Exposure to haloacetic acids via typical components of the Japanese diet and their allocations of drinking water ingestion to total exposure

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ABSTRACT

We conducted a national-scale survey on the haloacetic acids (HAAs) in the typical components of the Japanese diet. Combined with the findings of a previous study on multi-route HAA exposure, we estimated the actual relative contributions of drinking water ingestion to total HAA exposure and in this paper we discuss the necessary allocation factors for setting drinking water quality standard values of HAAs. The currently applied allocation (20%) was found to be unrealistically low and in need of appropriate adjustment. After determining the probability distribution of the relative contribution of each HAA, the rounded values corresponding to 0.05 and 0.1 cumulative probabilities were recommended for dichloroacetic acid (40%), trichloroacetic acid (30%), bromochloroacetic acid (30%) and bromodichloroacetic acid (60%) as their allocation factors. The direction of future investigations is discussed along with an overview of various sources of uncertainty. Ingestion exposure via diet and daily drinking water consumption were identified as priority factors.

Key words | allocation, drinking water quality standard value, haloacetic acids, multi-route exposure, relative contribution

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INTRODUCTION

Among the harmful chlorination by-products of drinking water, haloacetic acids (HAAs) have attracted extensive attention following trihalomethanes (THMs) as the second most frequently detected compounds (Singer *et al.* 1999; Richardson 2005). The best-known HAAs in drinking water are monochloroacetic acid (MCA), monobromoacetic acid (MBA), dichloroacetic acid (DCA), trichloroacetic acid (TCA), bromochloroacetic acid (BCA), bromodichloroacetic acid (BDCA), dibromoacetic acid (DBA), dibromochloroacetic acid (DBCA) and tribromoacetic acid (TBA). In Japan, MCA, DCA and TCA were first to be regulated by the national drinking water quality standards because more sufficient toxicological information is available for these compounds than for the other six species (Ministry of Health, Labour and Welfare Japan 2003).

As MCA and TCA are considered non-carcinogens (or non-genotoxic compounds), their standard values were derived by a threshold approach based on the tolerable daily intake (TDI) as shown in Equation (1). In contrast, DCA is regulated by a non-threshold approach based on the virtually safe dose (VSD) because of its genotoxicity. However, there is a different opinion on this point. DCA was classified as a possible human carcinogen by the International Agency for Research on Cancer (IARC) in 2002

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because of insufficient evidence of its carcinogenicity in experimental animals (IARC 2002). Therefore, the TDI approach may be taken if sufficient toxicological information on DCA is collected in the future. Similarly, the other six species of HAAs are currently not regulated because toxicological information on these compounds is very limited.

$$\text{Standard value (mg/L)} = \frac{\text{TDI } (\mu\text{g}/(\text{kg}\cdot\text{day})) \times \text{body weight (kg)} \times \text{allocation } (-)}{\text{Drinking} - \text{water consumption (L/day)} \times 1{,}000} \quad (1)$$

Allocation is a very important factor in the TDI approach, which reflects the proportion of TDI attributable to different exposure routes, such as ingestion, inhalation and transdermal exposure. This ensures that the total daily intake of each compound from all routes does not exceed the tolerable dose. However, because of the limited amount of information on multi-route exposure to HAAs, their allocation factors have not been accurately estimated, and a default value of 20% was applied in the establishment of the current standard values.

Itoh *et al.* (2008) conducted a survey to estimate multi-route (ingestion, inhalation and transdermal) exposure to HAAs in western Japan. The results indicate that because of the non-volatile property and long lag times (travel time of chemicals from skin permeation to blood circulation) of HAAs, inhalation and transdermal were minor routes of exposure. Ingestion exposure via the diet was the most important exposure route. Its contribution to the total exposure was more than 50%.

