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AMAP Workshop on Statistical Analysis of Temporal Trends of Mercury in Arctic Biota

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Report of the workshop, Swedish Museum of Natural History, Stockholm, 30 October- 3 November 2006

AMAP
Arctic Monitoring and Assessment Programme

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AMAP Workshop on Statistical Analysis of Temporal Trends of Mercury in Biota

Swedish Museum of Natural History, Stockholm

30 October - 3 November

Participants:

Canada: Birgit Braune, Marlene Evans, Lisa Loseto, Peter Outridge*, Jason Stow; Denmark (Greenland/Faroe Islands): Frank Rigét, Rune Dietz*; Finland: Matti Verta; Iceland: Eva Yngvadottir; Norway: Norman Green; Sweden: Anders Bignert; United Kingdom: Rob Fryer; AMAP: Simon Wilson

*meeting co-chairs (co-chairs of the AMAP Mercury Expert Group)

1 Opening of the meeting

Peter Outridge and Rune Dietz opened the meeting at 09:00 on 30 October 2006. Anders Bignert welcomed the participants (Annex 1) to the Swedish Museum of Natural History.

The workshop was viewed as a continuation of activities that were initiated at a workshop on the same topic held in November 2001 in Copenhagen as part of the AMAP (2002) heavy metals assessment process. An important development at the 2006 workshop, compared with the previous workshop, was the participation of a number of experts familiar with the datasets, as well as statistical experts. Another feature of the workshop was to focus greater attention on the analysis of co-variables such as biological parameters (age, sex, length, weight), and to consider new types of information, such as data on stable isotopes, which can provide additional insight into other factors that may influence trends such as changes in feeding behaviour, etc. Other potentially important factors that may affect trend interpretation, such as climate change/variability, were also covered in the discussions.

2 Main Objectives of the Workshop

The main objective of the workshop was to statistically analyse all available data concerning ‘short-term’ temporal trends (i.e. trends over the past few decades) of mercury in Arctic biota, in a statistically valid and consistent manner. The workshop also took note of some new century-scale datasets that are available, which could provide a context for some of the shorter-term datasets.

In addition, the results of the workshop would be used as a basis for recommendations concerning possible adjustments to the AMAP monitoring programme with respect to determination of temporal trends.

This report present results of the statistical workshop and discusses methodological issues rather than the trend results as such. Conclusions regarding temporal trends will be discussed in detail in future reports updating the AMAP assessment of Mercury in the Arctic.

3 Adoption of the Agenda and Organisation of the Work

The agenda distributed prior to the meeting (Annex 2) was discussed and accepted. It was agreed that the workshop would be arranged in an informal atmosphere, to provide opportunities for working in small break-out groups on specific issues and/or datasets.

3.1 Workshop Data Policy

The workshop data policy (Annex 3) was reviewed to ensure that all participants understood and confirmed (in writing) their agreement to the conditions outlined in the policy.

4 Workshop Materials and Methodology

4.1 Available Data and Preliminary Analyses

Simon Wilson presented an overview of the datasets available for consideration at the meeting: A total of 106 time-series (including 5 methyl-mercury time-series) with greater than or equal to 4 (not necessarily consecutive) years of data had been received prior to the meeting from Canada (56), Faroe Islands (4), Greenland (13), Iceland (21), Norway (6), and Sweden (6). These included mercury time-series in various tissues and organs of a range of species (including shellfish, freshwater and marine fish, marine mammals, terrestrial mammals, and seabirds). No time series were available from Alaska or Russia. The time-series covered periods ranging from 4 to 44 years (average 15.3 years; with data available for an average of 8.1 years within those periods). However, within this overall pattern, datasets were generally characterised either by series based on annual sampling, or by non-contiguous series of samples representing different 'time periods' with gaps of several years between them.

All datasets received prior to the workshop were subject to preliminary analyses, based on a regression of (log)concentration values against year, with no consideration of (or adjustment for) possible covariates. Results of the preliminary analyses were distributed for comment to all workshop participants and to the respective data providers prior to the meeting. The preliminary analyses were performed using the PIAW statistical application (see below). Details of this application can be found in Annex 4.

