reference values and our analyzed values. However, our analyzed values were outside of these ranges for selenium in the spinach leaves and the nonfat milk powder and for arsenic and molybdenum in the bovine muscle powder. The main cause of this disagreement may be that the certified or reference values are very low and near the detection limits of the present study. Because these values are very small, the effect of the disagreement was estimated to be negligible in the calculation of DMI in this study.

Detection limits were estimated to three times the standard deviation (SD) of the metal concentration derived from 10 measurements of the method blank. The method blank was a blank sample pretreated by the same procedure as for food composite samples.

**Estimation of DMI**

The National Nutrition Survey provides daily intake data for each food sample group (Figure 1). In the case of drinking water (14th group), the daily drinking water intake was assumed to be 2 L/day. To calculate DMI, the metal concentration in a food composite sample or drinking water was multiplied by the daily intake of each group, and the values were summed, as follows:

$$\sum_{i=1}^{14} a_i b_i$$

- $a_i$ = metal concentration in the food composite sample or drinking water of ith group
- $b_i$ = daily intake (consumption rate) of ith group

**Calculation of TDI and RDA values and their assumptions**

In this study, the values of TDI and RDA were calculated as for an average Japanese adult. Basically, the values of TDI (mg/kg/day) were derived from either the TDI reported in the *Guidelines for Drinking-water Quality* (WHO 2008) or from that proposed by JECFA (1982, 1983, 1989, 1993). TDI is not necessarily expressed as TDI but other similar terms, such as PTWI (provisional tolerable weekly intake) and PMTDI (provisional maximum tolerable daily intake). In this study, for simplification, the term “TDI” is used for all of these similar terms after weekly intakes were converted to daily intakes. Exceptions were the TDI values of manganese, molybdenum, and selenium.
TDI for manganese in the WHO guidelines is considered to apply only to drinking water, because it includes an uncertainty factor (UF) of 3 to take into consideration the possible increased bioavailability of manganese from water (WHO 2008). Therefore, the TDI of manganese was set to be 11 mg as daily intake per capita by excluding the UF of 3. This value was also applied as the upper limit for dietary reference intakes for Japanese (JMHLW 2005). For molybdenum, we applied the reference dose set for chronic oral exposure (0.005 mg/kg/day) by the U.S. Environmental Protection Agency (USEPA 1993). This value was also applied as the upper limit for Japanese (JMHLW 2005). For selenium, the no observed adverse effect level (NOAEL) of 4 μg/kg/day in humans was used directly for the derivation of the WHO guideline value (WHO 2003, 2008); this approach is the same as the TDI approach with a UF of 1. Therefore, we assume the NOAEL should be equivalent to TDI in the case of selenium. This value is more conservative than the upper limit for Japanese of 6.7 μg/kg/day (JMHLW 2005), which is based on a NOAEL of 13.3 μg/kg/day as determined by the study of Yang & Zhou (1994) and a UF of 2. To convert these TDI values to the unit of weight per capita, the average weight per capita was assumed to be 50 kg, which is the assumption used in establishing the drinking water quality standards in Japan.

The JMHLW (2005) provides separate RDA values for each gender and age group. In this study, the average of RDA values of both males and females from 18 to 69 years old was used as the RDA for an average Japanese person, except in the case of calcium and manganese. For these elements, JMHLW does not set a RDA but an adequate intake (AI) value. AI is defined as the amount of daily intake sufficient to maintain a stable nutritional state (JMHLW 2005). AI is set only when an RDA cannot be set because of the lack of experimental and epidemiological data. The values of TDI and RDA calculated in this study for an average Japanese person are shown in Table 1 with the results of DMI.

RESULTS AND DISCUSSION

Margin to TDI values for toxic metals

The mean and SD of DMI for the 17 elements, as well as the DMI of each food group, are presented in Table 1. The percentage of DMI to TDI was calculated for each of the 13 toxic metals (Figure 2). The mean DMIs of 10 toxic metals (excluding selenium, molybdenum, and arsenic) were less than 50% of TDI. A simple interpretation of this result indicates that there may be a substantial margin between TDI and mean DMI. Accordingly, for these 10 metals the total daily intake percentages allocated to drinking water could possibly be increased from the normal allocation percentage (i.e. 10–20% of TDI) in reviewing or establishing drinking water quality standards. In this case, however, only the mean intakes were considered. Variation of the intakes among individuals and areas should be further investigated and considered. Exposure from other possible sources such as air and soil should be considered as well. Furthermore, it may not be necessary to increase the allocation ratio if drinking water can meet the current standards with commonly-used drinking water treatment processes.

