National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Derivation of a water-based quality standard for secondary poisoning of mercury

RIVM Letter report 2015-0058
E.M.J. Verbruggen | R. van Herwijnen | C.E. Smit

National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Derivation of a water-based quality standard for secondary poisoning of mercury

RIVM Letter report 2015-0058
E.M.J. Verbruggen | R. van Herwijnen | C.E. Smit

Colophon

© RIVM 2015
Parts of this publication may be reproduced, provided acknowledgement is given to: National Institute for Public Health and the Environment, along with the title and year of publication.

E.M.J. Verbruggen (author), RIVM
R. van Herwijnen (author), RIVM
C.E. Smit (author), RIVM

Contact:
Eric Verbruggen
Centre for Safety of Substances and Products
eric.verbruggen@rivm.nl

This investigation has been performed by order and for the account of the Ministry of Infrastructure and the Environment, within the framework of the project "Chemical water quality, standard setting and Priority Substances Directive".

Published by:
National Institute for Public Health and the Environment
P.O. Box 1│3720 BA Bilthoven
The Netherlands
www.rivm.nl/en

Publiekssamenvatting

Afleiding van een waterkwaliteitsnorm voor kwik op basis van doorvergiftiging

Het RIVM doet een voorstel voor een nieuwe, Nederlandse waterkwaliteitsnorm voor kwik. Deze norm houdt rekening met de mate waarin kwik zich ophoopt in visetende dieren, en beschermt daardoor ook vogels en zoogdieren. De bestaande Europese norm voor kwik in oppervlaktewater gaat alleen over het acute directe effect van kwik op waterorganismen zonder rekening te houden met de stapeling in de voedselketen. Deze waternorm is niet laag genoeg om visetende vogels en zoogdieren te beschermen.

Van kwik is algemeen bekend dat het wereldwijd een probleem is. Het komt onder meer vrij bij de verbranding van steenkool. Kwik is opgenomen op de lijst van prioritair gevaarlijke stoffen onder de Kaderrichtlijn Water. Dit betekent dat de uitstoot naar het milieu moet worden voorkomen.

Naast de Europese norm voor oppervlaktewater, is er een Europese norm die een maximum stelt aan de hoeveelheid kwik in vis, de zogeheten biotanorm. Deze norm moet voorkomen dat visetende roofvogels en zoogdieren te veel kwik binnenkrijgen via het voedsel dat ze eten. De biotanorm is het gehalte van kwik in vis waarbij vogels en zoogdieren via hun voeding geen extra risico lopen.

Lidstaten moeten aantonen dat kwikgehalten in vis niet worden overschreden, maar mogen zelf bepalen hoe ze dat meten. Nederland geeft er de voorkeur aan om niet in vis, maar in water te meten. Daarom is berekend bij welke concentratie in water de biotanorm voor vis niet wordt overschreden. De berekende veilige concentratie in water is 0,07 nanogram opgelost kwik per liter. Deze norm is aanzienlijk strenger dan de norm voor de directe effecten op waterorganismen, die tot nu toe in Nederland is gebruikt. Het ministerie van Infrastructuur en Milieu (I&M) is van plan de voorgestelde norm dit jaar in de nieuwe wetgeving op te nemen.

Kernwoorden: kwik; doorvergiftiging; waterkwaliteitsnorm; Kaderrichtlijn Water

Synopsis

Derivation of a water-based quality standard for secondary poisoning of mercury

RIVM proposes a new water quality standard for mercury in Dutch surface waters. The standard protects fish eating animals by taking secondary poisoning into account. The current European standard for mercury in surface waters is based on acute direct effects on waterorganisms. Because accumulation in the food chain is not included, fish eating animals are not sufficiently protected.

Mercury is known for its worldwide environmental impact. Burning of charcoal is one of the emission routes. Mercury is listed as a priority hazardous susbtance under the Water Framework Directive, which means that environmental emissions should be prevented.

Next to the European surface water standard, a European biota standard has been set that limits the concentration of mercury in fish. This standard should protect predatory birds and mammals from adverse effects of mercury due to food intake.

European member states have to prove that mercury levels is fish are not exceeded, but can choose an alternative matrix. In the Netherlands there is a preference to monitor water instead of fish. Therefore, the biota standard has been converted into a water-based equivalent that offers adequate protection. The resulting value is 0.07 nanogram per liter, expressed as a dissolved concentration. This value is considerably lower than the standard for direct effects that has been used so far in the Netherlands. The ministry of Infrastructure and the Environment plans to include the proposed standard into new legislation this year.

Keywords: mercury; secondary poisoning; water quality standard; Water Framework Directive

Contents

Summary — 9

1 Introduction — 11
1.1 Water quality standards under the Water Framework Directive — 11
1.2 European biota standard for mercury — 11
1.3 Aim of this report: derivation of water-based risk limits — 12
1.4 Reader's guide — 12

2 Methodology: deriving EQS for bioaccumulating compounds — 13
2.1 General approach — 13
2.2 Uncertainty about published BAFs — 16
2.3 Determination of the BAF for mercury in relation to trophic level — 17

3 Results and discussion — 19
3.1 Summary of bioaccumulation field studies — 19
3.2 Correlation between water characteristics and bioaccumulation — 32
3.3 Correlation between BAF and trophic level — 34
3.4 Derivation of ERLs for mercury in water — 36
3.5 Implications of the new standards — 37

4 Summary and conclusions — 39

Acknowledgements — 41

References — 43

Summary

In this report, RIVM proposes a new water quality standard for mercury in Dutch surface waters within the context of the Water Framework Directive. The standard protects fish eating animals by taking secondary poisoning into account. The current European standard for mercury in surface waters is based on acute direct effects on waterorganisms. Because accumulation in the food chain is not included, fish eating animals are not sufficiently protected when only using this value.

For the protection of fish eating birds and mammals, a European biota standard for mercury is set at 20 µg/kg_{wwt} in Directive 2013/39/EU. This value represents a concentration in fish at which birds and mammals are protected against effects of mercury via secondary poisoning. However, compliance checking by means of monitoring in water has advantages over biota sampling in terms of reproducibility, costs and uniformity of sampling. Therefore, the biota standard for mercury has been converted into a water-based quality standard that offers the same level of protection.

For this, the relationship between concentrations in water and biota was investigated in this report. Bioaccumulation factors (BAFs) were derived for fish representing different trophic levels. The data show that bioaccumulation is positively correlated with trophic position. This means that small fish accumulate less mercury than organisms higher in the food chain. This correlation was used to establish BAFs for larger fish that are eaten by marine and freshwater predators and humans.

Log BAF-values based on dissolved total mercury and methylmercury are 5.47 and 6.69, respectively. Using these values, water-based quality standards are proposed of 0.07 ng/L for the sum of all dissolved mercury species (total mercury in filtered samples), and 0.004 ng/L for dissolved methylmercury.

The biota standard is exceeded in over 90% of the fish samples included in the present evaluation. Similarly, monitoring data of Dutch surface waters indicate that the proposed water quality standards will likely also be exceeded frequently.

1 Introduction

1.1 Water quality standards under the Water Framework Directive

The European Water Framework Directive 2000/60/EC (WFD) aims at "maintaining and improving the aquatic environment in the Community". Member States should achieve the objective of at least a "good ecological status" and a "good chemical status" by defining and implementing the necessary measures within integrated programs of measures. For a good chemical status the WFD requires that environmental quality standards (EQSs) are met. These EQSs serve as a benchmark to decide whether or not specific measures are required. The EQSs for priority (hazardous) substances are set on a European community level. For other compounds that are relevant to individual member states, standards are set on a national level.

The EQS for chronic exposure is aimed at the protection of ecosystems and human health. The derivation considers direct ecotoxicity to aquatic organisms, exposure of humans through consumption of fish and fishery products, and exposure of predators through secondary poisoning. The most critical of these routes determines the final standard. For compounds that have a strong potential to bioaccumulate in fish, human fish consumption and secondary poisoning routes are often most critical. Due to the characteristics of these compounds, concentrations increase along the food chain. Consumption of fish therefore leads to critical levels in humans or predators while at similar concentrations in water, aquatic organisms are not affected. For these compounds, concentrations in fish have been derived that will not cause adverse effects in humans or predatory birds and mammals upon lifetime consumption.

1.2 European biota standard for mercury

Also for the priority hazardous substance mercury, secondary poisoning is most critical, because of the high level of bioconcentration. According to the preamble of Directive 2008/105/EC (EC, 2008), EU community level EQSs based on surface water concentrations are sufficient for the majority of substances. An EQS based on surface water concentrations of 0.07 µg/L was set for mercury and its compounds. However, it was considered appropriate to establish EQSs for biota at the EU community level, because for this substance "it is not possible to ensure protection against indirect effects and secondary poisoning at Community level by EQS for surface water alone". A maximum concentration in biota for mercury of 20 $\mu g/kg_{wwt}$, expressed as total mercury (THg), was set in Art 3(2) of Directive 2008/105/EC, based on a substance data sheet that was compiled in 2005 (EC, 2005). The biota standard is based on the toxicity of mercury to birds and mammals. For human exposure via fish, the biota standard was set to 500 $\mu g/kg_{wwt}$ based on the European legal food limit for fish as laid down in Commission Regulation (EC) 1881/2006 (and its predecessor Commission Regulation 466/2001). The reason for setting standards based on concentrations in biota rather than concentrations in the water column was primarily the uncertainty surrounding bioconcentration and biomagnification factors. According to

Directive 2008/105/EC, if member states do not apply standards for biota they shall establish equal or stricter quality standards for water than those in the daughter directive, in order to achieve the same level of protection as the standards for biota.

The biota standard of 20 µg/kg_{wwt} is maintained in the new priority substances Directive 2013/39/EU (EU, 2013). The motivation for setting a biota standard is phrased differently and focuses on the analytical challenges when setting water-based standards for biota: "Some very hydrophobic substances accumulate in biota and are hardly detectable in water even using the most advanced analytical techniques. For such substances, EQS should be set for biota."
Similar to the previous directive, the option is given to apply alternative standards when stating: "Nevertheless, in order to take advantage of their monitoring strategy and adapt it to their local circumstances, Member States should have flexibility to apply an EQS for an alternative matrix."

1.3 Aim of this report: derivation of water-based risk limits

In the Netherlands, measuring water samples is preferred over biota monitoring. One of the arguments that is often used to promote biota monitoring is that the conversion of biota standards to water concentrations is uncertain because of the variation in accumulation between organisms. However, this variation will also be reflected in biota concentrations and the outcome of the biota monitoring will largely depend on the species that is sampled, its life-stage and home range, and the time and place sampling. This variation is hard to quantifiy without extensive sampling (Moermond and Verbruggen, 2012). Therefore, a better option is to address the variation in bioaccumulation by a thorough evaluation of bioaccumulation data and use this information when converting the biota standard into a single water-based value. The responsible ministry in the Netherlands therefore decided to investigate the possibility to rely on water-based quality standards for mercury and requested RIVM to propose water-based quality standards for these compounds. The methodology to convert biota standards into corresponding water concentrations is included in the European Technical Guidance For Deriving Environmental Quality Standards (EC, 2011).

1.4 Reader's guide

This report describes the derivation of an alternative quality standard for water based on a thorough evaluation of the relationship between mercury concentrations in water and accumulation in biota. Chapter 2 gives the theoretical background and outlines the methodoloy used. In Chapter 3, a summary is given of relevant literature and data are discussed and processed to derive water-based standards for mercury. The conclusions can be found in Chapter 4.

2 Methodology: deriving EQS for bioaccumulating compounds

2.1 General approach

The methodology for the derivation of EQSs for water is described in detail in the European Technical Guidance For Deriving Environmental Quality Standards (EC, 2011), further referred to as TGD-EQS. Starting point for the assessment is the quality standard for predatory birds or mammals, expressed as a concentration in fish ($QS_{biota,\ secpois}$). The $QS_{biota,\ secpois}$ was derived in 2005 as 20 µg/kg_{wwt}, expressed as total mercury (THg) based on chronic toxicity data for birds and mammals. Starting from the biota standard, corresponding water concentrations can be calculated. The biota standards as defined in the priority substances directive apply to large fish that are consumed by humans or freshwater predators, such as cormorants or otters. This $QS_{biota,\ secpois}$ aims to protect these predators by setting a limit for their food, which is 1 trophic level below this predator. For freshwater ecosystems, assuming the trophic level (TL) for algae, zooplankton, small fish and large fish are 1, 2, 3, and 4, respectively, the $QS_{biota,\ secpois}$ is set on TL4 to protect the birds and mammals at TL5.
Figure 1 depicts the relationships between water and biota at different trophic positions. Concentrations in TL4-fish depend on the accumulation of substances from the aqueous phase by lower aquatic organisms (bioconcentration) and accumulation in the food chain from TL1-3 to TL4 (biomagnification). These processes are represented by a bioconcentration factor (BCF) and biomagnification factors (BMF). The combination of these processes is represented by the bioaccumulation factor (BAF).