Additional time-series datasets, including 11 time-series from Finland and further series from Canada and Greenland were delivered at the meeting and included in the analyses completed during the workshop, resulting in a total of 138 time-series available for statistical analyses.

All time-series were given a unique identifying number consisting of a country code and sequence number (e.g. SWE-003). Where mercury was analysed in different tissues or organs of the same animals, the resulting datasets were treated as separate time series with different (but sequential) ID numbers; for example, CAN-011, CAN-012, … CAN-017 refer to different tissue-component time-series in beluga from Hendrickson's Island, Northwest Territories, Canada. In some cases, statistical analyses were re-run on the same 'time series' but with concentrations expressed on a different basis (e.g. dry weight basis vs. wet weight basis), or with additional years of data included/excluded, or run for some other defined sub-set of the dataset (e.g. large and small fish within the same time-series). In such cases a qualifying letter and/or number was added to the time-series ID to distinguish the run.

Updating of time-series

Based on comments received to the results of the preliminary analyses conducted prior to the meeting, a number of the time-series were updated during the workshop. In some cases, additional years of data or additional data on covariates were added, whereas in other cases

certain data were eliminated from the series to exclude 'incompatible' samples/years (e.g. where samples were collected at different locations in particular years). During the workshop the PIA and CTSM routines (see below) were run on the updated datasets. The results of these runs formed the basis for workshop discussions and conclusions. Following the workshop, all results were re-checked and in a few cases corrected results have been included in the final version of the workshop report.

4.2 Statistical Applications

Two statistical applications were available for use at the meeting:

- the (PIA) application developed for trend assessment of AMAP time-series data by Anders Bignert, and
- a collection of routines (CTSM) developed by Rob Fryer that have been applied in the temporal trend assessment work conducted under Oslo and Paris Commission (OSPAR).

Although based on a similar statistical method for trend detection (described in Nicholson et al., 1998), the two applications involve slightly different analyses:

(1) PIA

PIA is a custom application based on Fortran code and developed to run on Windows systems. It applies a trend assessment on an 'index' value (by default, the annual geometric mean concentration computed from an input set of concentration values for different years) and employs a 3-year running-mean smoother to test for non-linear trend components (as described in Bignert, 2001; see Annex 4). In addition to the statistical results, the application output includes a trend plot and a simplified 'trend interpretation'. A directive file specifies options for the analysis to be conducted, including options to vary the power and alpha values used in the statistical tests, and to apply an analysis based on median rather than geometric mean index values. During the workshop, the application was further developed to allow it to (optionally) employ a loess smoother with a defined window (for comparison with the CTSM methodology), and to analyze trends in concentrations adjusted for a selected covariate.

Test versions of the PIAW application were distributed to workshop participants prior to and during the meeting for them to use/test. An updated version of the application was prepared taking into account comments/suggestions received, and this is available from AMAP (instructions).

(2) CTSM

The CTSM routines are developed in the 'S' and/or 'R' statistical languages and can be run on systems with these software applications installed. The basic CTSM routines evaluate trends in annual 'index' values (typically median-log concentrations or geometric means) in a given year, employ a loess smoother with a specified (or variable) window, and include options for filling in missing values, among other features. They also include extensions for comparing observed levels with 'background' concentrations, weighting index values to take into account e.g. analytical quality assurance considerations, and other techniques used in the OSPAR trend assessments. For more information see Annex 5. The CTSM routines were introduced to workshop participants and used in the workshop to conduct statistical runs on covariate adjusted datasets (see below), due to the ability of the software to 'batch process' a large number of datasets simultaneously. They were also further developed during the workshop to allow more complex linear mixed models to be applied to selected time-series datasets.

4.3 Statistical Screening for Covariates Effects

Contaminant levels in biota often vary with some covariate, such as animal age/size (e.g. Dietz et al. 1998, Braune et al. 2005). Assessments of temporal trends in contaminant levels must therefore take into account changes in contaminant levels that are due to the covariate, or changes in the covariate effect over time.