The six-city average of daily arsenic intake exceeded TDI by a considerable degree (280% of TDI); the highest intake was 359% of the TDI. The toxicity of arsenic varies greatly according to its chemical form (i.e. inorganic or organic). The toxicity of organic arsenic is much lower than that of inorganic arsenic. In this study, the chemical form was not considered, and the DMI of arsenic was calculated on the basis of the total arsenic. According to the analysis of daily arsenic intake (Table 1), among the 13 food groups two groups (“Other vegetables and seaweeds” and “Fish and shellfish”) accounted for high ratios. The sum of arsenic intake from the other 11 food groups was lower than the TDI; therefore, the chemical forms of arsenic in these two groups should be further investigated. The mean molybdenum intake was 99% of the TDI, and maximum ratio was 134%. The excess intake of molybdenum does not pose an immediate health hazard to humans, because the TDI represents a tolerable intake for a lifetime, and short-term exposure to levels exceeding the TDI is not a cause for concern (WHO 2008). Uncertainties exist in risk assessment of long-term exposure, however, including large UFs generally involved in establishing TDIs (WHO 2008) and the lack of quantitative risk information on intakes exceeding the TDI. These uncertainties are limitations in the current risk assessment approaches and should be improved in the future.

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<table>
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<td>Rice</td>
<td>0.1 ± 0.04</td>
<td>14 ± 4.1</td>
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<td>8.8 ± 2.5</td>
<td>197 ± 148</td>
<td>1.0 ± 0.2</td>
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<td>Cereals and potatoes</td>
<td>0.09 ± 0.02</td>
<td>30 ± 7.0</td>
<td>0.4 ± 0.1</td>
<td>41 ± 10</td>
<td>22 ± 11</td>
<td>0.5 ± 0.2</td>
<td>0.9 ± 0.2</td>
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<td>0.2 ± 0.03</td>
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<td>Sugar and sweets</td>
<td>0.04 ± 0.01</td>
<td>7.2 ± 1.6</td>
<td>0.4 ± 0.3</td>
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<td>21 ± 24</td>
<td>0.1 ± 0.04</td>
<td>0.2 ± 0.1</td>
<td>4.0 ± 2.2</td>
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<td>Oils and fats</td>
<td>0.003 ± 0.0001</td>
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<td>0.004 ± 0.01</td>
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<td>29 ± 7.9</td>
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<td>Legumes</td>
<td>0.2 ± 0.05</td>
<td>47 ± 12</td>
<td>0.1 ± 0.04</td>
<td>52 ± 15</td>
<td>21 ± 7.3</td>
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<td>Fruits</td>
<td>0.3 ± 0.03</td>
<td>13 ± 0.8</td>
<td>0.03 ± 0.02</td>
<td>14 ± 1.7</td>
<td>10 ± 7.9</td>
<td>0.2 ± 0.09</td>
<td>0.1 ± 0.03</td>
<td>6.2 ± 2.8</td>
<td>0.05 ± 0.01</td>
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<tr>
<td>Green and yellow vegetables</td>
<td>0.2 ± 0.02</td>
<td>23 ± 7.6</td>
<td>0.2 ± 0.2</td>
<td>34 ± 14</td>
<td>11 ± 10</td>
<td>0.2 ± 0.06</td>
<td>0.3 ± 0.04</td>
<td>5.8 ± 5.1</td>
<td>0.04 ± 0.01</td>
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<td>Other vegetables and seaweed</td>
<td>0.4 ± 0.2</td>
<td>30 ± 10</td>
<td>0.6 ± 0.5</td>
<td>65 ± 21</td>
<td>23 ± 12</td>
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<td>0.6 ± 0.3</td>
<td>10 ± 1.2</td>
<td>0.09 ± 0.03</td>
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<td>Alcohols and beverages</td>
<td>0.2 ± 0.1</td>
<td>20 ± 13</td>
<td>1.1 ± 1.1</td>
<td>18 ± 23</td>
<td>34 ± 19</td>
<td>0.8 ± 0.6</td>
<td>0.2 ± 0.1</td>
<td>21 ± 18</td>
<td>0.08 ± 0.06</td>
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<td>Fish and shellfish</td>
<td>0.06 ± 0.02</td>
<td>36 ± 13</td>
<td>0.2 ± 0.1</td>
<td>109 ± 48</td>
<td>44 ± 19</td>
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<td>0.6 ± 0.2</td>
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<td>Meat and poultry</td>
<td>0.05 ± 0.04</td>
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<td>0.1 ± 0.1</td>
<td>35 ± 20</td>
<td>64 ± 25</td>
<td>0.03 ± 0.005</td>
<td>1.7 ± 0.6</td>
<td>3.0 ± 4.7</td>
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<td>Milk products</td>
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<td>17 ± 5.2</td>
<td>0.02 ± 0.02</td>
<td>188 ± 90</td>
<td>39 ± 11</td>
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<td>Seasoning</td>
<td>0.2 ± 0.05</td>
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<td>0.2 ± 0.1</td>
<td>22 ± 7.7</td>
<td>51 ± 10</td>
<td>0.4 ± 0.1</td>
<td>0.5 ± 0.2</td>
<td>25 ± 2.8</td>
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<tr>
<td>Drinking water</td>
<td>0.07 ± 0.03</td>
<td>6.1 ± 4.1</td>
<td>0.08 ± 0.07</td>
<td>31 ± 17</td>
<td>0.2 ± 0.1</td>
<td>0.004 ± 0.005</td>
<td>0.14 ± 0.30</td>
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<td>0.005 ± 0.003</td>
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<tr>
<td>DMI</td>
<td>1.93 ± 0.22</td>
<td>295 ± 38.1</td>
<td>3.60 ± 1.37</td>
<td>631 ± 126</td>
<td>568 ± 194</td>
<td>4.15 ± 0.66</td>
<td>6.13 ± 1.11</td>
<td>156 ± 0.03</td>
<td>1.20 ± 0.17</td>
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**Table 1** Daily metal intake of each food group, average of six cities
Daily essential metal intakes relative to RDA