The BCF is the ratio of the concentration in the organism divided by the water concentration, where the water phase is the only exposure route. BCF values are mostly determined in the laboratory. The concentration in the organism is expressed on a wet weight basis and preferably normalised to 5% lipids (ECHA, 2012). However, lipid normalisation is not relevant for mercury since it does not accumulate in lipids. If normalized, THg concentrations are usually normalized to dry weight content.
The BMF is the ratio of the concentration in a predator organism divided by the concentration in its prey. The BMF is usually determined on the basis of field studies and for hydrophobic organic chemicals commonly normalised to lipid content of prey and predator. Two BMFs are distinguished in the guidance document (EC, 2011). The first, BMF_1, describes the overall biomagnification from aquatic organisms to larger fish (TL4) in the aquatic environment that in turn is eaten by predators (including humans). For the marine environment, a second BMF_2 is included to account for accumulation in bird and mammals at TL5 (e.g. seals, dolphins, seabirds) that serve as food for top predators such as polar bears and killer whales.
For biomagnifying substances, only the first trophic level of primary consumers is in equilibrium with the water phase. The next trophic levels deviate from equilibrium if biomagnification occurs. The overall BMF up to the fourth trophic level in the aquatic environment thus

actually comprises three biomagnification steps. If biomagnification is expressed as the trophic magnification factor (TMF, which is the average increase in concentrations per trophic level) then the overall biomagnification step to TL 4 is equal to TMF^3 (Burkhard et al., 2013; Verbruggen, 2014).

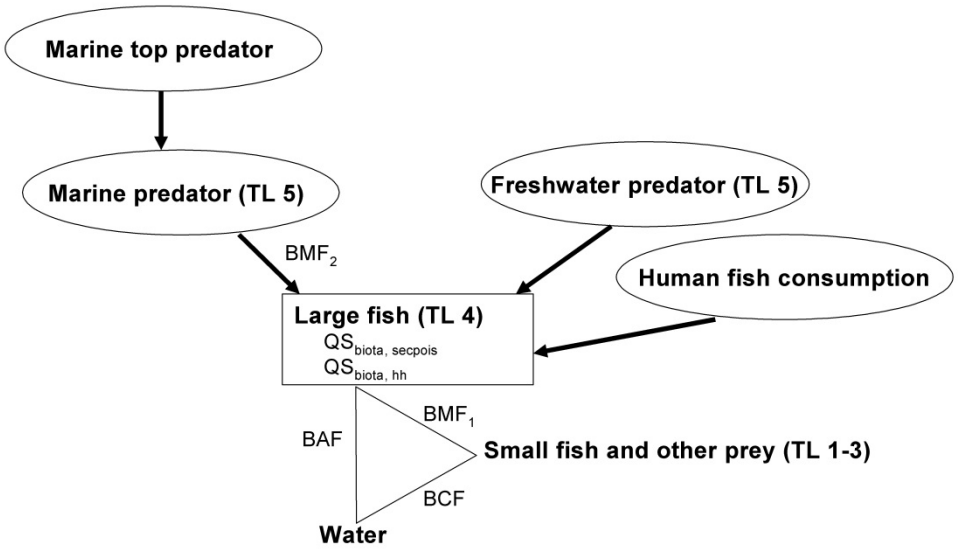

Figure 1 Scheme on how to recalculate biota standards into water concentrations. Ovals are protection goals (species to be protected); the rectangle is the trophic level on which the QS are set to protect the upper trophic levels. TL = trophic level; assuming trophic level 1 = algae; 2 = zooplankton; 3 = small fish; 4 = large fish; 5 = predatory birds, mammals, and large predatory fish. $QS_{biota,\ secpois}$ is the quality standard protecting predators (through secondary poisoning). $QS_{biota,\ hh}$ is the quality standard protecting humans (through the consumption of fish and fishery products). For $QS_{biota\ secpois}$ in freshwater, only the BMF1 is relevant. For $QS_{biota,\ secpois}$ in marine waters, the BMF1 and BMF2 are relevant because an additional trophic level should be included to protect marine top predators. Figure copied from Moermond and Verbruggen (2012).

In general, biomagnification, and thus total bioaccumulation, increases with increasing bioconcentration potential. The combination of bioconcentration and biomagnificiation is represented by the bioaccumulation factor (BAF). The BAF is a field-derived value that represents the resultant of bioconcentration and biomagnification. It is determined as the ratio of the concentration in an organism divided by the concentration in its surroundings (the water column), preferably normalised to 5% lipids (ECHA, 2012). For mercury, normalization to lipids is not applicable. If only fish are considered, differences in moisture content are limited (EFSA, 2009; Smit, 2005) and wet weight concentrations could be used for mercury in fish.

2.1.1 Calculations using BCF, BMF or BAF

According to section 4.4.4.1 of the TGD-EQS the biota-based QS for secondary poisoning should be calculated separately for the freshwater and saltwater environment. The $QS_{biota,\ secpois,\ fw}$ is derived using the lowest toxicity value for birds or mammals with the appropriate

assessment factor, while for the $QS_{biota,\ secpois,\ sw}$ the toxicity value should also be divided by the BMF_2 to account for the above described additional trophic level. The TGD-EQS gives the following equations:

$$QS_{biota,\ secpois,\ fw} = \frac{TOX_{oral}}{AF} \quad \text{Eq. 1}$$

$$QS_{biota,\ secpois,\ sw} = \frac{TOX_{oral}}{AF \times BMF_2} \quad \text{Eq. 2}$$

The corresponding concentrations in water, denoted as $QS_{fw,\ secpois}$ and $QS_{fw,\ secpois}$ are then calculated by dividing the $QS_{biota,\ secpois,\ fw}$ and $QS_{biota,\ secpois,\ sw}$ by the product of BCF and BMF_1 (see TGD-EQS, section 4.7.2.1):

$$QS_{fw,\ secpois} = \frac{QS_{biota,\ secpois,\ fw}}{BCF \times BMF_1} \quad \text{Eq. 3}$$

$$QS_{sw,\ secpois} = \frac{QS_{biota,\ secpois,\ sw}}{BCF \times BMF_1} \quad \text{Eq. 4}$$

Again, the product of BCF and BMF_1 may be replaced by the BAF for the appropriate trophic level.

Instead of using the product of BCF and BMF, a field based BAF may be used that includes both uptake from the water phase and uptake via food. The $QS_{water,\ secpois}$ can also be calculated according to Equation 5:

$$QS_{water,\ secpois} = \frac{QS_{biota,\ secpois}}{BAF} \quad \text{Eq. 5}$$

In this case, care should be taken that the BAF is derived for the appropriate trophic level. Forfish, a BAF at TL4 can replace the product of BCF and BMF_1. Deriving different biota standards for freshwater and marine waters has apparently not been considered in the EQS-dossier on mercury, since one value is presented for all waters, including marine. Therefore, in this report also a single value is derived, based on the EQS_{biota} for mercury fish.
In general, preference is given to the use of BAFs instead of using the product of BCF and BMF_1, because the BAF is based on field samples and includes all possible uptake routes and it can be directly derived from concentrations in biota at the appropriate trophic level. For a valid BAF, however, insight into the corresponding concentrations in water is needed. BMFs are generally also derived from field studies, which nowadays often study the transfer of a compound through the food chain as a function of trophic level. In that case, the BMF per trophic level is referred to as Trophic Magnification Factor (TMF). To apply a BMF in combination with a BCF value, the biomagnification factor should include all steps from the organisms that are in thermodynamic equilibrium with the water phase up to the trophic level that corresponds

to the biota standard (TL4). Usually, only algae (trophic level 1) are in equilibrium with the water concentration, if biomagnification occurs (e.g. Burkhard et al., 2013). This kind of biomagnification factors over the entire pelagic food chain are not often reported.

2.2 Uncertainty about published BAFs

As indicated in section 1.2, one of the reasons for not setting a water-based EQS for secondary poisoning of mercury was the uncertainty associated with the BAF. The EQS datasheet reports BAFs for MeHg that span four orders of magnitude (EC, 2005). A likely cause for this variation in data is the complex chemistry of mercury. In natural waters, mercury is predominantly present in its metallic and inorganic forms and about 1-10% is present as organic methylmercury (MeHg). In fish, 80-99% is present in the methylated form due to the biomagnification of MeHg from food, but also due to internal and external methylation of inorganic mercury (Slooff et al., 1995). Normally, for deriving a BAF, the concentrations measured in the organism and the corresponding water concentrations should be based on the same compound. For mercury, however, a BAF could be based on the summed concentration of all dissolved mercury forms in water, indicated as dissolved total[1] mercury (THg), because all mercury forms in water will contribute to the internal MeHg levels in fish. If BAFs are based solely on MeHg concentrations in water, resulting values will be much higher, because MeHg concentrations in water are only small compared to the dominant inorganic mercury species. Whether THg of MeHg concentrations in fish are used is less relevant, because the fraction of MeHg is high in fish. However, at lower trophic levels, fractions of MeHg will be lower as well. This may partly explain a wide range and high values of observed BAF values based on MeHg as described in the EQS dossier (EC, 2005).

In a previous Dutch national assessments of secondary poisoning it was assumed that the BAF for mercury, although based on dissolved THg in water, should also be used to estimate accumulation of MeHg in fish as described above (Slooff et al., 1995; Smit et al., 2000). However, the previously used BAF value of 21700 L/kg_{wwt} seems to be rather low as compared to the range presented in the EQS dossier. This value was based on monitoring data in fish and surface waters in the Netherlands from 1988-1989 (Romijn et al., 1991; Slooff et al., 1995). Reported geometric mean mercury concentrations in water (0.01 and 0.06 µg/L dissolved THg) seem to be rather high as compared to more recent data (Van Duijnhoven, 2011), which may explain the relatively low BAF obtained at that time. Moreover, it is not fully clear if the water concentrations refer to the sites or regions where fish were caught.

Another major influence on the value of the BAF values is the trophic level of the species. In the EQS dossier no distinction is made between the trophic level for the reported BAF values. Mercury is known for its high biomagnification potential, with average increase in concentration per trophic level for aquatic ecosystems worldwide by a factor of 3.5 for THg and 6.5 for MeHg (Lavoie et al., 2013). From these values, also the

[1] Note that 'total' in this context refers to the summed concentration of all mercury species and is not meant as the opposite of dissolved. Therefore, the term "dissolved THg" will be used.

increase in the fraction MeHg with trophic level becomes apparent. The influence of trophic level will be discussed further below.

2.3 Determination of the BAF for mercury in relation to trophic level

2.3.1 Literature search

Numerous studies on mercury accumulation have been published during the past years. For the purpose of this assessment, laboratory studies on bioconcentration are considered less relevant and only field bioaccumulation studies were selected. The available literature was screened for studies that could be used to establish a relationship between BAF and trophic level, and/or trophic magnification studies from which TMFs could be derived. From these, only studies were evaluated in which measured concentrations in organisms were reported together with dissolved mercury concentrations measured in (filtered) water samples taken during organism sampling. A total of around 20 scientific publications together with underlying reports and data was selected for further evaluation.

2.3.2 Evaluation and assessment

The selected studies were evaluated with respect to scientific reliability and relevance for the present assessment. Reliability indices (Ri) were assigned according to (Klimisch et al., 1997), with Ri1 being fully reliable, Ri2 reliable with restrictions, Ri3 not reliable and Ri4 not assignable. BAFs were calculated from the reported concentrations of THg in fish and dissolved concentrations of THg and/or MeHg. If fish concentrations were only reported on a dry weight basis, corresponding wet weight based BAFs were calculated using the reported moisture content or default values (EFSA, 2009; Smit, 2005). If data could only be retrieved from figures, the datapoints were extracted using the TechDig software program (Jones, 1998)

Sometimes both MeHg and THg concentrations were reported in fish. The BAF values for MeHg were then calculated from the MeHg concentrations in fish and water.However, the EQS for mercury is for THg and contrary to the concentration of MeHg, the concentration of THg in fish was always reported. Therefore, all BAF values were based on the THg concentration in fish, and if necessary recalculated. The BAF for THg refers thus to the THg concentration in fish divided by the THg concentration in water and the BAF for MeHg to the THg concentration in fish divided by the MeHg concentration in water. Because the fraction MeHg is high in fish, differences are small.

In case only total THg or MeHg were reported from measurements in unfiltered samples, BAFs were taken into account if data indicated that filtration would only have made a minor difference. This is the case if reported Dissolved Organic Carbon (DOC) and Total Organic Carbon (TOC) are similar, or when it can be assumed that TOC is low e.g. in oceanic regions. In those cases, it can be assumed that unfiltered and filtered mercury concentrations are similar.

To be able to establish the relationship between BAF and trophic level, only studies with information on the latter are relevant. Preferably, the trophic position is determined by measuring stable isotopes in the biota samples; the enrichment in nitrogen isotope ratio ($\delta^{15}N$) in those samples is a measure of trophic position, see e.g. (Jardine et al., 2006; Vander Zanden et al., 1997). When trophic levels were not reported

they were calculated from the reported $\delta^{15}N$-values for zooplankton or periphyton, respectively, as:

$TL = 2+(\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton})/3.4$, or

$TL = 1+(\delta^{15}N_{consumer} - \delta^{15}N_{periphyton})/3.4$.

With these formulas, zooplankton or periphyton are set at TL 2 or 1 respectively. Which formula is used depends on the data available, when $\delta^{15}N$ values are available for both zooplankton and periphyton, preference is usually given to zooplankton. The factor 3.4 is the average increase in $\delta^{15}N$ per trophic level used in most trophic magnification studies.
For the derivation of the BAF at TL 4, a regression was made between the logarithm of BAF values and trophic level. For this purpose, individual fish samples were used as much as possible. In case only average values per species were available, these were used with inclusion of the number that underlie this value. In the regression method, this number is included in the weighting of the data, meaning a BAF based on e.g. 18 samples gets a weight 18 times higher than a BAF based on an individual sample.