An initial overview of the covariates available for the time-series under consideration yielded the following general patterns:

- A number of time-series purposely restricted sampling to eliminate or control for biological covariates (e.g. age/sex specific sample collection; length-stratified sampling).
- Typically, time-series included covariates only for incomplete sub-sets of the data, i.e. covariate measurements were missing for some years or for some samples. Therefore, adjustment for covariates would mean that in many cases the fundamental structure of the underlying time-series was altered (reduced number of years, selective exclusion of data-points with no associated covariate measurements, etc.).
- Available covariates included biological parameters (age, length, weigh, sex, etc.), measures of trophic level and feeding behaviour (stable isotopes: $\delta^{15}N$, $\delta^{13}C$), and measures reflecting moisture or lipid content of tissues and organs (concentrations expressed on a particular basis, % moisture, % lipids, etc.). Availability of covariate data differed between species groups monitored.

Most of the mercury time-series available at the workshop included several covariates of interest, so parametric methods, particularly analysis of covariance and linear mixed models, were employed to investigate:

- the relationship between mercury concentrations and the covariates
- the effect of covariate-adjustment on temporal trend assessments of mercury concentrations

As an initial step, a quick screening of data sets to identify significant covariates was performed. This screening focussed on those datasets where covariate effects were potentially important (for biological parameters: fish and mammals, unless the sampling programmes involved restricted animals to a specific size/age/sex; for stable isotopes: freshwater fish, marine mammals, and seabird egg time-series)

The purpose of this analysis was to identify those covariates that were always important and those which could be ignored. For each time series and covariate, the linear model:

log mercury concentration ~ year + covariate + year * covariate

was fitted. Year was treated as a factor, so the model fitted a separate slope and intercept for each year. Length and weight were log transformed and age was square root transformed. The significance of the following relationships were tablualted:

- covariate main effect (i.e. does the covariate affect mercury concentrations)
- the interaction between year and the covariate (i.e. is the relationship consistent across time).

Note that we did not fit several covariates at once, so the effect of some covariates might be masked. For example, the significance of a stable isotope might emerge only when concentrations have first been adjusted for age. This was addressed in the third set of analyses using a linear mixed model, described below.

Results of the covariate screening tests are presented in Annex 6.

4.4 Trend Analyses

Trend analysis of time-series with no significant covariate effects

Unadjusted runs (i.e. statistical analyses of trends that ignored possible covariates) were conducted on all updated time series datasets, including additional time series delivered at the meeting, using the PIAW application. Analyses were also repeated on some time series (e.g. Icelandic mussel datasets) where runs were conducted for concentrations expressed on a dry weight basis.

Trend analysis of time-series with significant covariate effects

There are many ways of taking covariates into account in the statistical analyses (see the introduction to Fryer & Nicholson, 2002). For the purposes of the workshop, two methods were employed:

(1) Assessing covariate-adjusted concentrations for temporal trends

Mercury concentrations in fish and mammals are typically related to length, weight and/or age (i.e. some measure of duration of exposure). The exceptions are typically those time series where individuals have been sampled in a restricted size or age range (in order to avoid the effect of the covariate). We therefore considered the effect on temporal trend assessments of adjusting mercury concentrations in fish for length and in mammals for age.

When there was no interaction between year and covariate, the linear model:

log mercury concentration ~ year + covariate

was fitted, and log mercury concentrations were adjusted to the median value of the covariate across the whole time series. (Again, year was treated as a factor, length was log transformed, and age was square root transformed). The annual mean size-adjusted log mercury concentrations were then assessed for temporal trends using the standard methodology.

When there was an interaction between year and covariate, the linear model:

log mercury concentration ~ year + covariate + year * covariate

was fitted and log mercury concentrations adjusted first to the lower (25%) quartile value of the covariate (i.e. small or young individuals) and then to the upper (75%) quartile value of the covariate (i.e. large or old individuals). Temporal trends in the annual mean size-adjusted log mercury concentrations were then assessed for both small/young individuals and large/old individuals using the standard methodology.