The importance of ingestion exposure via the diet has also been supported by other studies performed in other countries. Schroll (1994) and Sutinen *et al.* (1995) reported that TCA may be assimilated by vegetables via their roots or leaves by uptake from the soil and air. MCA and TCA were actually detected in a limited number of dietary component samples (Reimann *et al.* 1996). HAAs have been found in foods disinfected with chlorine during production and processing (USEPA 1994). In addition, Krasner and Wright (2005) reported that increased mono- and di-halogenated acetic acid formation and degradation of tri-halogenated acetic acids occur during boiling of chlorinated tap water. These observations suggest that HAAs can be formed during the cooking of food using tap water under real-world conditions. However, none of these studies gave a comprehensive conclusion on HAA ingestion exposure via diet. Furthermore, the only domestic study (Itoh *et al.* 2008) did not take the spatial variation of HAA ingestion exposure into consideration (i.e. the survey was only conducted in one sampling spot). Thus, investigation of HAA ingestion exposure via the diet with consideration of spatial variation is crucial. Therefore, in this study, a survey on HAA concentrations in the typical Japanese diet was performed over a wide sampling scale. Based on the results, the relative contribution of drinking water ingestion to total HAA exposure was estimated. The legitimacy of the current default allocation factor was then discussed.

Also, uncertainty analysis was conducted to identify important factors in estimating the allocations of DCA and TCA to drinking water.

METHODS

Survey protocol

For wider sample scale, market-basket surveys were conducted in six regions of Japan (Regions A–F, located from latitudes 24.3° to 35.7° N), with populations ranging from nearly 1 million to 14 million, between June and October 2008. Since our strategy was to focus on spatial variation over the whole country instead of a specific region, only one survey was conducted for each region. Following the explanation of the National Nutrition Survey (Ministry of Health, Labour and Welfare Japan 2004), foods were randomly purchased to represent the 13 groups of typical dietary classification as shown in Table 1. In four of the sampling regions (A–D), raw foods in each food group were prepared with local tap water and blended according to their intake proportions. All of the homogenized samples were stored as 13 groups at −20 °C and shipped from each region to our laboratory for analysis. Samples from the other two regions (E and F) were similarly prepared, blended, stored and sent to our laboratory except that cooking was done in reagent water.

Table 1 | Dietary groups and their daily intake amounts

Dietary groups	Daily intake amounts (g)
Grain	343
Potato	167
Sweetener and snacks	7
Lipid	11
Bean	62
Fruit	119
Vegetable	254
Seaweed	13
Beverage	616
Seafood	83
Meat and egg	112
Dairy	135
Seasoning	92

Source: Ministry of Health, Labour and Welfare Japan (2004).

Extraction and analysis of HAAs

Reagent water was purified with a Millipore Academic-A10 purification system (Millipore, Tokyo, Japan). HAA standards (water analysis grade) were from Kanto Chemical (Tokyo, Japan). All other reagents (water analysis grade) were purchased from Wako Pure Chemical Industries (Osaka, Japan). HAAs in dietary components were extracted with methyl tert-butyl ether (MTBE) based on the procedure developed by Raymer *et al.* (2000). HAAs extracted in the MTBE phase were converted to their methyl esters in acidic methanol followed by heating for 2 h.

As the sample matrixes were much more complex than drinking water, HAAs (methyl esters) were analysed and quantified with internal standard (IS) calibration (1,2,3-trichloropropane) by gas chromatography/mass spectrometry (GC/MS) (GCMS-QP2010 Plus; Shimadzu, Kyoto, Japan) with a J&W DB-5MS capillary column (30 m × 0.32 mm × 0.25 μm film; Agilent Technologies, Santa Clara, CA) for separation. The GC/MS parameters included an injection temperature of 230 °C and a detection temperature of 200 °C. The initial oven temperature was 40 °C, which was held constant for 6 min before being ramped at 2.5 °C/min to 65 °C, and to a final oven temperature of 205 °C at 20 °C/min. The flow rate of carrier gas (helium) was 2.04 mL/min. The quantification and monitoring ions for the HAA esters are shown in Table 2. HAA extraction recoveries of each dietary sample were determined by spiking appropriate amounts of HAAs into dietary samples and conducting the entire extraction and quantification procedure. The measured HAA concentrations were corrected based on their recoveries. The method quantification limit (MQL) of each HAA was defined as the minimum HAA concentration in MTBE that gave a signal-to-noise ratio of 10 (Table 2). Concentrations below the MQL (ND) were calculated as zero in the following exposure assessment.