3 Results and discussion

3.1 Summary of bioaccumulation field studies

3.1.1 Freshwater studies

Bowles et al. (2001) and Apte et al. (2000) examined the accumulation of mercury in Lake Murray in Papua New Guinea. Water was sampled in June 1995 and November 1996. Only during the second sampling round water samples were filtered (Apte et al., 2000). DOC concentrations were around 3.6 mg/L with values ranging from 1.9 to 5.7 mg/L. Total suspended solids (TSS) varied strongly from 2.0 to 42 mg/L with an average of around 10 mg/L. There was a strong correlation between TSS and unfiltered THg concentrations. Dissolved THg concentrations were 30 to 100% of unfiltered THg concentrations. For MeHg dissolved concentrations were 14% to 100% of unfiltered concentrations (51±30%). Dissolved THg concentrations were reported to be 0.9±0.4 ng/L THg (n=18) and dissolved MeHg concentrations 0.05 ng/L (Apte et al., 2000). The latter value might still be an overestimation, as 50% of all samples for unfiltered MeHg had concentrations lower than 0.05 ng/L. These dissolved concentrations reported in Apte et al. (2000) for 1996 are lower than the 1.42 and 0.067 ng/L reported for THg and MeHg, respectively, that were reported by both Bowles et al. (2001) and Apte et al. (2000) as average values over June 1995 and November 1996. Because water concentrations from June 1995 were not filtered (Apte et al., 2000; Bowles et al., 2001), these average concentrations for 1995 and 1996 have to refer to total concentrations. Because of the influence of TSS on these unfiltered concentrations, the dissolved concentrations reported by Apte et al. (2000) were used instead.
Plants and algae were sampled in June 1995 and November 1996. Seven species of fish from different trophic levels were collected in August 1996 and November 1996. The sampled fish included fly river gizzard shad (*Nematalosa flyensis*), *strickland river gizzard shad* (*Nematalosa papuensis*), groove-snouted catfish (*Arius berneyi*), seven-spotted archerfish (*Toxotes chatareus*), giant freshwater anchovy (*Thryssa scratchleyi*), Sepik garpike (*Strongylura kreffti*), barramundi (*Lates calcarifer*). Ratios for stable isotopes were presented and from these data trophic levels were calculated. Macroalgae were used as the base of the food chain (TL=1) and not seston, because this can be a mix of phytoplankton and zooplankton:

$$TL = 1+(\delta^{15}N_{consumer} - \delta^{15}N_{macroalgae})/3.4$$

Seston ended up at trophic level 1.4. The 7 fish species levels ranging from 2.1 to 3.4. The Appendix to the report by Apte et al. (2000) could not be retrieved. Therefore, data are not based on individual fish, but on species means instead. For fish, average log BAF per species ranged from 4.7 to 5.7 for THg and 6.0 to 7.0 for THg concentrations in fish relative to the MeHg concentration in water, both on wet weight basis. *The slope of the logarithm of wet-weight MeHg concentrations in fish versus* $\delta^{15}N$ was 0.28. With the assumption of an enrichment in $\delta^{15}N$ of 3.4‰ per trophic level, the TMF for MeHg in fish in Lake Murray is 9.0. (Ri=2)

Campbell et al. (2003a) determined the bioaccumulation of mercury in Napoleon Gulf (Uganda) and Winam Gulf (Kenya) in northern Lake Victoria. Water and organisms were sampled in October and November 1998 for Napoleon Gulf and December 1998 for Winam Gulf. Water samples were not filtered. Particulate organic carbon and total suspended particles were not reported. Besides that, mercury concentrations were high. Concentrations for THg in Napoleon Gulf varied from 1.9 to 5.8 ng/L, while MeHg concentrations ranged from 0.2 to 1 ng/L. THg concentrations in Winam Gulf varied from 2.9 to 4.5 ng/L.
Trophic levels were calculated from the presented data on stable isotopes. For this purpose, the data for planktivorous shrimp *Caridina nilotica* (Campbell et al., 2003b) was used for both sites as reference for the food chain (TL=2):

$$TL = 2+(\delta^{15}N_{consumer} - \delta^{15}N_{shrimp})/3.4$$

In Napoleon Gulf, six fish species were sampled: cichlids (*Haplochromis* spp.), Nile perch (*Lates niloticus*), Nile tilapia (*Oreochromis niloticus*), marbled lungfish (*Protopterus aethiopicus*), silver cyprinid (*Rastrineobola argentea*), and redbelly tilapia (*Tilapia zilli*), with Nile perch and Nile tilapia each divided in three size classes. Trophic level varied from 2.3 from the smallest group of Nile tilapia (5.1-20 cm) to 3.8 for the largest Nile perch (60.1-100 cm). In Winam Gulf, some other species were sampled (Fischer's Victoria squeaker (*Synodontis afrofischeri*), semutundu (*Bagrus docmak*), North African catfish (*Clarias gariepinus*) and silver catfish (*Schilbe intermedius*) instead of silver cyprinid and redbelly tilapia). The trophic levels varied from 2.4 for the middle group of Nile tilapia (20.1–40 cm) to 3.7 for a group of 4 large Nile perches (60.1-100 cm).
For fish in Napoleon Gulf, average log BAF per group ranged from 3.8 to 4.9 for THg and 4.6 to 5.7 for MeHg, both on wet weight basis. For Winam Gulf, the log BAF values for THg varied from 3.5 to 5.0. The slope of the logarithm of wet-weight THg concentrations in fish versus $\delta^{15}N$ was 0.163 for Napoleon Gulf and 0.165 for Winam Gulf. With the assumption of an enrichment in $\delta^{15}N$ of 3.4‰ per trophic level, the TMFs for THg are 3.6. Indeed, these values are low, probably reflecting a high association with suspended particles. Because of the influence of POC and TSS, BAF calculated from the reported water concentrations are considered unreliable. (Ri=3)

Dominique et al. (2007) reported concentrations for THg and MeHg in *Curimata cyprinoides* and *Triportheus rotundatus* collected in a water reservoir and downstream of the reservoir in French Guyana in March 2003. Water samples were collected in the same period. Water concentrations are reported both for unfiltered and filtered samples and both for THg and MeHg. Both THg and MeHg concentrations in fish were reported for skeletal muscle only based on a dry weight basis. The numbers of *C. cyprinoides* for which THg concentrations were determined was 25 for the reservoir site and 41 for the downstream site, and the number of *T. rotundatus* was 10 for both sites. On the basis of the reported average dry weight concentrations in the two fish species and filtered water concentrations at both locations, log BAF values were calculated ranging from 6.0 to 6.7 for THg and from 6.5 to

7.5 for THg in fish and MeHg in water. Stable isotopes were determined for the fish, benthic invertebrates (insects), biofilm and seston. Seston was not further specified. However, it was assumed that this was trophic level 1. Trophic levels were estimated with $\delta^{15}N$ values for seston as basis:

$$TL = 1+(\delta^{15}N_{consumer} - \delta^{15}N_{periphyton})/3.4.$$

This resulted in erroneously low values for biofilm and benthic invertebrates. However, trophic level for fish was 1.9 and 2.8 for *C. cyprinoides* and 2.9 and 2.4 for *T. rotundatus*. This low trophic level for both species is in accordance with the low trophic level reported by fishbase (2.4), but also with the analysis of stomach content determined in the study itself, which shows that the species are mainly detrivorous. Carbon isotope analysis shows however a strong link with biofilm, at least for *C. cyprinoides*, but $\delta^{15}N$ signatures for biofilm are depleted compared to atmospheric nitrogen and are thus erroneously low. Data for the two fish species and seston show a consistent pattern with an increase in dry weight concentrations of around 4 per trophic level for the data from the two sites combined. (Ri = 2)

Poste et al. (2012) examined mercury uptake in the food web in two different bays of Lake Victoria (Murchison Bay and Napoleon Gulf). Water and biota samples were taken between September 2008 and February 2009. Water concentrations were determined in unfiltered water samples. Because of the high plankton content (260 and 50 mg/L at Murchison Bay and Napoleon Gulf, respectively), dissolved concentrations will be substantially lower. Using the reported concentration of THg in phytoplankton, concentrations were corrected from 1.3 and 0.53 ng/L to 0.81 and 0.43 ng/L for Murchison Bay and Napoleon Gulf, respectively. Wet weight concentrations for 10 different fish species in the Murchison Bay and 14 species in the Napoleon Gulf are reported: cichlids (*Haplochromis* spp.), Nile perch (*Lates niloticus*), blue spotted tilapia (*Oreochromis leucostictus*), Nile tilapia (*O. niloticus*), marbled lungfish (*Protopterus aethiopicus*), silver cyprinid or dagaa (*Rastrineobola argentea*), Fischer's Victoria squeaker (*Synodontis afrofischeri*), Lake Victoria squeaker (*S. victoriae*), and redbelly tilapia (*Tilapia zilli*) from both embayments, African sharptooth catfish (*Clarias gariepinus*) from Murchison Bay and Alluaud's haplo (*Astatoreochromis alluaudi*), silver catfish (*Bagrus docmac*), Sadler's robber (*Brycinus sadleri*), elephant-snout fish (*Mormyrus kannume*) and Victoria tilapia (*O. variabilis*) from Napoleon Gulf). For the Murchison Bay log BAFs for fish are determined ranging from 4.0 to 4.8 and for the Napoleon Gulf from 3.9 to 5.4. Trophic levels were calculated from reported values for $\delta^{15}N$ with Nile tilapia as basis:

$$TL = 2+(\delta^{15}N_{consumer} - \delta^{15}N_{Nile\ tilapia})/3.4$$

In the study itself, it is indicated that trophic levels of the fish species are not significantly different between the two embayments, when normalized to Nile tilapia. Further, in fishbase it is reported that the trophic level of this species is 2.0±0.00. Trophic levels of the fish species range from 2.0 to 2.8 for Murchison Bay and from 1.8 to 3.4 for Napoleon Gulf. Calculated trophic levels are 2.0 and 2.2 for zooplankton

and 1.4 and 0.9 for periphyton, in Murchison Bay and Napoleon Gulf, respectively. TMFs, calculated from the presented slopes of the regression of log concentrations with $\delta^{15}N$, assuming an enrichment of 3.4‰ per trophic level, are 2.8 for Murchison Bay and 4.8 for the Napoleon Gulf. (Ri = 2)

Cheng et al. (2011) studied biomagnification of mercury in four aquaculture ponds with two different food chains (omnivorous and predatory) in the Pearl River Delta, China. For each food chain two different ponds were sampled, water samples were collected in May 2009, collection date of biota samples is not reported, but was possibly performed together with sediment sampling in November 2008. In all ponds zooplankton was sampled, in the ponds with omnivorous food chains grass carp (*Ctenopharyngodon idellus*) and bighead carp (*Aristichthys nobilis*) were sampled, in the ponds with the predatory food chain mud carp (*Cirrhina molitorella*) and mandarin fish (*Siniperca chuatsi*) were sampled. In the pond with the omnivorous food chain, the fish were fed with fish feed. In the predatory food chain, the mandarin fish were fed with juvenile mud carp. It is reported that these mud carp were purchased as well, just as the fish food from the omnivorous food chain. Thus, in both food chains the food at the bottom of the food chain is at least partly not related to the food ecosystem.
Filtered water concentrations and concentrations in biota on dry weight basis were reported for both THg and MeHg. Especially filtered THg concentrations were extraordinarily high with values ranging from 18 to 35 ng/L in the four ponds (in various studies reported here water concentrations stayed below 6 ng/L for all types of ecosystems).
Only one value for $\delta^{15}N$ for zooplankton was available for all four ponds and for the fish species only one value for two ponds with the same food chain. However, the number of total zooplankton samples did not match the data presented in the figures. The reported trophic levels of the fish had a range of 2.5 to 3.9, but calculated from the reported $\delta^{15}N$ data these should rather be 2.5 to 4.2.
Biota THg and MeHg concentrations for each pond were only reported as pooled values. Individual data for the biota concentrations versus $\delta^{15}N$ were shown in figures, but the data for the two omnivorous food chains and for the two predatory food chains were pooled in the figures too. Consequently, it was not clear which of the ponds the data referred to.
Log BAF for the fish (based on dry weight) were in the range of 3.0 to 4.0 for THg and 4.8 to 5.8 for MeHg. These BAFs are clearly lower than all other valid BAF considered in this report. The reported TMFs for the omnivorous food chain were 1.94 and 2.34 for THg and MeHg, respectively, based on pooled data for both ponds. For the predatory food chain, these values were 2.04 and 2.60, respectively. Since the fish in the ponds were fed with food that was not originating from the ponds and the because the aqueous concentrations in the pond were extraordinarily high, these BAFs (and TMFs) should considered unreliable (Ri = 3).