(2) More detailed modelling of covariates

For selected time series, the effect of multiple covariates on log mercury concentrations was investigated using linear mixed models. The fixed and random components of the model were:

fixed ~ s(year) + covariate1 + covariate2 + …

*random ~ year + year*covariate1 + year * covariate2 + …*

In the fixed component, s(year) denotes that both a smooth and a linear trend in time were considered. In the random component, year represents random between-year variation in contaminant levels, year * covariate1 represents random between-year variation in the relationship between contaminant levels and covariate1, and so on. A full model was first fitted with all the available covariates and the model was then simplified in a backwards stepwise procedure. (Note that only one size covariate was ever used – log length for fish and sqrt age for mammals.)

5 Workshop Trend Results

5.1 Results of Screening for Covariate Effects

On the basis of the statistical screening for covariate effects (see 4.3), the following observations and conclusions were made (for a complete tabular summary of the screening results see Appendix 6):

Shellfish monitoring (in Iceland and Norway) involved collection of animals in specific length/weight classes. The limited size ranges available make it difficult to establish relationships between the covariates and mercury concentrations. For this reason, no adjustment for biological covariates in these samples was performed. Information received from Iceland indicated that, due to possible depuration problems, trends of mercury in Icelandic mussel series are best considered using dry weight concentrations. This is not the case for Norwegian mussel samples where trends are determined on wet weight concentrations.

Fish (marine and freshwater) typically showed a strong relationship between mercury (in muscle tissue) and size of fish. Covariate availability in the datasets under consideration was: length > weight > sex > age. Length and weight were fairly consistently measured, age and sex less so; age determination of fish is complicated and thus size parameters were typically used as a surrogate for age. Under some programmes, samples were collected in specific length classes (e.g. freshwater fish from Swedish and some Canadian lakes, marine fish from Iceland and Norway). Stable isotopes were only available in freshwater fish from Canada, and then often not for all years; moisture content measurements were available for most samples, but mercury concentrations were normally reported on a wet weight basis, and trends were therefore investigated using wet weight concentrations.

Length was identified as a significant covariable for tissue mercury in the majority of freshwater fish (16 out of 18 available time-series), but only a few cases (2 out of 18) exhibited significant interactions with year of collection. Length was also a significant covariable for all seven of the marine fish time series, three of these also having significant interactions with year of collection, i.e. the Hg concentration - length regression slope was significantly different between at least two of the sample years.

Weight was identified as a significant covariate in the majority of time-series (12 of 17) where weight data was available for freshwater fish, and only one of these time-series showed significant interaction between weight and year of collection. Weight was also a significant covariable with mercury concentration in all four time-series for marine fish, with none of these showing an interaction between year of collection and weight.

Thus, using length and/or weight as a surrogate for age, the mercury – age relationships were generally stable between years for freshwater fish. The between year relationship was less stable for marine fish, particularly with length.

Sex was not correlated with mercury in the time series for freshwater or marine fish, however, the interaction with year of collection for marine fish was significant.

Ten of the time series for freshwater fish included stable isotope data. Seven time series were significantly correlated with $\delta^{15}N$ and one of those showed a significant interaction with year of collection. Two of the time series that were significantly correlated with $\delta^{15}N$ were also correlated with $\delta^{13}C$ and a separate time series showed a significant interaction with year of collection.

Terrestrial mammal time-series included data on the biological covariates age and sex; % moisture was also available for some series. Sex data were generally available, age less consistently so. The Swedish monitoring programme is based on animals of specific age and gender. Stable isotope data were not available for any of the terrestrial mammals. Of the five terrestrial mammal time series considered, three displayed significant mercury – age

correlations, and two of these time series showed a significant interaction with year of collection, along with one other. The interaction effect means that the mercury vs. age regression slope was significantly different between at least two years of sampling. Due to time limitations, sex was not examined as a covariable for terrestrial mammals; however this should be included in any future analyses of those terrestrial mammal datasets that are not based on gender specific sampling schemes.

Marine mammal time-series included the widest range of possible covariate measurements, including age and sex (measured in most time-series, but with gaps in the data), length and weight measured in some series, and stable isotopes included in some Canadian and Greenlandic monitoring programmes (but again with gaps in the available datasets). In 29 of the 31 marine mammal time series investigated, mercury concentrations were correlated with age, and 7 (23%) showed a significant interaction with year of collection. As age was not determined for most narwhal time series, length was tested for this species. For 5 of the 6 narwhal time series, length was a significant covariable, with only one showing a significant interaction with year of collection. Sex was a significant covariable in only one of the 6 marine mammal time series for which sex data were available; none of the interactions with year were significant.