Exposure assessment and estimation of allocations of HAAs

The dietary intake values for each food group obtained from the literature were applied in the ingestion exposure assessment using Equation (2) (Ministry of Health, Labour and Welfare Japan 2004). The regional variation in the diet intake amounts among the six target regions was not considered. The HAA concentrations in Equation (2) were those detected from local diet. After the HAA ingestion exposures via diet of each region were obtained, their average values were calculated as the national representative values to derive the relative contribution of each HAA. Exposure levels via other routes were obtained from a

Table 2 | HAA ester quantification conditions and method performance

Compounds	Quantification ions	Monitoring ions	Retention times (min)	MQLs (μg/L)
MCA	49	77, 108	3.00	10
MBA	59	121, 152	4.60	2
DCA	59	83, 85	4.92	1
TCA	59	117, 119	8.64	1
BCA	59	127, 129	9.20	2
Internal standard[a]	75	110	9.50	NA[b]
DBA	59	173, 109	15.3	1
BDCA	59	161, 163	15.7	2
DBCA	59	207, 209	21.4	5
TBA	59	251, 253	23.3	10

[a]1,2,3-trichloropropane.
[b]Not applicable.

previous report (Itoh *et al.* 2008). A total of 22 subjects participated in this previous study. To derive the individual total exposure and relative contribution of drinking water (Equation (3)), tap water ingestion, inhalation and transdermal exposure levels in each of the 22 subjects were combined with the average values of dietary ingestion exposure determined above.

$$\text{Ingestion exposure via diet of each region } (\mu g/\text{day}) = \sum(\text{HAA concentration in local diet } (ng/g) \times \text{intake } (g/\text{day}))/1{,}000 \quad (2)$$

$$\text{Relative contribution of drinking water ingestion to total exposure } (-) = \text{ingestion exposure via drinking water } (\mu g/\text{day})/\text{total exposure } (\mu g/\text{day}) \quad (3)$$

The present study was performed to determine how the allocation factors of HAAs should be established. Attempts to answer this question should focus on investigating whether the current default (20%) is appropriate, and finding an alternative(s) if it is deemed inappropriate. Therefore, it is necessary to approximate the probability distributions of the relative attributions of each HAA. Then, it would be possible to translate between any given pair of cumulative probabilities and the corresponding relative contribution in the approximated distributions. Finally, an appropriate allocation factor can be determined by the acceptable cumulative probability (5 percentiles in this study) for each HAA. Building histograms of the relative contributions, approximating the probability distributions to the histograms, and measuring the goodness of the approximations were performed using Crystal Ball Version 11 (Oracle, Redwood Shores, CA).

RESULTS AND DISCUSSION

HAA concentrations and ingestion exposure via the diet

The HAA extraction recoveries varied between the dietary samples (e.g. DCA extraction recoveries were 0.64 in grain and 0.96 in beverage samples). However, the relative standard deviations in all cases were less than 30%. Therefore, measured HAA concentrations were corrected based on the determined recoveries. In all the dietary samples from Regions A–D, DCA (0–18.7 ng/g) and BCA (0–60.9 ng/g) were detected most frequently (DCA and BCA were detected from 27 and 12 diet samples, respectively, from all 52 samples (13 samples from each region)). TCA was detected in a lower concentration range (0–17.6 ng/g) but more frequently (24 samples) compared with BCA (Figure 1). The others were either detected from specific samples (BDCA and MBA in the lipid group and DBCA in the lipid and seaweed groups) or not detected at all (MCA, DBA and TBA).

In dietary samples of Regions E and F, DCA (0–7.97 ng/g) and BCA (0–7.58 ng/g) were present in slightly lower concentrations than in Regions A–D. TCA was detected in a lower range (0–12.1 ng/g) but a higher median value (4.88 ng/g) than in Regions A–D (0.03 ng/g) (Figure 2).