Watras et al. (1998) examined bioaccumulation in small fish in 14 different lakes in Vilas County (USA). Microseston (phytoplankton and bacterioplankton), zooplankton and small fish (yellow perch (*Perca flavescens*) and Golden shiner (*Notemigonus crysoleuca*) were sampled during spring and summer 1994. Average concentrations in fish for the

different lakes were plotted in figures. BAF values for THg calculated from these data varied between 4.2 and 5.6 on a wet weight basis and between 4.8 to 6.2 on a dry weight basis. BAF values for THg in fish relative to MeHg concentration in water were between 5.0 and 6.6 on a wet weight basis and between 5.6 and 7.2 on a dry weight basis. BAF did not correlate with pH , but there was a significant negative correlation with DOC. Age of the fish varied up to seven years, and therefore the exact trophic level will most likely be variable as well. Trophic levels and $\delta\ ^{15}N$ levels were not determined. Neither there is information on the number of fish for each lake. Therefore this publication does not contain enough information to be used in the regression. (Ri = 4)

Doetzel (2007) examined the accumulation of mercury in zooplankton, benthic invertebrates and fish, mainly lake trout and lake whitefish from lakes in Northern Canada. For 10 lakes from the Mackenzie River Basin (MRB) water concentrations and stable isotopes are available. Data were read from figures with some additional data for lake trout (*Salvelinus namaycush*) from the tables. Data from figures matched very well with the corresponding data from the tables. Biota sampling in these lakes occurred in the summers in the period of 2001-2003 (one or two consecutive years). Water sampling was performed in the same period. In total this yielded 401 data for individual fish, being 2 burbots (*Lota lota*), 196 lake trouts, 194 lake whitefish (*Coregonus clupeaformis*), 5 longnose suckers (*Catostomus catostomus*), 1 ninespine stickleback (*Pungitius pungitius*), 1 northern pike (*Esox lucius*), and 2 slimy sculpins (*Cottus cognatus*). Trophic levels were calculated from the presented data on $\delta\ ^{15}N$ as:

$$TL = 2+(\delta^{15}N_{consumer} - \delta\ ^{15}N_{zooplankton})/3.4,$$

This resulted in a range from 2.4 for burbot to 5.3 for very old lake trout. Concentrations in biota were reported on a wet weight basis. The log BAF (L/kg) values for THg varied from 3.2 to 6.7 based on wet weight concentrations. BAF values for THg concentration in fish relative to MeHg in water varied from 4.5 to 8.1 on wet weight basis. Water samples have not been filtered. Lakes were oligotrophic to mesotrophic, and thus the influence of particulate matter will be rather limited. (Ri = 2)

Evans et al. (2005) present similar data for a number of lakes. The water chemistry for the lakes is the same as in the above study by Doetzel (2007). It is stated that the majority of the sampling occurred between 1996 and 2000. However, for the lakes Cli, Little Doctor and Willow there is an almost complete overlap with the data for lake whitefish and lake trout from Doetzel (2007). Therefore, only 13 additional lake whitefish from Lake Willow, and the other specimens not mentioned by Doetzel (2007) were included (7 burbots and 30 northern pikes from Lake Willow and 6 white suckers, 5 walleyes and 6 northern pikes from lake Little Doctor). For the other two lakes (Sibbeston and Tsetso), data on 7 white suckers, and respectively 48 and 50 lake whitefish, 5 and 43 walleyes, and 2 and 3 northern pikes were read from the presented figures. (Ri=2)

Herrin et al. (1998) studied the uptake of MeHg in the food chain of Devil's lake in Wisconsin (USA). Water samples were not filtered. BAF values could be calculated from the presented THg concentrations in fish and THg and MeHg concentrations in unfiltered water. In 1994 and 1995, log BAF values for mimic shiners (*Notropis volucellus*) were 5.4 and 5.4 for for THg in fish compared with THg in water and 6.0 and 6.7 compared with MeHg in water, both on a dry weight basis. On a wet weight basis, log BAF values were 4.8 for THg water concentrations and 5.4 and 6.1 for MeHg water concentrations. For bluegill (*Lepomis macrochirus*) dry weight based log BAF values were 5.4 to 5.5 for THg water concentrations and 5.8 to 6.7 for MeHg water concentrations over the years 1994 and 1995. Based on wet weight, log BAF values were 4.8 for THg and 5.2 to 6.0 for MeHg.
Trophic levels ($\delta^{15}N$) were not determined and thus no information on the variability of trophic level is available. All fish were yearlings. In the study, it is assumed that these are strictly feeding on zooplankton and consequently, fish should be assigned to trophic level 3. However, this assumption might be an approximation. Stomach content showed that at least 15% consisted of zooplankton. In May and October this was 85% or more. So, stomach content does not provide unequivocal information on trophic levels as well.
Also the influence of particulate matter on the concentration is not clear. Water concentrations of MeHg were 0.52 ng/L in 1994 and 0.07 ng/L in 1995, while concentrations of MeHg associated to particulate matter were 0.07 and 0.05 ng/L in 1994 and 1995 respectively. So, the MeHg concentrations and the fraction of MeHg associated to particulate matter is highly variable over the years of monitoring. The influence of particulate matter on THg concentrations is also unknown. It appeared that THg concentrations per liter of water associated to particulate matter were as high as or even higher than filtered concentrations. This was thought to be due to contaminated filter, which was not an issue for MeHg concentrations. Nevertheless, a significant fraction of THg might be associated to particulate matter as well. For the reasons described above, the study was not further used. Furthermore, the number of fish that was sampled is unknown. (Ri = 3)

Chasar et al. (2009) examined bioaccumulation of mercury in eight streams in the states Oregon (OR), Wisconsin (WI), and Florida (FL). Water samples were collected 18 times in the period 2002-2004. Water samples were filtered and both THg and MeHg were determined. Invertebrates and forage fish were sampled in both spring and fall of 2003, predator fish were collected once in the summer or fall of 2003 or 2004. In fish, THg concentrations were reported as dry weight concentrations, moisture content of each fish was reported as well. The fish that were sampled in the different streams were: reticulate sculpin (*Cottus perplexus*), mosquitofish (*Gambusia affinis*), cutthroat trout (*Oncorhynchus clarkii*), and redside shiner (*Richardsonius balteatus*), mottled sculpin (*Cottus bairdii*), blacknose dace (*Rhinichthys atratulus*), brown trout (*Salmo trutta*), warmouth (*Chaenobryttus gulosus*), seminole killifish (*Fundulus seminolis*), eastern mosquitofish (*G. holbrooki*), redbreast sunfish (*Lepomis auritus*), bluegill (*L. macrochirus*), spotted sunfish (*L. punctatus*), largemouth bass (*Micropterus salmoides*), coastal shiner (*Notropis petersoni*), sculpin (*Cottus* sp.), speckled dace (*Rhinichthys osculus*), rainbow trout

(*O. mykiss*), creek chub (*Semotilus atromaculatus*), green sunfish (*L. cyanellus*), sailfin molly (*Poecilia latipinna*), and pumpkinseed (*L. gibbosus*). Data for individual fish were retrieved from an underlying report (Chasar et al., 2008).
Trophic levels were determined from the presented data on $\delta^{15}N$ with glossosomatid and hydropsychid caddisfly, baetid mayfly, and chironomid larvae and amphipods as reference (TL=2):

$$TL = 2+(\delta^{15}N_{consumer} - \delta^{15}N_{invertebrtes})/3.4$$

For the non-urban streams this worked well, with trophic levels for individual fish ranging from 3.1 to 4.1 for Lookout Creek (OR), from 2.5 to 3.2 for Pike River (WI), from 3.0 to 4.0 for Evergreen River (WI), from 1.7 to 4.0 for St. Marys River (FL), and from 2.2 to 4.5 for Santa Fe River (FL). However, for the urban streams $\delta^{15}N$ at the level of the invertebrates was already high, i.e. around 10‰ or even higher. The enrichment of ^{15}N in fish relative to this trophic level, was limited leading to rather low trophic levels, ranging from 1.7 to 2.9 for Beaverton Creek (OR), from 1.2 to 2.9 for Oak Creek (WI), and from 1.3 to 3.0 for Little Wekiva River (FL). In summary, estimated trophic levels for fish from $\delta^{15}N$ are too low for these systems, differentiation in trophic levels is low and estimated values do not correlate with reported trophic levels (from fishbase). Because trophic levels could not be accurately estimated from $\delta^{15}N$ the data from these urban streams were not further taken into account in the final regression analysis.
The data set resulted in data combinations of BAF and trophic level for 579 individual fish, of which 357 were used in the final regression analysis. These 357 BAF values for THg for individual fish ranged from 3.3 to 5.6 on wet weight basis, and from 3.8 to 6.3 on dry weight basis, BAF values for THg concentrations in fish compared to MeHg water concentrations ranged from 4.5 to 6.6 on wet weight basis, and from 5.0 to 7.3 on dry weight basis.
Slopes of of logarithm of THg concentrations based on dry weight versus $\delta^{15}N$ ranged from 0.14 to 0.26 in seven systems and was not significant for the data of one of the urban streams. Assuming a trophic enrichment in $\delta^{15}N$ of 3.4‰ per trophic level, the TMF for these seven streams ranges from 3.0 to 7.7. (Ri=2).

Stewart et al. (2008) examined uptake of MeHg in the food web of the Camp Far West Reservoir located in an historic gold mining region in California (USA). MeHg concentrations in three fish species (collected in August 2002 and August 2003), crustacean and zooplankton were reported. The concentrations of dissolved THg and MeHg in the water samples (collected in the same period as the fish samples) were measured and reported with more detail in an underlying report by Alpers et al. (2008). To calculcate the BAF values the water concentrations for the water concentrations for the epilimnion (excluding Dairy Farm Mine Pit Lake and Impoundments) were used. These concentrations were 0.96±0.49 ng/L for filtered THg and 0.039±0.014 ng/L for filtered MeHg. This water concentration for MeHg is also reported by Stewart et al. (2008). The numbers of fish used in the study were 60 threadfin shad (*Dorosoma petenense*), 15 bluegill (*Lepomis macrochirus*), and 20 spotted bass (*Micropterus punctulatus*), received by personal communication (Dr. Stewart from USGS Water Resources

Division). The mean ratios of MeHg:THg and the moisture contents were retrieved from Saiki et al. (2010). The ratios MeHg:THg were 0.78, 0.93, and 0.87 and the moisture fractions were 0.777, 0.739, and 0.736, for threadfin shad, bluegill, and spotted bass respectively. The log BAFs for threadfin shad, bluegill and spotted bass are 5.2, 5.4 and 5.9 on basis of wet weight THg concentrations and 5.8, 6.0, and 6.4 on dry weight basis, respectively. With the MeHg concentrations in water, the log BAFs were 6.6,6.8, and 7.3 on wet weight basis and 7.2, 7.4, and 7.8 on dry weight basis. These fish species represent trophic levels of 3.2, 3.5 and 4.3 respectively, based on an average increase in $\delta^{15}N$ of 3.4‰ per trophic level with suspended matter as trophic level 1 (with these values zooplankton (>75 µm) has trophic level 2.1, mayfly nymphs (Baetidae) 1.9, midge larvae (Chironimidae) 2.4 and crayfish (*Orconectes virilis*) 2.8). On the basis of the linear correlation $^{10}\log (MeHg) = 0.20 * (\delta\ ^{15}N) + 0.55$ ($r^2 = 0.83$; $p < 0.0001$), based on dry weight, the TMF was determined to be 4.8 (Ri = 2).

Cui et al. (2011) examined transfer of mercury in food web in wetlands of the Yellow River Delta (China). Plants, molluscs, crustacean, fish and birds were collected in August 2008. Water samples were taken in the same period but concentrations did not exceed the detection limit of 5 ng/L for filtered (0.45 µm) in water. The six fish species, catfish (*Chaeturichthys sitgmatias*, TL=2.85), common carp (*Cyprinus carpio*, TL=2.65), javelin goby (*Acanthogobius hasta*, TL=3.65), redeye mullet (*Liza haematocheila*, TL=2.18), silver carp (*Hypophthalmichthys molitrix*, TL=2.29) and weever (*Lateolabras japonicas*, TL=2.74) had log BAF values > 4.3 to > 5.4, based on dry weight concentrations of THg. These species covered the trophic range 2.2 to 3.7. The TMF, determined on the basis of the correlation $^{10}\log (Hg) = -1.76 + 0.45 * TL$ ($r^2 = 0.45$; $p = 0.10$), was 2.8 for the food web consisting of aquatic plants, invertebrates, fish, and birds (Ri = 2).