Stable isotope data was available for ten marine mammal time series - four ringed seal and six beluga. Mercury concentrations for three of the four ringed seal time series were significantly, positively correlated with $\delta^{15}N$, and while none of those three showed a significant interaction with year of collection, the fourth time series – that which was not correlated with $\delta^{15}N$ – did show a significant interaction with year of collection. Mercury concentrations were not correlated with $\delta^{13}C$ in any of the ringed seal time series, however, two time series showed a significant interaction with year of collection.

Mercury concentrations for all of the beluga time series were significantly correlated with both $\delta^{15}N$ and $\delta^{13}C$, and of those, none showed a significant interaction with year of collection. It should be noted that the six time series for beluga are all from one site in the western Canadian Arctic and represent different tissues, including liver, muscle and muktuk, each of which have results for both total mercury and methylmercury, though not in all years.

Seabird time-series were based mainly on egg sampling programmes, with stable isotopes (and % moisture) available for the Canadian time series, but no covariates available for the Faeroe Islands series. None of the four time series for mercury in seabird eggs were correlated with $\delta^{15}N$ or $\delta^{13}C$.

The guillemot time series from Greenland, where mercury is analysed in liver tissue, included both age and sex data. Neither age, length nor sex were relevant covariables in time series for seabird eggs and were, therefore, not examined

5.2 Evaluation of Temporal Trends

Results of the Workshop analyses – unadjusted trends for datasets where covariates were not available, not significant, or where covariates were otherwise taken into account (e.g. by selective sampling schemes); and (biological) covariate-adjusted trends using the methods described in section 4 (above) – are tabulated in Annex 7. Annex 7 Table A presents results of the 27 time series that had significant temporal trends, the results for time-series where trends were not significant are tabulated in Table B.

The purpose of this report is to present the results of the Workshop and discuss methodological issues, rather than the meaning of trend results as such, which are the subject of interpretation in AMAP assessment reports. Considering results for individual time-series is also outside the scope of this report; these are considered in far greater detail in related scientific papers and publications. However, the following brief comments and observations are made on the combined results of the analyses conducted during the Workshop:

The results are in accordance with other studies presented in papers circulated as background documents to the workshop (Braune et al. 2005; Braune 2007; Dam and Rigét (2006, pers. comm.); Dietz et al. 2006a, 2006b; Evans et al. 2005a, 2005b; Rigét et al. 2006; Lockhart et al. 2005; Muir et al. 2005), and with the geographical patterns in mercury temporal trends identified in the 2002 AMAP Heavy Metals Assessment (AMAP, 2002; 2004).

Of the 12 significant time trends in Arctic Canada and West Greenland, all show increasing trends in mercury in biota, with average annual increases of between 1.9 to 10% (Annex 7, Table A and Figure 1). In some cases, such as the 10% increasing trend observed in Yukon caribou, the trends were strongly influenced by large increases during the early part of the time series, this being one possible reason for significant non-linear trend components that were identified in several time-series. East of Greenland, 12 out of 15 significant time trends showed decreasing trends in mercury of between -0.7 and -7% per year (Table A and Figure 1). Five of these significant decreasing trends, however, reflected recovery of lake systems following mercury contamination from sediments during construction of hydroelectric reservoirs at sites in Finland. A further six time series demonstrated considerable statistical power and were considered to have an adequate number of years of data, however these time series did not produce a discernable trend, i.e. mercury concentrations remained unchanged. All six of these time series are from the region east of Greenland.