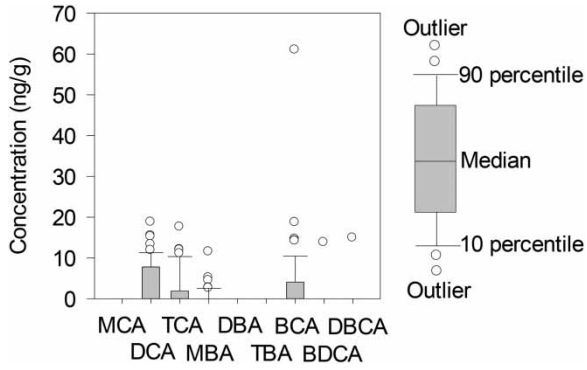

Figure 1 | HAA concentrations in dietary samples (Regions A–D).

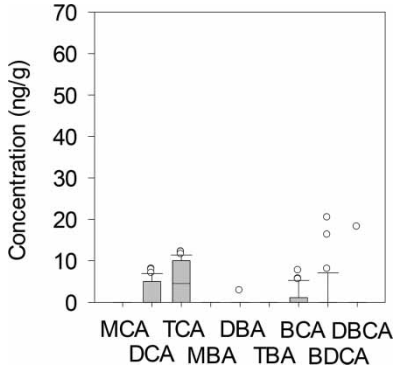

Figure 2 | HAA concentrations in dietary samples (Regions E and F).

There are several reasons for these observations: extra HAAs were not formed because of a lack of chlorine in the cooking water of Regions E and F; higher TCA concentration in raw food of Regions E and F; and the degradation of HAAs with higher molecular weight during heating. However, the strategy of this study is to involve spatial variation in the estimation of ingestion exposure to HAAs but not to find the change mechanism for them. Therefore, differences of HAA concentrations between Regions A–D and E and F were not further investigated. Spatial variation will be consequently discussed in this paper as one of the uncertainties in the estimation procedure.

Ingestion exposures of each HAA were determined by summing their ingestion via each food group (Table 3). Hereby, the average values of HAA ingestion exposure were applied in the following estimation of the relative contributions. The exposure level of DCA was the highest (15.6 μg/day), followed by TCA (12.2 μg/day) and BCA (11.6 μg/day). For these three species, beverage (1.1–9.35 μg/day) and vegetable (1.97–4.74 μg/day) groups were identified as the major media of exposure. The higher exposure levels were mainly because of their higher daily intakes (616 and 254 g/day, respectively) compared with the other groups.

Relative contribution of drinking water ingestion to total HAA exposure

The estimated relative contributions for all 22 subjects and their observed frequency histograms are shown in Figure 3. As the inhalation and transdermal exposures of MCA, MBA and TBA were not estimated in the previous study (Itoh *et al.* 2008), their relative contributions could not be calculated here.

Probability distributions were approximated to the histograms by the goodness-of-fit (GoF) test function using Oracle Crystal Ball software. On selecting the appropriate distributions, more attention was paid to the Anderson–Darling (A–D) test results than other GoF tests (e.g. χ^2 test and Kolmogorov–Smirnov test) because it is more sensitive than other GoF tests in the tails rather than the mid-range of the probability distribution (USEPA 2001; Alqam *et al.* 2002). With respect to the decision regarding drinking water quality standard values, because a smaller allocation factor leads directly to stricter regulation (a smaller standard value), the lower tail of the cumulative distribution is of greatest concern. Therefore, the A–D test should be particularly useful.

The probability distributions chosen for each HAA are summarized in Table 4. The A–D values indicated that the chosen probability distributions gave satisfactory approximations to the frequency histograms of the relative contributions, except DBA (A–D 2.38) and DBCA (A–D 4.16) (a value less than 1.5 generally indicates a good fit). Therefore, DBA and DBCA were excluded from the following discussion of identifying allocation factors using this method.