Rolfhus et al. (2011) has reported an assessment of bioaccumulation of mercury in the food web of lakes in the Great Lakes regions in Canada and the USA based on the study by Wiener et al. (2006). Although in the publications themselves no data are presented from which BAFs could be derived, through personal communication (Dr. Rolfhus, Dr. Wiener and Dr. Sandheinrich from the USGS Water Science Center in Minnesota and the University of Wisconsin-La Crosse) a data set was received from which BAFs and related TLs could be gathered for 13 lakes in Voyageurs National Park (MN, USA) as reported by Wiener et al. (2006). The data set contains dry weight biota concentrations of THg in yellow perch (*Perca flavescens*) and northern pike (*Esox lucius*) for 16 different lakes and total (unfiltered) water concentrations for THg and MeHg. In total 68 BAFs for individual perch samples and 125 BAFs for individual pike samples were determined.
Log BAF values (dry weight) for 1-year-old perch were in the range of 4.8 to 5.8 for BAF based on THg in water and 5.8 to 7.0 for BAF based on MeHg in water and for pike these were 5.4 to 7.0 for THg and 6.5 to 8.3 for MeHg. Reported concentrations for fish were on basis of MeHg, but this was based on the assumption that 100% of the THg concentration in fish is MeHg (Rolfhus et al., 2011). Further, the dry weight concentrations were calculated assuming a general moisture

content of 80% (Rolfhus et al., 2011). This value was also used to calculate the original wet weight concentrations.
It appears that the baseline value for $\delta^{15}N$ of zooplankton is strongly variable with vary low values, sometimes even negative. Estimated trophic levels for 1-year old yellow perch varied from 2.5 to 4.6, while trophic levels for northern pike ranged from 2.9 to 5.4. According to Wiener et al. (2006) these 1-year-old yellow perch, approximately 5 cm and 1.5 g, are largely feeding on zooplankton and benthic invertebrates. Zooplankton and benthic invertebrates can be considered to form trophic level 2, and thus 1-year old perch belong to trophic level 3. Therefore, trophic levels were calculated from the presented data on $\delta^{15}N$ as:

$$TL = 3+(\delta^{15}N_{consumer} - \delta\ ^{15}N_{1\text{-y perch}})/3.4$$

The average value for $\delta^{15}N$ of 1-y old perch was taken, resulting in trophic levels for perch with a standard deviation of 0.1, which confirms the homogeneity within this group. Trophic levels for northen pike determined in this way ranged from 3.0 to 4.3.
For one lake, the MeHg levels were below the limit of determination of 0.04 ng/L at all sampling times and thus, the BAFs determined for this lake could only be calculated for THg. The DOC levels as well as TOC levels were reported for the lakes, DOC levels range from 4.4 to 18.5 mg/L and TOC levels from 4.2 to 18.9 mg/L (Goldstein et al., 2003). In general the TOC and DOC levels differ little from each other (mostly within 5% but often only 2 or 3%), which indicates that the level of particulate matter is probably very low in these lakes. Therefore, the water concentrations determined in unfiltered samples can be regarded equivalent to filtered water concentrations. (Ri = 2)

Gorski et al. (2003) reported on the bioaccumulation of mercury in two lakes on an island (Isle Royale) in lake Superior. Crustaceans, insects and fish were sampled between June 1998 and August 1999. Fish species were adult yellow perch (*Perca flavescens*) and northern pike (*Esox lucius*). Water samples were taken in the same period. In both lakes, the trophic level of Yyellow perch was 3.7, for northern pike, the trophic level differed slightly between the two lakes ranging from 4.2 to 4.3. Trophic levels were determined with caddisfly as basis of the food web:

$$TL = 2+(\delta^{15}N_{consumer} - \delta^{15}N_{caddisfly})/3.4$$

Zooplankton was also sampled, but had slightly enriched $\delta^{15}N$ values, which was attributed to not completely planktivorous species. Stable isotopes were not determined for age-1 yellow perch (31-40 mm), thus trophic levels could not be determined directly. However, from the data for Voyageurs National Park (Wiener et al., 2006), it appeared that the trophic level was very homogenous (see above). Therefore, also for this study, age-1 yellow perch were assigned trophic level 3 and which was used in combination with the presented aggregated data (n=10 for Lake Sargent and n=13 for Lake Richie).
For the other species only an average value for $\delta^{15}N$ was presented together with the standard deviation. For perch, $\delta^{15}N$ was determined in only 6 fish in each lake. However, it appeard that for adult perch the spread in trophic level was low too (standard deviation of 0.1 trophic

level). Concentrations for individual fish were shown in figures, from which they were retrieved. Concentrations for adult perch differed more than for age-1 yellow perch, but were rather well log-normally distributed with a relatively small standard deviation (0.2 log units). Therefore, the geometric mean concentrations in adult perch (n=33 for Lake Sargent and n=15 for Lake Richie) were used as aggregated data together with the average trophic level.
Also for northern pike, the spread in trophic level was only 0.1 to 0.2. $\delta^{15}N$ was determined in 14 fish in Lake Sargent and 11 fish in Lake Rich, which equals almost all northern pike sampled in the two lakes (n=16 for Lake Sargent and n=11 for Lake Richie). Also in this case, the concentrations are rather well log-normally distributed with a relatively small standard deviation (0.2-0.3 log units). Therefore, the data for northern pike were treated in the same way as for adult yellow perch. However, the reported data from an earlier study dating back to the year 2000 were excluded for further analysis.
The log BAF values for THg based on wet weight were 4.3 and 4.0 for age-1 yellow perch, 4.4 and 4.4 for adult yellow perch and 5.3 and 4.8 for northern pike in Lake Sargent and Lake Richie, respectively. Based on MeHg concentration in water, these values were 5.5 and 5.3 for age-1 yellow perch, 5.7 and 5.4 for adult yellow perch and 6.6 and 6.1 for northern pike. The reported BAF value in the study were based on dry weight, calculated from estimates for the moisture content of the fish. (Ri = 2)

Gantner et al. (2010b) examined the accumulation of mercury in Arctic char (*Salvelinus alpinus*) from periphyton in 18 different arctic lakes in the Canadian arctic. The majority of sampling was conducted during July and August of 2005, 2006 and 2007. Water sampling was performed in the same period and concentrations for 17 lakes were obtained through personal communication (Dr. Gantner, University of Northern British Columbia). Water concentrations in the lakes varied from 0.3 to 0.8 ng/L. Concentrations were determined in dorsal muscle of adult char and in whole body homogenates for juvenile char and sticklebacks (*Pungitius pungitius*). Concentrations were reported as wet weight concentrations for adult Arctic char and as dry weight concentrations for all other groups. For the recalculation of wet weight concentrations into dry weight concentrations, the reported average moisture weight content of 77% for muscle of adult Arctic char was used (Gantner et al., 2010a). For juvenile Arctic char and stickleback an average moisture content for fish of 73.7% was used (EFSA, 2009). If no value for moisture content is reported in the underlying study itself, this value has been used throughout this report.
Water samples are reported to have been filtered. Besides that, particulate organic carbon is very low (0.1 to 0.4 mg/L, concentrations for DOC and particulate organic carbon (POC) are probably erroneously reported as µg/L, i.e. 1000 times lower), and thus unlikely to affect the dissolved concentrations significantly. Trophic levels were determined as:

$$TL = 1+(\delta^{15}N_{consumer} - \delta^{15}N_{periphyton})/3.4$$

Trophic level for juvenile Arctic char in Char lake was reported as 0.7, but according to the $\delta^{15}N$ this should be in the order of 2.7 and so this

value was used in further analysis. The resulting trophic levels lie in a range from 1.8 to 4.3 for the 49 pooled data for in total 499 fish samples , with trophic levels of 2.1 to 4.3 for 33 pooled data of Arctic char, 1.8 to 3.3 for 14 pooled data of juvenile arctic char and 2.8 and 3.5 for 2 pooled data of sticklebacks.The log BAF (L/kg) values for total mercury (THg) in the Arctic char in 17 of the 18 lakes range from 5.1 to 6.4, based on wet weight concentrations. The log BAF values for the juvenile Arctic char range from 4.9 to 5.9, the log BAF values for the two composite stickleback samples were 5.9 and 5.7, for both juvenile Arctic char and stickleback based on dry weight (Ri = 2).
On the basis of the TLs determined, TMF values for MeHg were determined for the different lakes ranging from 3.6 to 64.3 based on MeHg concentrations on a dry weight basis. All 18 lakes showed a significant correlation between. There was no correlation between TMF and the food chain length, after removing the value of 64.3, which was an outlier (Gantner et al., 2010a). However, several food chains were very short (up to trophic level 2.1) with many fish and few invertebrate samples, which might have resulted in some very high TMF values (Borgå et al., 2012).

Paterson et al. (1998), Monson and Brezonik (1998) and Hall et al. (2009) studied bioaccumulation of mercury in (zoo)plankton. Similarly Back et al. (2003) and De Wit et al. (2012) examined crustaceans and insects and Watanabe et al. (2008) examined insects only. Since in these studies mercury was not determined in fish, they are not relevant for the present assessment.

3.1.2 Marine studies

Kim et al. (2012) determined THg and MeHg concentrations of 12 fish species in Masan Bay, a temperate estuary in Korea. Fish species, polychaete, bivalves, crustacean and cephalopod as well as water samples were collected in August and September 2009. The sampled fish were common mullet (*Mugil cephalus*), marbled flounder (*Pleuronectes yokohamae*), ridged-eye flounder (*P. cornutus*), ocellate spot skate (*Okamejei kenojei*), rudder fish (*Girella punctata*), yellow striped flounder (*P. herzensteini*), Korean rockfish (*Sebastes schlegeli*), conger eel (*Conger myriaster*), common silver-biddy (*Gerres oyena*), red seabream (*Pagrus major*), fat greenling (*Hexagrammos otakii*), and spotbelly rockfish (*S. pachycephalus*). Water concentrations were determined at the surface and bottom of the bay (depth 16 m). Water concentrations did not differ significantly. Both filtered and unfiltered water concentrations were measured, but only unfiltered water concentrations were reported. However, filtered concentrations could be calculated from the unfiltered concentrations in combination with the reported total suspended solids (TSS) and the partition coefficient to TSS. Concentrations in biota are expressed on a dry weight basis. BAF values are calculated for the different fish species ranging from 4.5 to 5.7 for THg and 5.9 to 7.1 for THg fish concentrations relative to MeHg water concentrations. The fish species had TL levels ranging from 2.5 to 3.7 based on *Mytilus edulis* as TL 2. $\delta^{15}N$ levels ranged from 10.3 to 16.2 and the correlation between dry weight concentrations and $\delta^{15}N$ resulted in TMF values of 2.5 for THg and 3.7 for MeHg, calculated from the presented slopes with an assumed average increase in $\delta^{15}N$ of

3.4‰ per trophic level. If only fish are considered, the TMFs are higher, being 9.0 for THg and 16 for MeHg. (Ri = 2)

Pethybridge et al. (2012) examined biomagnification of mercury in 16 cartilaginous fish (class Chondrichthyes) species southeast of Australia: 2 spiny dogfish (*Squalus acanthias*), 2 shortnose spurdogs (*S. megalops*), 1 broadnose sevengill shark (*Notorynchus cepedianus*), 2 southern dogfish (*Centrophorus zeehaani*), 2 shortspine spurdogs (*S. mitsukurii*), 2 Australian sawtail catsharks (*Figaro boardmani*), 1 South China catshark (*Apristurus sinensis*), 21 longnose velvet dogfish (*Centroselachus crepidater*), 2 roughskin dogfish (*Centroscymnus owstoni*), 2 Portuguese dogfish (*C. coelopsis*), 2 carpenter's chimaeras (*Chimaera lignaria*), 2 birdbeak dogfish (*Deania calcea*), 2 kitefin sharks (*Dalatias licha*), 20 New Zealand lanternsharks (*Etmopterus baxteri*), 2 Plunket's sharks (*Proscymnodoms plunketi*) and 2 Pacific spookfish (*Rhinochimaera pacifica*). Also three individual of other fish species (bony fish, class Osteichthyes) were examined: Hector's lanternfish (*Lampanyctodes hectoris*), cardinal fish (*Epigonus lenimen*) and redbait (*Emmelichthys nitidis*). Further, data for zooplankton, crustaceans, cephalopods (squids and octopus), and several groups of fish were presented but not further specified.
Biota samples (muscle tissue) were collected between 2004 and 2006. Water concentrations for this area were not reported in Pethybridge et al. (2012), but a mercury concentration in water for the relevant aera could be obtained from Cossa et al. (2011) for the year 2008. In this large oceanic compartments mercury concentrations are not expected to fluctuate significantly on a yearly basis. Water samples were not filtered, but POC is considered to be low enough to assume that filtration would not have influenced the results. The median THg concentration from all concentrations measured on the transect from Tasmania to Antarctica was 0.24 ng/L. From the presented figure, it appears that this concentration is representative for ocean water up to a latitude of 45 °S and 2000 m of depth. The concentrations in this area are rather homogeneous. Nevertheless, it was suggested by Pethybridge et al. (2012), referring to the same study by Cossa et al. (2011), that the concentration of MeHg increase with depth and that the relative amount of MeHg increases with depth as well. Indeed, the concentration of MeHg seems to increase with depth. The median MeHg concentration was 0.046 ng/L for the same traject as mentioned above for THg. This MeHg concentration is representative for ocean water up to a latitude of 45 °S and 1000 m of depth. However, whether cartilaginous fish species lived in shelf (<300 m depth), upper-slope (200-600 m depth) or mid-slope (600-2000 m depth),did not not seem to influence THg concentration (P=0.29). Thus, the assumption of a similar exposure concentration for all species, as is used below to calculate the BAFs seems not to be contradicted.
Very high log BAFs on a wet weight basis were calculated for in total 67 individuals of cartilaginous fish, ranging from 5.9 to 7.3, while the TLs range from 3.4 to 4.7. The log BAF values for the individual bony fish were 5.4 for Hector's lanternfish, 5.6, and 5.7 for cardinal fish. Of all BAFs considered in this report based on THg concentration in water, the highest 56 are for the cartilaginous fish. The difference between these BAFs and the rest is highly significant. No such difference was found for the remaining three fish species. Also the BAF relating the THg

concentration in fish to the MeHg concentration in water showed very high values for cartilaginous fish, ranging from 6.6 to 8.0. Although some of the highest BAFs here were for freshwater fish, these fish had also very high trophic levels. When regressed against trophic level, the BAFs for cartilaginous fish, were significantly higher than the rest. Therefore, the BAF values for the 67 cartilaginous fish are not further included in the assessments.
A series of TMF values on basis of wet weight THg concentrations were presented. The TMF for the food chain including zooplankton, crustaceans, all bony fish and squid groups was only 3.04. Similarly, the 16 shark and chimaeras showed a TMF of 2.84 and if only sharks were considered 4.84. However, a TMF of 13.4 was determined for the whole community including zooplankton and crustaceans, other fish and squid, and shark and chimaeras species, and a TMF of even 23.83 for other fish and squid, and shark species. This confirms the high accumulation in the cartilaginous fish, by showng the discrepancy between bony fish and cartilaginous fish in the food chain. Further TMFs were reported for the benthic food web (including chimaeras), the shelf/upper-slope food web, and the mid-slope food web, which were 7.70, 11.01, and 16.83, respectively. (Ri=2)