The analysis of covariance showed that age, or some surrogate for age, such as length or weight, was a powerful covariate and indicated the need to normalize the mercury data prior to conducting trend analysis. Age/length/weight adjusted mercury concentrations were therefore used in the analysis of temporal trends for all time series, with the exception of those involving bivalves or seabird eggs. In most cases the trend analysis was done on both unadjusted and age/length/weight adjusted data, which allowed us to compare outcomes. In many cases, using unadjusted or adjusted mercury concentrations did not have a significant impact on the outcome of the trend analysis, e.g. insignificant trend results remained insignificant. Of the 27 significant trends, only four became significant after co-variation in age, length or weight had been accounted for. In these four cases, the data adjustment removed enough variation in the data to allow us to detect a significant trend. On the other hand, the temporal mercury trends in nine time series that showed significance with the use of unadjusted data, were no longer found to be significant after age adjustment. This result implies that the initial trend was related to differences in age/length/weight of the specimens over time, not changes in mercury concentrations. The time series where data adjustments made a difference in the outcome were evenly distributed between marine mammals and fish, which represent the bulk of time series examined at the workshop.

5.3 Evaluation of Adequacy of Time Series to Detect Trends

For the purposes of this Report, we use a concept called 'Adequacy' which is closely related to, yet distinct from, the 'statistical power' of datasets. 'Adequacy' (expressed as a percentage) is defined as the number of actual monitoring years in a time-series divided by the number of years of sampling required to detect a 5% annual change in Hg concentrations, with a significance level of $P<0.05$ and 80% statistical power. Adequacy should not be confused with the concept of power, which is only one component of adequacy albeit a very important one. Statistical power is defined as 1.0 minus the probability of a false positive result (β or Type II Error) (Sokal and Rohlf 1981); it is affected by factors such as the numbers of samples taken each year, variability of Hg concentrations within and between years of sampling, and the desired detectable rate of change of Hg levels. The desired power level can also be predetermined (which we have set here at a conventional value of 80%). When this is done, other parameters of practical interest can be calculated, such as the number of years of data required to statistically detect a 5% annual change in Hg concentrations (i.e. our measure of adequacy). Time-series which have fewer than the minimum number of years required to detect a 5% annual rate of change are