Each HAA has a different type of probability distribution. Therefore, it is necessary to seek alternatives. After integrating the probability distributions, the cumulative probabilities and their corresponding relative contributions were determined as shown in Table 5 and Figure 4, along with that of the current default allocation (0.2). It should be noted that the four species of HAA except MCA and TCA do not necessarily require allocation factors as they are not regulated or not included in the TDI approach, as explained above. However, their estimated relative contributions could be used in future reconsideration or establishment of new standard values.

In the case of DCA, the currently applied allocation factor (0.2) corresponds with a cumulative probability of 0.001. For TCA, BCA and BDCA, the allocation factor virtually corresponds to a cumulative probability of zero.

Table 3 | HAA ingestion exposures via dietary intake (μg/day)

	MCA	DCA	TCA	MBA	DBA	TBA	BCA	BDCA	DBCA
Maximum	0.00	15.6	12.2	2.16	0.03	0.00	11.6	4.84	0.19
Minimum	0.00	0.89	0.00	0.00	0.00	0.00	1.98	0.00	0.00
Average	0.00	6.49	6.55	0.43	0.00	0.00	4.70	0.83	0.06

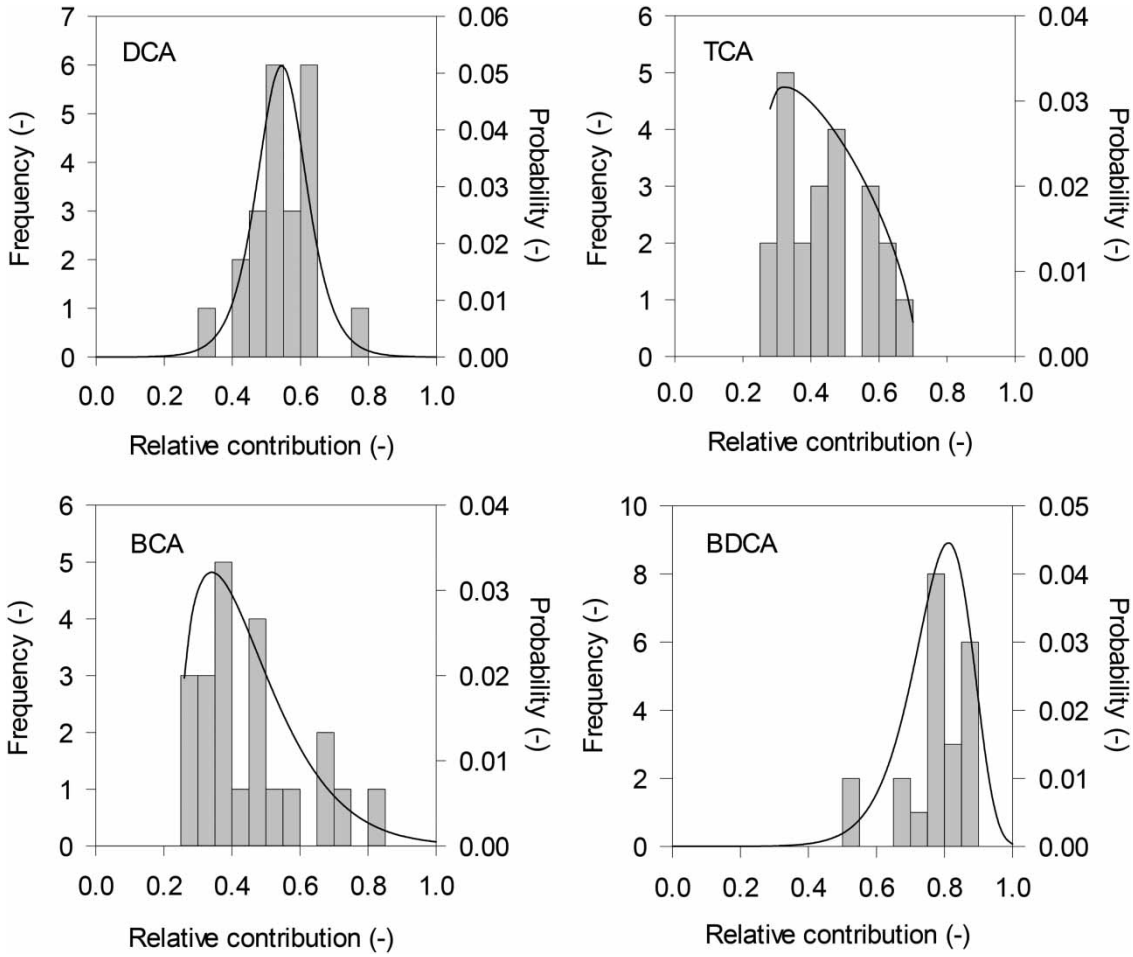

Figure 3 | Histograms of the estimated relative contribution of drinking water ingestion to total HAA exposure and their goodness-of-fit test results.

Table 4 | Goodness-of-fit test results of the relative contributions of HAAs

Compounds	Probability distributions	Parameters	A–D values
DCA	Logistic	Average = 54.5, Scale = 4.87	0.159
TCA	Beta	Min = 27.0, Max = 71.1, α = 1.09, β = 1.63	0.149
BCA	Weibull	Location = 24.5, Scale = 22.9, Shape = 1.41	0.193
BDCA	Weibull	Location = −42.3, Scale = 124, Shape = 15	0.551
DBA	Logistic	Average = 89.3, Scale = 5.43	2.376
DBCA	Logistic	Average = 79.3, Scale = 19.7	4.163

Thus, an allocation factor greater than 0.2 is acceptable to more than 99.9% of the studied population. This implies that 0.2 is unnecessarily low and should be increased. In addition, making decisions regarding public health issues, such as setting an allocation factor, should exclude the outliers, and 0.05 and 0.1 would become the