Lavoie et al. (2010) collected bird, fish, molluscs and crustacean samples at the east coast of Canada in the Gulf of St. Lawrence within a 60 km radius of Corossol Island, Canada. Fish samples were collected in August 2006. Invertebrate and water samples were taken in 2007. Birds were sampled in May and June 2006 and two additional herring gulls in 2007. Littoral and benthic macroinvertebrates, capelin (*Mallotus villosus*) and American sandlace (*Ammodytes americanus*) were collected from the surface near shore. Other fish and decapod samples were collected from a depth between 112 and 282 m and included American plaice (*Hippoglossoides platessoides*), witch flounder (*Glyptocephalus cynoglossus*) and Atlantic herring (*Clupea harengus*). Zooplankton was collected between 0 and 250 m depth. Concentrations in biota were reported in dry weight as well as wet weight. Water samples were taken at the surface, at 85 meter depth and 170 meter depth. Water concentrations for THg were explicitly mentioned to be based on unfiltered water samples. MeHg concentrations were measured after filtration on a glass fiber. The studied food web was an arctic marine environment and therefore, the concentration of suspended matter will be low. Consequently, the difference between total and dissolved concentrations is expected to be marginal.
BAFs for fish were calculated from the mean of the reported water concentrations at the three depths and reported dry or wet weight concentrations in biota. The values for log BAF for the five fish species range from 3.9 to 4.6 for THg on wet weight basis, and 4.8 to 5.5 for THg fish concentrations relative to MeHg water concentrations on wet weight basis, from 4.5 to 5.3 for THg on dry weight basis, and 5.4 to 6.2 for THg fish concentrations relative to MeHg water concentrations on dry weight basis. The water samples and biota samples for fish were taken in two consecutive years and it is considered unlikely that this will strongly influence the BAFs because of the large scale and remote area of the study location. The reported trophic levels are based on the mollusc *Tectura testudinalis* and cover a range of 3.4 to 4.2 for fish.

$TL = 2+(\delta^{15}N_{consumer} - \delta^{15}N_{mollusc})/3.4$.

Trophic levels for birds were calculate slightly differently assuming an enrichment in $\delta^{15}N$ of 2.6‰ in birds compared to their diets:

$TL = 3+(\delta^{15}N_{bird} - \delta^{15}N_{mollusc} – 2.6)/3.4$.

The TMF (sometimes referred to as food web magnification factor, FWMF) determined for the whole system is 3.8 for THg and 6.5 for MeHg based on a wet weight basis, and 2.9 for THg and 5.0 for MeHg based on a dry weight basis, calculated from the presented slopes on basis of trophic levels. (Ri = 2)

3.2 Correlation between water characteristics and bioaccumulation

Several studies reviewed for this report, mention an increase in biota concentrations with increasing DOC levels. Wiener et al. (2006) examined the influence of several parameters, including pH and total organic carbon (TOC), which was almost equal to DOC (Goldstein et al., 2003), on the THg-concentration in 1-year-old yellow perch collected in 17 different lakes in Voyageurs National Park (Minesota, USA). The best correlation was observed for a combination of the parameters TOC in water, pH and dissolved SO_4 (r^2 = 0.63). For TOC alone, r^2 was 0.48 and for pH alone r^2 was 0.37. An increase in TOC is correlated with an increase in the concentration in fish. Similarly, a positive correlation (r^2 = 0.33) between THg concentrations in 3-year-old yellow perch and DOC was shown for 15 different lakes in Vilas County (Wisconsin, USA) by Watras et al. (1998). Chasar et al. (2009) reported that mercury concentrations in the tissue of algae, daphnia and fish correlate strongly and positively to DOC concentrations in the water (p values < 0.0001). This relation is however not always observed. In a study into the uptake of mercury in fish, insects, crustacean and algae in 19 different arctic lakes, no correlation was found between DOC and THg-concentration in Arctic char (Gantner et al., 2010a). Also, from the data available in the paper of Evans et al. (2005), no clear trend could be observed between DOC level and mercury concentration in biota.

In studies where an increase in DOC is correlated with increased internal concentration is fish, an opposite trend seems to be occurring for the BAF. From the data obtained for Voyageurs National Park (Wiener et al., 2006) it appears that BAFs for THg decreased with increasing DOC. In the above mentioned study of Watras et al. (1998), the correlation between log BAF values for MeHg and DOC was also examined. For fish, zooplankton and microseston a negative correlation was found between DOC and log BAF (r^2 0.61, 0.70 and 0.64 respectively). Hall et al. (2009) also showed a significant ($p < 0.05$, r^2 = 0.24) negative correlation between log BAF values for MeHg in zooplankton and log DOC (µmol/L). The negative correlation between DOC and BAF was also shown by Rolfhus et al. (2011) for seston (r^2 = 0.46) and zooplankton (r^2 = 0.26). Gorski et al. (2003), only reported data for two DOC levels but the data showed a decline in the BAF value with increasing DOC levels.

Although the information presented above is not consistent, a mechanism for the relation between DOC and mercury uptake can be proposed. Higher DOC levels are correlated to higher mercury concentrations in water due to association with DOC (Dittman and Driscoll, 2009; Evans et al., 2005; Rolfhus et al., 2011). At the same time, warm water temperatures and high concentrations of labile organic matter are known to enhance microbial methylation of inorganic mercury, resulting in higher MeHg-levels (Chételat et al., 2014). Since in aquatic food chains uptake of MeHg is favoured over inorganic or elemental mercury (US-EPA, 1997), an increase in DOC concentration is related to higher mercury concentrations in biota (Lavoie et al., 2013). Although concentrations in biota are increasing with increasing DOC levels, the BAF seems to decrease with increasing DOC levels as reported by Watras et al. (1998), Hall et al. (2009) and Rolfhus et al. (2011). This could be explained by the fact that although THg and MeHg-concentrations are increasing with increasing DOC levels, MeHg is also adsorbed to DOC, which therefore reduces the overall bioavailability and uptake by fish. The data presented above suggest that the decrease in the bioavailable fraction with increasing DOC outweighs the increase in methylation of mercury, which then results in a decline in the BAF.

pH is another important parameter frequently examined. Watras et al. (1998) reported that higher pH values resulted in lower concentrations in fish (r^2 = 0.72) and reported a number of other parameters that influence the relationship between THg and MeHg, such as the presence of aluminum, copper, iron and other elements. Wiener et al. (2006) also showed that the best correlation for THg in 1-year-old yellow perch was observed for a combination of parameters where pH was included, in combination with dissolved sulfate and TOC.
Hall et al. (2009) showed an increase in BAF with increasing pH, whereas data from Monson and Brezonik (1998) show a decline in BAF and biota concentration with increasing pH for plankton in 12 different lakes. Other potential relevant parameters are: level dissolved SO_4 (Wiener et al., 2006); water shed or catchment area (Dittman and Driscoll, 2009; Gantner et al., 2010b; Gantner et al., 2010a); latitude (Lavoie et al., 2013); elevation (Dittman and Driscoll, 2009); fish body conditions (Dittman and Driscoll, 2009); chlorophyll-a concentration (Lavoie et al., 2013); and food web structure (Ferriss and Essington, 2014). Some of these parameters are linked with each other like lake area and DOC level (Evans et al., 2005) or pH and DOC level (Hall et al., 2009). In the end, the concentration in biota will be the net result of the complex water chemistry that determines the relative importance of MeHg as compared to THg and bioavailability of mercury species.

The number of parameters influencing the uptake of mercury is large and the information in our dataset is limited. Nevertheless DOC and pH are important parameters which are likely associated with part of the variation observed in BAF-levels. Salinity is another factor reported to influence mercury accumulation (Wang and Wang, 2010). A future challenge will be to further explore the correlations between these and other parameters in order to refine the BAF used here for different water types.

3.3 Correlation between BAF and trophic level

In order to select an appropriate BAF for the calculation of a water-based EQS for mercury, a regression was made of the available combinations of BAF and TL. This was done for on basis of THg concentrations in fish (because THg was always reported and MeHg not) and both THg and MeHg in water. The obtained correlations are thus suitable to extrapolate a concentration of THg in fish to an equivalent concentration of either THg or MeHg in water. Because no differences were observed, data for marine and freshwater fish were combined except for the data for cartilaginous fish (class Chondrichthyes), which were excluded from the regression.

In Figure 2, the correlation between BAFs for fish and their trophic level is plotted. It appears that the correlation for the BAF values based on MeHg in water is better than the correlation based on THg in water. This can be seen from the fact that for BAFs with THg as basis 9% of the BAF values lies more than 1 order of magnitude from the regression line, while for MeHg-based BAFs this is only 3%. A further observation is that the slope for BAFs based on MeHg (0.525±0.020) is much steeper than that for BAFs based on THg (0.279±0.016). There is no simple explanation for this observation, because both sets of BAFs are based on the same THg concentrations in fish and the same trophic levels for these fish, while only the concentration in water is a different one. However, the set for BAFs based on MeHg water concentrations (n=1516) is only a subset of the dataset for THg water concentrations (n=2370). Further, the dataset for THg is more scattered than that for MeHg. The increased slope might be just because of a better statistical fit.

If it is assumed that variability in the estimated trophic level is similar to that in the BAFs, then a better procedure might be to minimize the least squares of the perpendicular offsets from the line instead of the vertical offsets. Indeed, this yields two fits that have much more comparable and steeper slopes, i.e. 0.605 for BAFs based on THg in water and 0.882 for BAFs based on MeHg in water. These regression lines are plotted in Figure 2.

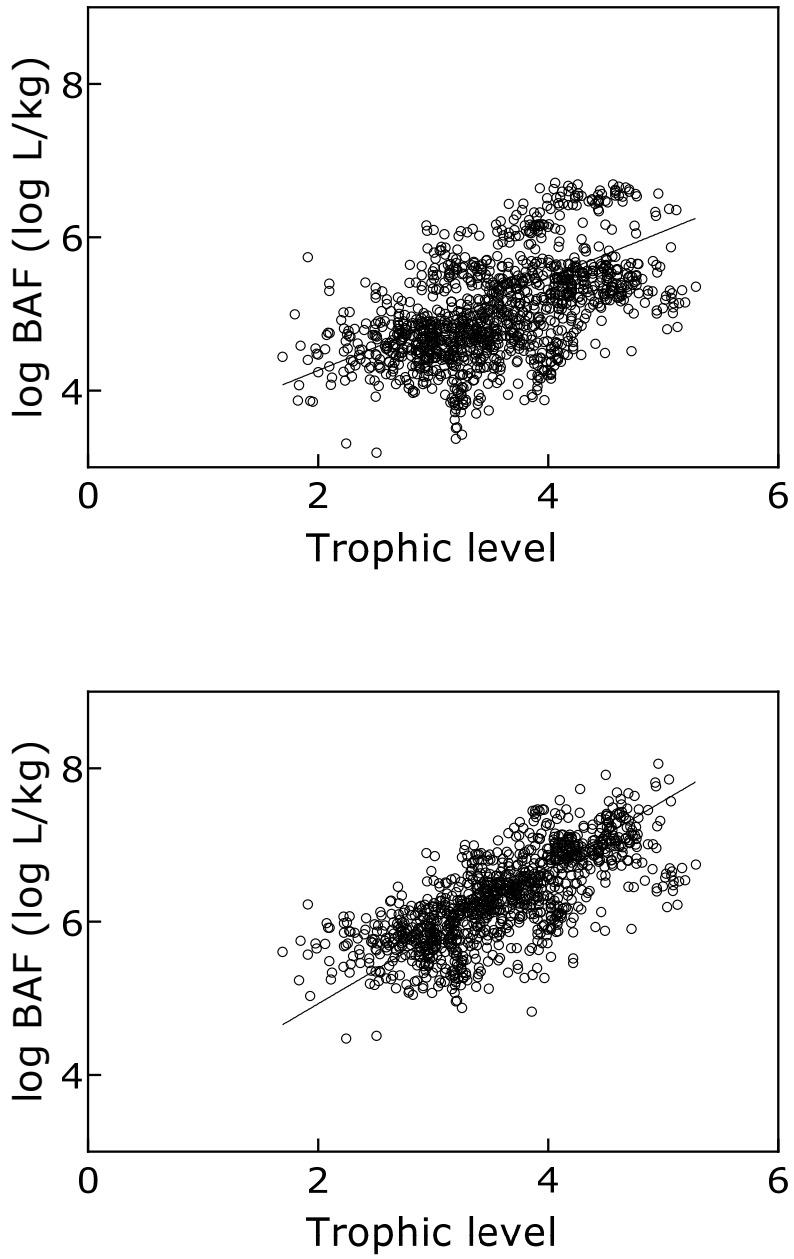

Figure 2 Correlation between BAFs and trophic level. BAFs are based on concentrations of THg in fish. The upper figure shows the correlation based on THg in water, the lower figure shows the correlation based on MeHg in water.