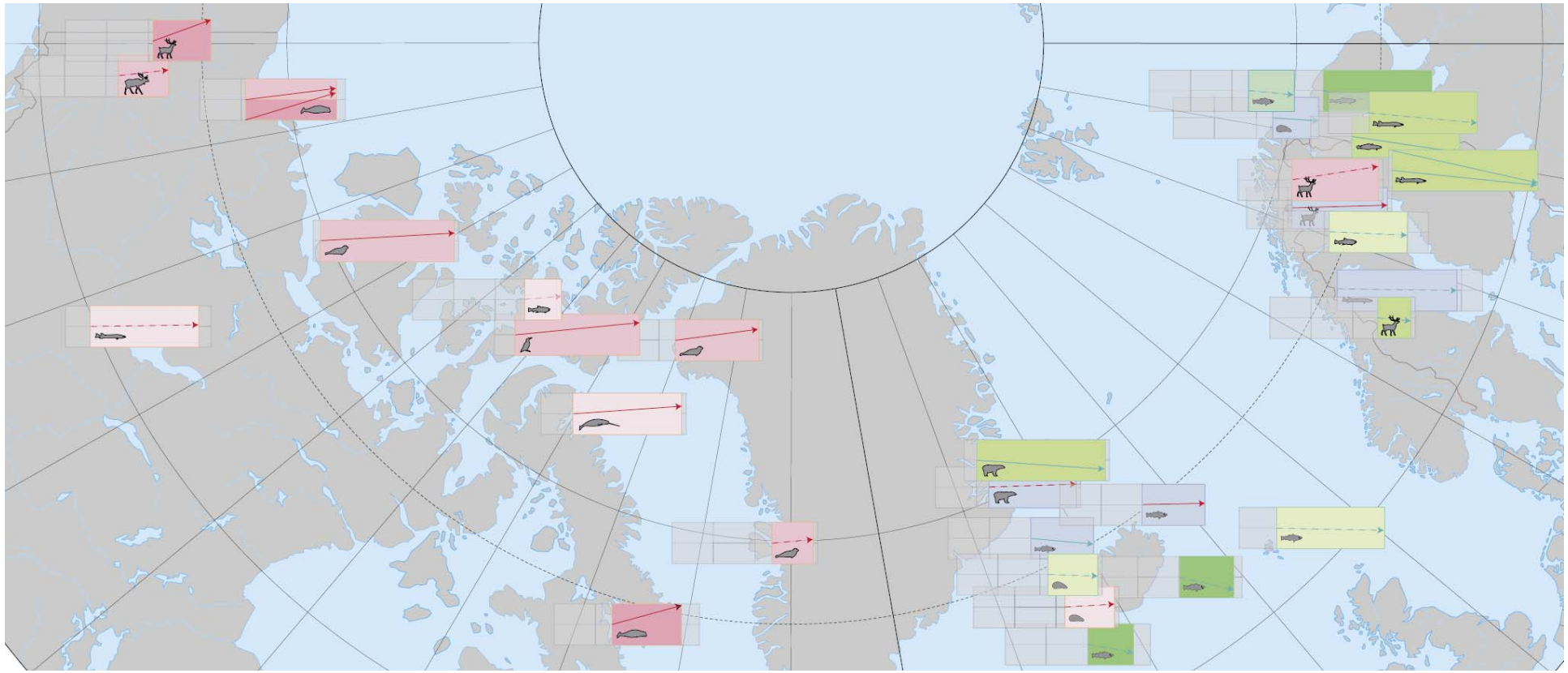

Figure 1. Map of significant trends (see Table A for details), also showing results for 6 non-significant trends for time-series where adequacy was > 1 (see section 5.7). Decreasing trends are indicated by green arrows and shading, increasing trends by red arrows and shading (the greater the trend the darker the shading; non-significant trends have blue shading); size of shaded areas represents the period covered by the time series within a frame that corresponds to the period 1970 to 2005/2006; dashed arrows indicate significant non-linear trend components; and symbols indicate species. Individual frames that show two trend lines represent time series where mercury concentrations were adjusted to two baseline ages/lengths, for example the lower 25th percentile and the upper 75th percentile for length in the case of Beaufort Sea beluga.

classed as ‘inadequate’, while those with years of data equal to or more than the minimum required are ‘adequate’.

Following the workshop in Copenhagen in 2001, an evaluation of the adequacy of the time-series available at that time was conducted. The conclusions of this work were that “*in general the investigated time-series were too short to possess an acceptable statistical power*” and that “*the consequence of sampling less frequently than once a year is considerable loss of power*” (Bignert et al., 2004). A further conclusion was that although at that time only four of the 42 time-series examined were considered adequate to detect trends at the desired level of power, this number should increase to 14 (i.e. from 10 to 32%) with a further five years of (annual) sampling. Consequently, as part of the 2006 workshop activities, a re-evaluation of the adequacy of available biota time-series was conducted to update the status of these datasets, and to see if there were further recommendations that would be relevant to future monitoring programme design and implementation.

Annex 7 contains two summary tables which includes adequacy values for all time-series available at the 2006 workshop. Figure 2 shows the same information in a plot of the adequacy of datasets submitted to the 2006 Workshop. Five of the 2006 datasets could be classified as “adequate”. This number would have been higher except that for 3 of the 4 adequate datasets in 2001, no new data were available for the 2006 Workshop (Figure 3a). Note that for two Swedish fish datasets (the two longest 2001 datasets), samples continue to be collected and archived for future analysis. In addition to the 3 unavailable ‘adequate’ datasets, another 13 datasets which were ‘inadequate’ in 2001 were also unavailable for the 2006 Workshop. The lack of new data may indicate that these time-series have been discontinued; no explanations were available. Compared to the predicted adequacy based on 2001 data, the actual adequacy of 2006 datasets overall was generally lower than expected (Fig. 3b).