probabilities of interest. In the case of DCA, as the 0.05 and 0.1 cumulative probabilities correspond to relative contributions of 0.402 and 0.438, respectively, a rounded value of 40% is recommended as the allocation factor. Based on the same consideration, the allocation factors of TCA, BCA and BDCA were recommended as 30, 30 and 60%, respectively (Table 6). Although raising the allocation factor leads to higher standard values (i.e. looser regulation on drinking water quality) according to Equation (1), these recommendations have a sound scientific rationale. Although allocations of MCA, MBA and TBA were not estimated because of their low concentrations in the environment, a much wider survey should be applied to obtain information

Table 5 | Relative contributions and cumulative probabilities of interest

Compounds	Relative contributions (–)	Cumulative probabilities (–)
DCA	0.20	0.001
	0.402	0.050
	0.438	0.100
TCA	0.20	0.000
	0.287	0.050
	0.303	0.100
BCA	0.20	0.000
	0.273	0.050
	0.292	0.100
BDCA	0.20	0.000
	0.597	0.050
	0.648	0.100
DBA[a]	0.20	0.000
	0.733	0.050
	0.774	0.100
DBCA[a]	0.20	0.047
	0.212	0.050
	0.360	0.100

[a]Excluded due to poor approximation (A–D values >1.5).

on these compounds. In conclusion, the allocation factors of DCA, TCA, BCA and BDCA are recommended based on the distributions of their relative contribution in drinking water ingestion to total exposure. The current allocation factors should be adjusted to 30% (TCA, BCA), 40% (DCA) and 60% (BDCA).

Uncertainty analysis in the estimation of DCA and TCA

The calculation of relative contributions involved a number of uncertainties, such as the use of average dietary ingestion exposure values for DCA and TCA. The effects of these uncertainties could be reduced by further individual investigations, because they do not have equal influences on the estimates. Therefore, it is necessary to rank their priority and to conduct purpose-designed investigations accordingly. In this subsection, the following factors that may significantly contribute to the uncertainties in the estimation of the allocations of DCA and TCA to drinking water are discussed.