Almost all of TLs on which these slopes are based were calculated from an increase of 3.4‰ in $\delta^{15}N$ per trophic level, Cui et al. (2011) used a values of 3.8‰. On a $\delta^{15}N$ basis, the slopes are thus 3.4 times lower, i.e. 0.177 and 0.259, for THg and MeHg, respectively. These values are very comparable to the average value for the slope of 0.16 for THg from an analysis of 127 trophic magnification factors worldwide (Lavoie et al., 2013). The slope for 124 values of MeHg was on average 0.24 (Lavoie et

al., 2013). The higher TMF for MeHg can be explained by the increasing fraction of MeHg as compared to THg with increasing trophic level, thus increasing the TMF.

At trophic level 4, the log BAF from THg in water to THg in fish is 5.47, while the BAF from MeHg in water to THg in fish is 6.69. It should be noted that there is indeed a highly significant increase BAF with trophic level. In Figure 3 the data are binned according to trophic levels. Only the average BAF values of THg for the bins with trophic level <2.5 and from 2.5 to <3.5 are not significantly different. All other groups differ significantly with increasing BAF with trophic level. The average log BAF values for the bins with trophic level ranging from 3.5 to <4.5 are 5.27 for log BAF based on THg water concentrations and 6.54 for log BAF based on MeHg water concentrations, which is similar to the values derived from the regression.

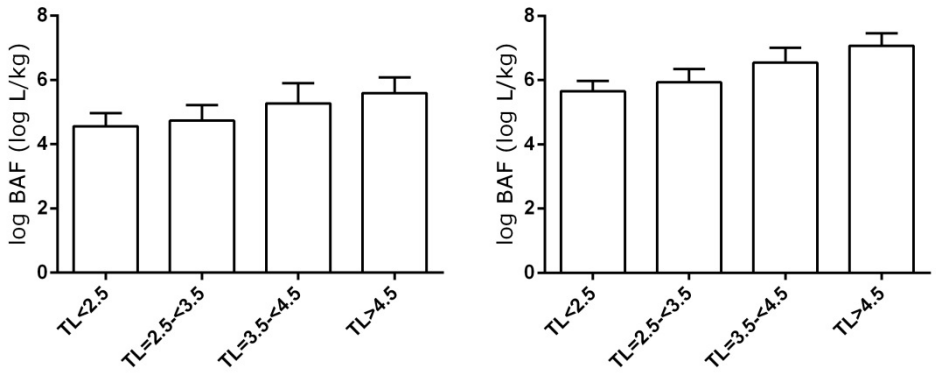

Figure 3 Statistical analysis of BAFs and trophic level. BAFs are based on concentrations of THg in fish. The left figure shows the BAF based on THg in water, the right figure shows the BAF based on MeHg in water.

3.4 Derivation of ERLs for mercury in water

The biota standard of 20 µg/kg_{wwt} as maintained in the new priority substances Directive 2013/39/EU (EU, 2013) remains as the $QS_{biota,\ secpois}$. To calculate this value into a water concentration, the above derived log BAF-values of 5.47 based on THg, and 6.69 based on MeHg are used according to Equation 7.
The $QS_{fw,\ secpois}$ for THg in freshwater = $QS_{biota,\ secpois}$ / BAF_{fw} = 20 / 10^5.47 = 6.7 x 10^{-5} µg/L =67 pg THg/L = 0.07 ng THg/L.
If this number is calculated for concentrations of MeHg in freshwater, the $QS_{fw,\ secpois}$ = $QS_{biota,\ secpois}$ / BAF_{fw} = 20 / 10^6.69 = 4.0 x 10^{-5} µg/L = 4.0 pg MeHg/L = 0.004 ng MeHg/L.
These values are derived using BAFs that are based on dissolved mercury concentrations including background levels, and should thus also be applied in that way. In view of the somewhat smaller variation in BAF-data, the value based on MeHg may be favoured from a scientific point of view. However, from the viewpoint of the current monitoring practice in which dissolved THg is measured, the proposed standard of 0.07 ng THg/L is considered to be most appropriate.

3.5 Implications of the new standards

An important observation for the translation of the biota standard to water is that in the more than 2000 fish that are the basis for the BAF values, the $QS_{biota,\ secpois}$ is exceeded in 92% of the cases. It should be noted that these fish were not from polluted areas, but to the contrary often from very remote areas worldwide. The dataset does not include studies from the Netherlands because no studies were available with simultaneous analysis of fish and water samples. However, data from Van Leeuwen et al. (2013) show that concentrations in fillet from Dutch North Sea fish fit into the observed pattern with minimum levels ranging from 30 $\mu g/kg_{wwt}$ for herring to 220 $\mu g/kg_{wwt}$ for dab. Although levels in fillet may not be fully comparable to those in whole fish, these data suggest that the biota standard will be exceeded. This is also the case for the equivalent water-based standard. In 2013, annual average concentrations of dissolved mercury at drinking water intake locations in the Netherlands ranged from 0.53 to 0.66 ng/L, individual measurements ranged from <0.3 to 2.9 ng/L (RIWA, 2014). A similar range of <0.2 – 2.7 ng/L is reported for various locations in Dutch surface waters in 2013 (data from Waterbase[2]). In over 60% of the cases, concentrations were lower than the current limit of determination (LOD) of 0.2-1 ng/L. Since the proposed standards are lower than the LOD, non-detection may still mean that the standard is exceeded. It can thus be foreseen that the derived quality standard for water will often be exceeded.

The proposed values are also lower than the currently used background concentrations for mercury in Dutch surface waters. These values are 10 ng/L for both inorganic and organic mercury in freshwater. For marine waters, the background concentration for inorganic mercury is set at 3 ng/L[3]. It should be noted that for mercury, being an anthropogenic contaminant, a background does not represent a naturally occurring level, but rather points at the worldwide presence of contamination. It can be argued that the currently used Dutch background concentrations, which data back to the late 1990s, are (much) too high. They are based on monitoring data of total metals in unpolluted areas, which were converted to dissolved concentrations using a generic partition coefficient and a suspended matter concentration of 30 mg/L (Osté, 2013). In 2013, new background concentrations were derived for a number of metals by taking the 10^{th} percentile of recent monitoring data. For mercury, it was concluded that the 10^{th} percentile of dissolved concentrations would be < 1 ng/L, but a definitive value could not be set due to the high number of samples with concentrations below the limit of detection (Osté, 2013). These data confirm that the majority of concentrations in Dutch surface waters will likely be higher than the proposed standards.

In view of this, it may be questioned if the biota standard of 20 $\mu g/kg_{wwt}$ is adequate. OSPAR's Ecological Assessment Criterion for mercury in fish is set higher at 35 $\mu g/kg_{wwt}$ (OSPAR, 2009). It should be noted, however, that the OSPAR EAC reflects a Background Assessment

[2] http://live.waterbase.nl/
[3] www.stoffen-risico.nl

Concentration (BAC). The BAC is a statistically derived value that is based on a time series of monitoring data and represents the concentration at which it is possible to conclude with high probability that measured levels are near background (OSPAR, 2008). Given the fact that mercury is a compound with long-term widespread occurrence, these background levels are not necessarily related to no-effect levels.

The biota standard of 20 µg/kg_{wwt} is derived by putting an assessment factor of 10 on the lowest dietary NOEC for mammals of 0.22 mg/kg_{wwt} (EC, 2005). Data are taken from previous reviews by RIVM and underlying literature dates back to the late 1980's. A lot of relevant studies have been published since then, and an update of the literature might be considered. In that case, a more refined biota standard could be obtained using the recently published energy-based approach developed by the Netherlands (Verbruggen, 2014). Most likely enough data will be available to perform statistical extrapolation, but it is questionable if this leads to a substantially higher value. Based on an extensive review of laboratory and field studies, Depew et al. (2012) showed that the NOAEL for the Common loon (*Gavia immer*, *ijsduiker* in Dutch) is around 20 to 30 µg/kg fish. This value for a single species is thus similar to the current generic WFD biota standard, which should protect all birds. It should also be noted that even if a higher biota standard would be used, this standard will likely still be exceeded in a large number of fish and water samples worldwide.
Mercury in fish is also an issue from a human health perspective. The US EPA very actively communicates about the risks of mercury exposure via consumption of fish, and advices against consumption of certain species[4]. In 2010, the European Food Safety Authority concluded that the mean dietary exposure of methylmercury across age groups does not exceed the tolerable weekly intake (TWI), with the exception of toddlers and other children in some surveys. However, high fish consumers may exceed the TWI (EFSA, 2010).
Taken this information together, it seems unlikely that a refined biota standard would be much higher than the current one.

[4] see http://www.epa.gov/mercury/

4 Summary and conclusions

Within the context of the Water Framework Directive, a biota standard for mercury is set at 20 µg/kg_{wwt} in Directive 2013/39/EU. This value represents a concentration in fish at which birds and mammals are protected against effects of mercury via secondary poisoning. However, compliance checking by means of monitoring in water has advantages over biota sampling in terms of reproducibility, costs and uniformity of sampling. Therefore, the biota standard for mercury was converted into a water-based quality standard that offers the same level of protection.

To that end, the relationship between concentrations in water and biota was investigated in this report. Bioaccumulation factors (BAFs) were derived for fish representing different trophic levels. The data show that bioaccumulation is positively correlated with trophic position. This correlation was used to establish BAFs for larger fish at the trophic level that is representative of the protection goals aimed for by the biota standard, i.e. marine and freshwater predators and humans.

Log BAF-values based on dissolved total mercury and methylmercury are 5.47 and 6.69, respectively. Using these values, water-based quality standards are proposed of 0.07 ng/L for the sum of all dissolved mercury species (i.e. total mercury in filtered samples), and 0.004 ng/L for dissolved methylmercury. Monitoring data indicate that these levels are likely to be exceeded frequently. This is not surprising, given the fact that the biota standard was also exceeded in over 90% of the fish included in the present evaluation, including all trophic levels and very remote areas worldwide.

Acknowledgements

The authors thank the following people for providing us with valuable information additionally to their publications:

Kristofer Rolfhus, Mark Sandheinrich and James Wiener from the University of Wisconsin-La Crosse, River Studies Center, La Crosse, WI, United States,
Mark Brigham from U.S. Geological Survey (USGS), Minnesota Water Science Center, Mounds View, MN, USA,
Nikolaus Gantner from University of Northern British Columbia, Prince George, BC, Canada,
Robin Stewart from U.S. Geological Survey (USGS), Water Resources Division, Menlo Park, CA, USA, and
Raphaël Lavoie of Queen's University, Department of Biology, Kingston, ON, Canada.

The members of the Scientific Advisory Group for standard setting in water and air (*Wetenschappelijke Klankbordgroep Normstelling water en lucht*) are gratefully acknowledged for reviewing the draft report.

References

Alpers CN, Stewart AR, Saiki MK, Marvin-DiPasquale MC, Topping BR, Rider KM, Gallanthine SK, Kester CA, Rye RO, Antweiler RC, De Wild JF. 2008. Environmental Factors Affecting Mercury in Camp Far West Reservoir, California, 2001–03. Sacramento, CA, USA: U.S. Geological Survey. Report nr. U.S. Geological Survey Scientific Investigations Report 2006–5008. Survey USG.

Apte SC, K.C. Bowles KC, W.A. Maher WA, Smith REW. 2000. Mercury cycling in Lake Murray, Papua New Guinea. CSIRO, Centre for Advanced Analytical Chemistry. Report nr. ET/IR301.

Back RC, Gorski PR, Cleckner LB, Hurley JP. 2003. Mercury content and speciation in the plankton and benthos of Lake Superior. Sci Total Env 304: 349-354.

Borgå K, Kidd KA, Muir DCG, Berglund O, Conder JM, Gobas FAPC, Kucklick J, Malm O, Powellkk DE. 2012. Trophic magnification factors: Considerations of ecology, ecosystems, and study design. Integrated Environmental Assessment and Management 8 (1): 64-84.

Bowles KC, Apte SC, Maher WA, Kawei M, Smith R. 2001. Bioaccumulation and biomagnification of mercury in Lake Murray, Papua New Guinea. Can J Fish Aquat Sci 58: 888-897.

Burkhard LP, Borgå K, Powell DE, Leonards P, Muir DCG, Parkerton TF, Woodburn KB. 2013. Improving the quality and scientific understanding of Trophic Magnification Factors (TMFs). Environ Sci Technol 47: 1186−1187.

Campbell LM, Hecky RE, Nyaundi J, Muggide R, Dixon DG. 2003a. Distribution and food-web transfer of mercury in Napoleon and Winam Gulfs, Lake Victoria, East Africa. Journal of Great Lakes Research 29 (SUPPL. 2): 267-282.

Campbell LM, Hecky RE, Wandera SB. 2003b. Stable isotope analyses of food web structure and fish diet in Napoleon and Winam gulfs, Lake Victoria, East Africa. Journal of Great Lakes Research 29 (SUPPL. 2): 243-257.

Chasar LC, Scudder BC, Bell AH, Wentz DA, Brigham ME. 2008. Total Mercury, Methylmercury, and Carbon and Nitrogen Stable Isotope Data for Biota from Selected Streams in Oregon, Wisconsin, and Florida, 2002–04. Reston, VA, USA U.S. Department of the Interior, U.S. Geological Survey.