The percentages of DMI to RDA or AI were calculated for nine essential metals (Figure 3). The mean dietary intakes of magnesium, calcium, and iron were less than the RDA or AI. In terms of minimum daily intake among the six cities, the intakes of five metals (magnesium, calcium, manganese, iron, and zinc) were lower than the RDA or AI. Magnesium and calcium intakes were lower than the RDA or AI in five of the six cities. The differences between the RDA or AI and the intakes, however, were not very large, and the variation of the intake among cities and among years should be further investigated. As discussed for TDI, RDA and AI values also include many uncertainties that should be improved in the future, which limit our ability to discuss risks associated with deficiency of essential metals.

Iron intake was lower than the RDA in all six cities, and manganese and zinc intakes were lower in one city. Iron, manganese, and zinc are considered to be both essential and toxic metals. In terms of toxicity, the DMI values of these three metals were less than 50% of the TDI. Therefore, Japanese may consume more of these metals to satisfy the RDA or AI value.

Daily intakes of chromium, selenium, and molybdenum were far greater than the RDA values. In terms of toxicological aspects, the DMI values of selenium and molybdenum were near, or a slightly higher than, the TDI (Figure 2). Thus, it is not recommended that Japanese consume more selenium or molybdenum. For chromium, JMHLW (2005) does not provide a tolerable upper intake level, and the WHO does not describe the quantitative risk of chromium intake, although it has established a provisional drinking-water quality guideline value 0.05 mg/L for total chromium (WHO 2006).
Contribution of drinking water to TDI and/or RDA

For the 13 toxic metals analyzed, the contribution of drinking water to TDI was 2% or less in all six cities (Figure 4). Basically, in establishing the Japanese water quality standards, 10 of the TDI is allocated to intake via drinking water for most metals. The actual contribution is much less than 10% of the TDI, because the metal contents are controlled to be about 20–30% of the Japanese drinking water quality standard values in most water purification plants in Japan (Japan Water Works Association 2009).

The drinking water contribution to the RDA or AI of essential metals is less than 10% in all cities. On average, drinking water contributed only about 5% of the AI of calcium. Hardness in water is derived from calcium and magnesium. According to the WHO Guidelines for Drinking-water Quality, very soft waters may have an adverse effect on mineral balance and cardiovascular health (WHO 2008). The calcium concentration in drinking water is low in many cities in Japan (Japan Water Works Association 2009), and calcium compounds are added at some drinking water treatment plants where the hardness of the water is very low. This calcium addition was originally designed to prevent the corrosion of water pipes, but it may also help to reduce the health risks posed by very soft water. In general, the process increases the calcium concentration by ~8 mg/L (Japan Water Works Association 2009), which is equivalent to a daily intake of 16 mg/day or 2.3% of the AI of calcium. Therefore, this addition of calcium to drinking water does not help with calcium intake deficiency; it is better to consume calcium in foods such as milk.

CONCLUSION

The daily intakes of 17 metals in six cities in Japan were estimated by analyzing the concentrations of metals in locally obtained food composite samples and drinking water samples. The mean daily intake of 10 of the 13 toxic metals was less than 50% of TDI, except in the cases of arsenic, selenium, and molybdenum. The allocation ratio of intake to drinking water in establishing drinking water quality standards could possibly be increased from the normal allocation of 10–20% of TDI for these 10 metals. However, not only the mean intakes but also variation of the intakes among individuals and areas should be further investigated and considered. For the 13 toxic metals analyzed in this study, the contribution of drinking water to TDI was 2% or less in all six cities. For essential metals, the DMI values of magnesium, calcium, iron, manganese, and zinc were lower than RDA or AI in at least one city. The drinking water contribution to RDA or AI was less than 10% for all essential metals in all the six cities, indicating that drinking water did not contribute very much to essential metal intake.

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