Spatial variation of ingestion exposure via diet

Using the average ingestion exposure via the diet could influence the estimation. Table 7 shows the calculation results of the relative contributions (0.05 cumulative probability) based on the maximum and minimum exposure levels. In the case of DCA, the maximum and minimum ingestion exposures not only led to different results (0.220 and 0.707, respectively), they also varied markedly from the originally estimated value (0.402). Similar results were also found for TCA. Thus, ingestion exposure via the diet is an important uncertainty factor. Therefore, further investigations of ingestion exposure via the diet over a wider scale with larger numbers of subjects are necessary.

Daily drinking water consumption

In the present study, as daily drinking water consumption was not investigated in each subject, the currently applied value of 2 L was used for evaluation of ingestion exposure. However, there have been previous reports on this issue in Japan. Yano *et al.* (2000) reported an arithmetic mean value of 209.2 mL/day for direct drinking water consumption based on a questionnaire survey. Song (2011) re-estimated the survey results and concluded that the daily direct drinking water consumption could be described as a Weibull distribution (location = 0, scale = 0.33, and shape, Weibull slope = 0.895) with an average value of 321 mL/day. Furthermore, Okashita (2010) estimated that daily indirect tap water consumption via the diet (as cooking matrix) is 732 mL. Therefore, a daily drinking water consumption level of 1.053 L (321 + 732 mL) was used here to examine its influence on the allocations. As shown in Table 7, smaller allocations of DCA (0.252) and TCA (0.175) were obtained compared with their originally estimated values (0.402 and 0.287, respectively). As the daily drinking water consumption could vary between individuals and seasons, further investigations of this issue are required.

Choice of probability density function

There are other choices of probability distribution in the A–D test results. As selecting different functions can lead to different estimates of allocations, this also introduces uncertainty. Here, the second options (beta and Weibull distributions for DCA and TCA, respectively) were compared with the original results. As shown in Table 7,