Chasar LC, Scudder BC, Stewart AR, Bell AH, Aiken GR. 2009. Mercury cycling in stream ecosystems. 3. Trophic dynamics and methylmercury bioaccumulation. Environmental Science and Technology 43 (8): 2733-2739.

Cheng Z, Liang P, Shao D-D, Wu S-C, Nie X-P, Chen K-C, Li K-B, Wong M-H. 2011. Mercury biomagnification in the aquaculture pond ecosystem in the Pearl River Delta. Arch Environ Contam Toxicol 61 (3): 491-499.

Chételat J, Amyot M, Arp P, Blais JM, Depew D, Emmerton CA, Evans M, Gamberg M, Gantner N, Girard C, Graydon J, Kirk J, Lean D, Lehnherr I, Muir D, Nasr M, J. Poulain A, Power M, Roach P, Stern G, Swanson H, van der Velden S. 2014. Mercury in freshwater ecosystems of the Canadian Arctic: Recent advances on its cycling and fate. Science of the Total Environment.

Cossa D, Heimbürger LE, Lannuzel D, Rintoul SR, Butler ECV, Bowie AR, Averty B, Watson RJ, Remenyi T. 2011. Mercury in the Southern Ocean. Geochimica et Cosmochimica Acta 75 (14): 4037-4052.

Cui B, Zhang Q, Zhang K, Liu X, Zhang H. 2011. Analyzing trophic transfer of heavy metals for food webs in the newly-formed wetlands of the Yellow River Delta, China. Environ Pollut 159: 1297-1306.

De Wit HA, Kainz MJ, Lindholm M. 2012. Methylmercury bioaccumulation in invertebrates of boreal streams in Norway: Effects of aqueous methylmercury and diet retention. Environmental Pollution 164: 235-241.

Depew DC, Basu N, Burgess NM, Campbell LM, Evers DC, Grasman KA, Scheuhammer AM. 2012. Derivation of screening benchmarks for dietary methylmercury exposure for the common loon (Gavia immer): Rationale for use in ecological risk assessment. Environ Toxicol Chem 31 (10): 2399-2407.

Dittman JA, Driscoll CT. 2009. Factors influencing changes in mercury concentrations in lake water and yellow perch (Perca flavescens) in Adirondack lakes. Biogeochemistry 93 (3): 179-196.

Doetzel LM. 2007. An investigation of the factors affecting mercury accumulation in lake trout, *Salvelinus namaycush*, in Northern Canada. Saskatoon, Canada, University of Saskatchewan.

Dominique Y, Maury-Brachet R, Muresan B, Vigouroux R, Richard S, Cossa D, Mariotti A, Boudou A. 2007. Biofilm and mercury availability as key factors for mercury accumulation in fish (Curimata cyprinoides) from a disturbed Amazonian freshwater system. Environ Toxicol Chem 26 (1): 45-52.

EC. 2005. Common Implementation Strategy for the Water Framework Directive. Environmental Quality Standards (EQS) Substance Data Sheet. Priority Substance No. 21 Mercury and its Compounds CAS-No. 7439-97-6. Final version. Brussels, 15 January 2005.

EC. 2008. Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European Parliament and of the Council. http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32008L0105:EN:NOT.

EC. 2011. Common implementation strategy for the Water framework Directive (2000/60/EC) - Guidance Document No. 27 - Technical guidance for deriving environmental qualilty standards. Brussel: Europese Commissie. Report nr. 2011-055.

ECHA. 2012. Guidance on information requirements and chemical safety assessment. Chapter R.7c: Endpoint specific guidance. Version 1.1. Helsinki, Finland: European Chemicals Agency. Report nr. ECHA-12-G-23-EN.

EFSA. 2009. Guidance of EFSA. Risk Assessment for Birds and Mammals. On request from EFSA, Question No EFSA-Q-2009-00223. First published on 17 December 2009. EFSA Journal 7 (12): 1438.

EFSA. 2010. Scientific Opinion on the risk for public health related to the presence of mercury and methylmercury in food. EFSA Panel on Contaminants in the Food Chain (CONTAM). EFSA Journal 10 (12): 2985.

EU. 2013. Directive 2013/39/EU of the European Parliament and of the Council of of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2013:226:0001:0017:EN:PDF.

Evans MS, Lockhart WL, Doetzel L, Low G, Muir D, Kidd K, Stephens G, Delaronde J. 2005. Elevated mercury concentrations in fish in lakes in the Mackenzie River Basin: The role of physical, chemical, and biological factors. Science of the Total Environment 351-352: 479-500.

Ferriss BE, Essington TE. 2014. Does trophic structure dictate mercury concentrations in top predators? A comparative analysis of pelagic food webs in the Pacific Ocean. Ecological Modelling 278: 18-28.

Gantner N, Muir DC, Power M, Iqaluk D, Reist JD, Babaluk JA, Meili M, Borg H, Hammar J, Michaud W, Dempson B, Solomon KR. 2010a. Mercury concentrations in landlocked arctic char (Salvelinus alpinus) from the Canadian arctic. Part II: Influence of lake biotic and abiotic characteristics on geographic trends in 27 populations. Environ Toxicol Chem 29 (3): 633-643.

Gantner N, Power M, Iqaluk D, Meili M, Borg H, Sundbom M, Solomon KR, Lawson G, Muir DC. 2010b. Mercury concentrations in landlocked arctic char (Salvelinus alpinus) from the Canadian arctic. Part I: Insights from trophic relationships in 18 lakes. Environ Toxicol Chem 29 (3): 621-632.

Goldstein RM, Brigham ME, Steuwe L, Menheer MA. 2003. Mercury Data from Small Lakes in Voyageurs National Park, Northern Minnesota, 2000–02. Mounds View, MN, USA: U.S. Geological Survey. Report nr. Open-File Report 03–480. Survey USG.

Gorski PR, Cleckner LB, Hurley JP, Sierszen ME, Armstrong DE. 2003. Factors affecting enhanced mercury bioaccumulation in inland lakes of Isle Royale National Park, USA. Science of the Total Environment 304 (1-3): 327-348.

Hall BD, Cherewyk KA, Paterson MJ, Bodaly RA. 2009. Changes in methyl mercury concentrations in zooplankton from four experimental reservoirs with differing amounts of carbon in the flooded catchments. Canadian Journal of Fisheries and Aquatic Sciences 66 (11): 1910-1919.

Herrin RT, Lathrop RC, Gorski PR, Andren AW. 1998. Hypolimnetic methylmercury and its uptake by plankton during fall destratification: A key entry point of mercury into lake food chains? Limnology and Oceanography 43 (7): 1476-1486.

Jardine TD, Kidd KA, Fisk AT. 2006. Applications, considerations, and sources of uncertainty when using stable isotope analysis in ecotoxicology. Environ sci Technol 40 (24): 7501-7511.

Jones RB. 1998. TechDig (computer program). Version 2.0d. Mundelein, USA,

Kim E, Kim H, Shin K-H, Kundu SR, Lee B-G, Han S. 2012. Biomagnification of mercury through the benthic food webs of a temperate estuary: Masan Bay, Korea. Environ Toxicol Chem 31 (6): 1254-1263.

Klimisch H-J, Andreae M, Tillman U. 1997. A systematic approach for evaluating the quality of experimental toxicological and ecotoxicological data. Regulatory Toxicology and Pharmacology 25 (1): 1-5.

Lavoie RA, Hebert CE, Rail J-F, Braune BM, Yumvihoze E, Hill LG, Lean DRS. 2010. Trophic structure and mercury distribution in a Gulf of St. Lawrence (Canada) food web using stable isotope analysis. Sci Total Env 408 (22): 5529-5539.

Lavoie RA, Jardine TD, Chumchal MM, Kidd KA, Campbell LM. 2013. Biomagnification of mercury in aquatic food webs: A worldwide meta-analysis. Environ Sci Technol 47 (23): 13385-13394.

Moermond CTA, Verbruggen EMJ. 2012. An evaluation of bioaccumulation data for hexachlorobenzene to derive water quality standards according to the EU-WFD methodology. IEAM 9 (1): 87-97.

Monson BA, Brezonik PL. 1998. Seasonal patterns of mercury species in water and plankton from softwater lakes in Northeastern Minnesota. Biogeochemistry 40 (2-3): 147-162.

OSPAR. 2008. CEMP Assessment Manual Co-ordinated Environmental Monitoring Programme Assessment Manual for contaminants in sediment and biota.

OSPAR. 2009. CEMP assessment report: 2008/2009 Assessment of trends and concentrations of selected hazardous substances in sediments and biota.

Osté L. 2013. Derivation of dissolved background concentrations in Dutch surface water based on a 10th percentile of monitoring data. Utrecht, the Netherlands: Deltares. Report nr. 1206111.005-2.

Paterson MJ, Rudd JWM, St. Louis V. 1998. Increases in total and methylmercury in zooplankton following flooding of a peatland reservoir. Environmental Science and Technology 32 (24): 3868-3874.

Pethybridge H, Butler ECV, Cossa D, Daley R, Boudo A. 2012. Trophic structure and biomagnification of mercury in an assemblage of deepwater chondrichthyans from southeastern Australia. Mar Ecol Prog Ser 451: 163-174.

Poste AE, Muir DCG, Mbabazi D, Hecky RE. 2012. Food web structure and mercury trophodynamics in two contrasting embayments in northern Lake Victoria. J Great Lakes Res 38: 699-707.

RIWA. 2014. Jaarrapport 2013. De Rijn. Nieuwegein, the Netherlands: RIWA-Rijn, Vereniging van Rivierwaterbedrijven.

Rolfhus KR, Hall BD, Monson BA, Paterson MJ, Jeremiason JD. 2011. Assessment of mercury bioaccumulation within the pelagic food web of lakes in the western Great Lakes region. Ecotoxicology 20 (7): 1520-1529.

Romijn CAFM, Luttik R, Van de Meent D, Slooff W, Canton JH. 1991. Presentation of a general algorithm for effect-assessment on secondary poisoning. Bilthoven, the Netherlands: National Institute for Public Health and the Environment. Report nr. 679102002.

Saiki MK, Martin BA, May TW, Alpers CN. 2010. Mercury concentrations in fish from a Sierra Nevada foothill reservoir located downstream from historic gold-mining operations. Environmental Monitoring and Assessment 163 (1-4): 313-326.

Slooff W, Van Beelen P, Annema JA, Janus JA. 1995. Integrated criteria document mercury. Bilthoven, the Netherlands: National Institute for Public Health and the Environment. Report nr. 601014008.

Smit CE. 2005. Energy and moisture content and assimilation efficiency of bird and mammal food. Bilthoven, the Netherlands: National Institute for Public Health and the Environment. Report nr. 601516013.

Smit CE, Van Wezel AP, Jager T, Traas TP. 2000. Secondary poisoning of cadmium, copper and mercury: implications for the Maximum Permissible Concentrations in water, sediment and soil. Bilthoven, the Netherland: National Institute for Publich Health and the Environment. Report nr. 601501009.

Stewart RA, Saiki MK, Kuwabara JS, Alpers CN, Marvin-DiPasquale M, Krabbenhoft DP. 2008. Influence of plankton mercury dynamics and trophic pathways on mercury concentrations of top predator fish of a mining-impacted reservoir. Can J Fish Aquat Sci 65: 2351-2366.

US-EPA. 1997. Mercury Study Report to Congress - Volume III: Fate and Transport of Mercury in the Environment. US-EPA. Report nr. EPA-452/R-97-005.

Van Duijnhoven N. 2011. Inzicht in landelijke kwaliteitsdata 2006-2009. Achtergronddocument t.b.v. update stoffenlijst MR monitoring. Utrecht, the Netherlands: Deltares.

Van Leeuwen SPJ, Van der Roest J, Van der Lee MK, Hoogenboom LAP. 2013. Contaminanten in vis en schaaldieren uit de Noordzee. Wageningen, the Netherlands: RIKILT Wageningen UR (University & Research centre). Report nr. 2013.011.

Vander Zanden MJ, Cabana G, Rasmussen JB. 1997. Comparing trophic position of freshwater fish calculated using stable nitrogen isotope ratios (δ15N) and literature dietary data. Can J Fish Aquat Sci 54: 1142-1158.

Verbruggen EM, J. 2014. New method for the derivation of risk limits for secondary poisoning. Bilthoven, the Netherlands: National Institute for Public Health and the Environment (in press).

Wang R, Wang W-X. 2010. Importance of speciation in understanding mercury bioaccumulation in Tilapia controlled by salinity and dissolved organic matter. Environ Sci Technol 44 (20): 7964-7969.

Watanabe K, Monaghan MT, Takemon Y, Omuraa T. 2008. Biodilution of heavy metals in a stream macroinvertebrate food web: Evidence from stable isotope analysis. Sci Total Env 394: 57-67.

Watras CJ, Back RC, Halvorsen S, Hudson RJM, Morrison KA, Wente SP. 1998. Bioaccumulation of mercury in pelagic freshwater food webs. Science of the Total Environment 219 (2-3): 183-208.

Wiener JG, Knights BC, Sandheinrich MB, Jeremiason JD, Brigham ME, Engstrom DR, Woodruff LG, Cannon WF, Balogh SJ. 2006. Mercury in soils, lakes, and fish in Voyageurs National Park (Minnesota): Importance of atmospheric deposition and ecosystem factors. Environmental Science and Technology 40 (20): 6261-6268.

RIVM
Committed to *health and sustainability*