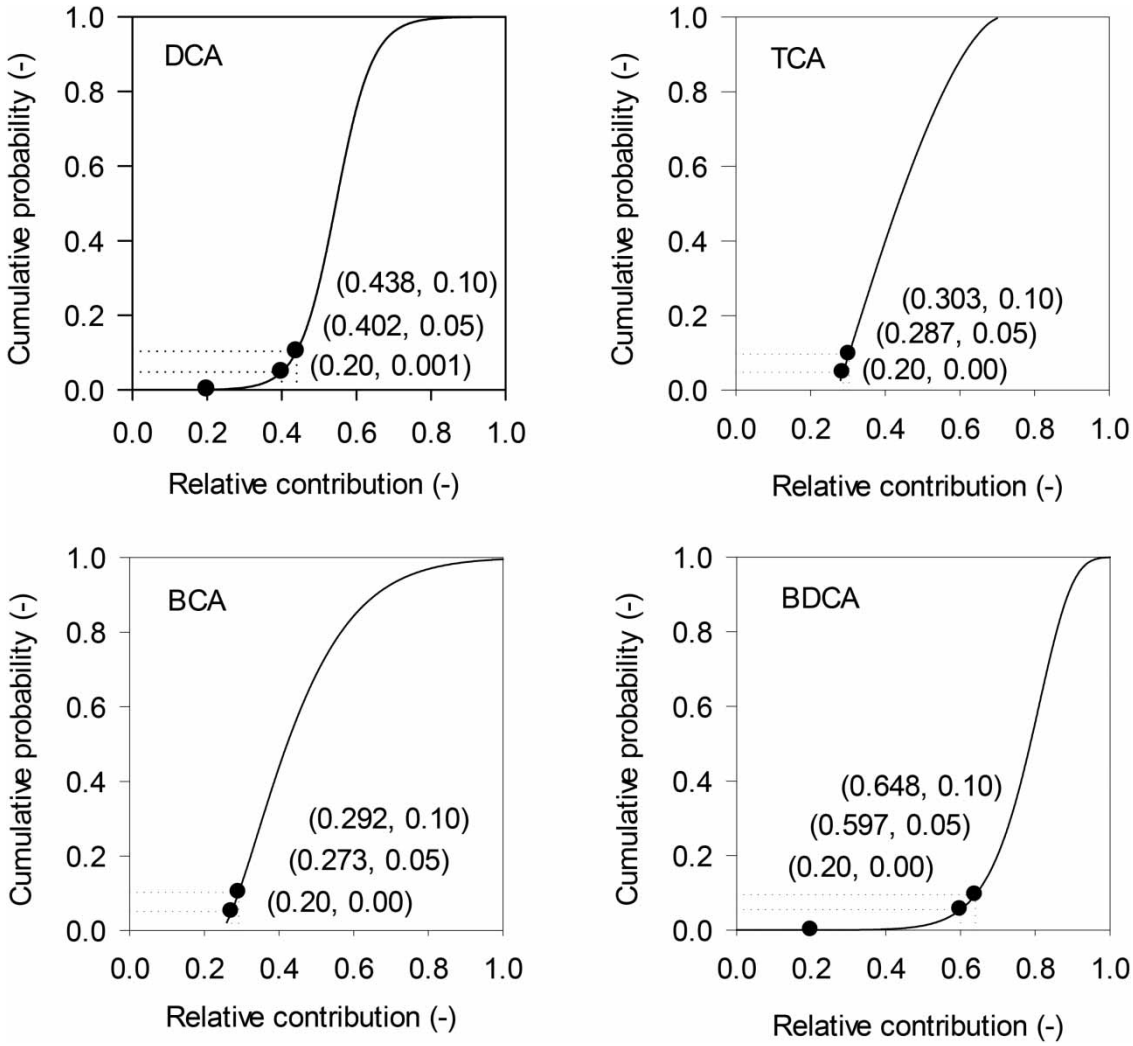

Figure 4 | Cumulative probabilities of the estimated relative contribution of drinking water ingestion to total HAA exposure.

Table 6 | Recommended allocation factors of HAAs

Compounds	Allocation factors (%)
DCA	40
TCA	30
BCA	30
BDCA	60

there were no marked differences between the two distributions. Therefore, the choice of probability density function does not appear to be an important source of uncertainty.

Relevant parameters applied in inhalation and transdermal exposure

In the previous study of Itoh *et al.* (2008), an average breathing frequency of 15 m^3/day was universally applied in inhalation exposure assessment. Therefore, individual differences among subjects were not considered. Yasutaka & Matsuda (2007) provided a relationship between breathing frequency and body weight to reduce the uncertainty. In addition, using average occupation times in indoor and outdoor environments results in similar uncertainty. Furthermore, in the transdermal exposure assessment, because of a lack of information on skin permeability coefficient and lag time of

Table 7 | Summary of uncertainty analysis results

Uncertainty sources		Relative contribution of DCA (–)	Relative contribution of TCA (–)
Originally estimated values without consideration of uncertainty sources		0.402[a]	0.287[a]
Ingestion exposure via diet	Max	0.220	0.186
	Min	0.707	0.876
Probability density function	Beta	0.399	–
	Weibull	–	0.292
New daily drinking-water consumption	–	0.252	0.175

[a]0.05 cumulative probability.

BDCA and DBCA, those of DBA were applied because of the similar molecular weights of these compounds, and this also introduces uncertainty into the estimation. However, because inhalation of and transdermal exposure to HAAs contribute much less to the total exposure than ingestion, they are not considered as important sources of uncertainty.

Different uncertainties have different effects on the estimation results. In setting drinking water quality standards of HAAs, further efforts to determine the ingestion exposure via the diet and daily tap water consumption should be given higher priorities.

CONCLUSIONS

Together with the results of previous studies, the allocation factors of HAAs were discussed and recommendations were made based on the probability distributions of the relative contributions of drinking water ingestion to total exposure. The currently applied default value of 20% was found to be unrealistically low and needs to be adjusted for each HAA. The rounded relative contributions corresponding to 0.05 and 0.1 cumulative probabilities were recommended as the allocation factors for DCA (40%), TCA (30%), BCA (30%) and BDCA (60%). Advice on future directions of study was provided based on discussion and overview of various uncertainties. Ingestion exposure via the diet and daily drinking water consumption were shown to be priority factors for further studies.

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