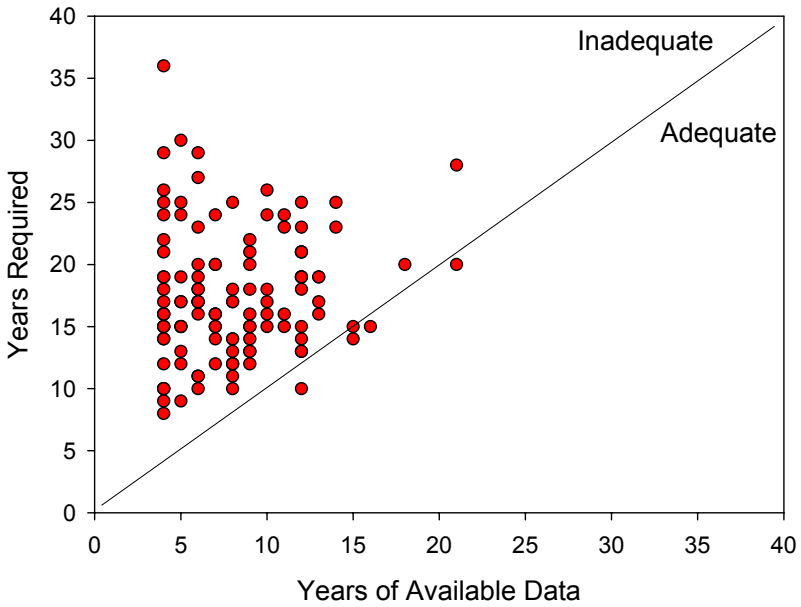

Figure 2. Statistical adequacy of the datasets analysed in the 2006 AMAP biotic Hg trends workshop

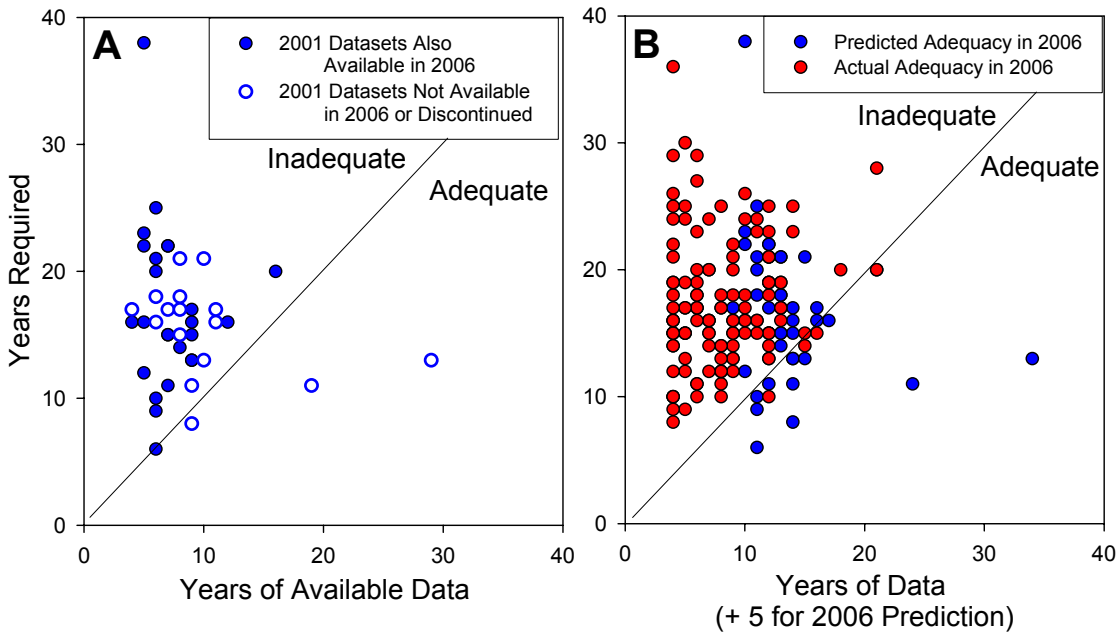

Figure 3. Comparison of temporal trend dataset adequacy from the 2001 workshop (A), the predicted adequacy for 2006 (i.e. plus five years of data), and the actual adequacy calculated in 2006 (B) for common datasets analysed both the 2001 and 2006 AMAP Hg trend workshops.

Figure 4 shows the improvement or decline in adequacy of time-series that were available to both the 2001 and 2006 workshops. If the adequacy of the data were identical in both years, the points would lie on the diagonal. Generally the mercury data markedly improved in adequacy since 2001, although there were 6 series for which adequacy either declined or remained constant, largely attributable to increased inter-year variability in the extended time-series. The improvements in adequacy were mainly due to more years of data being collected, so that inter-annual variability was better characterized than in 2001. The corollary was that declines in adequacy were due to greater inter-annual variability being apparent with more years of data.

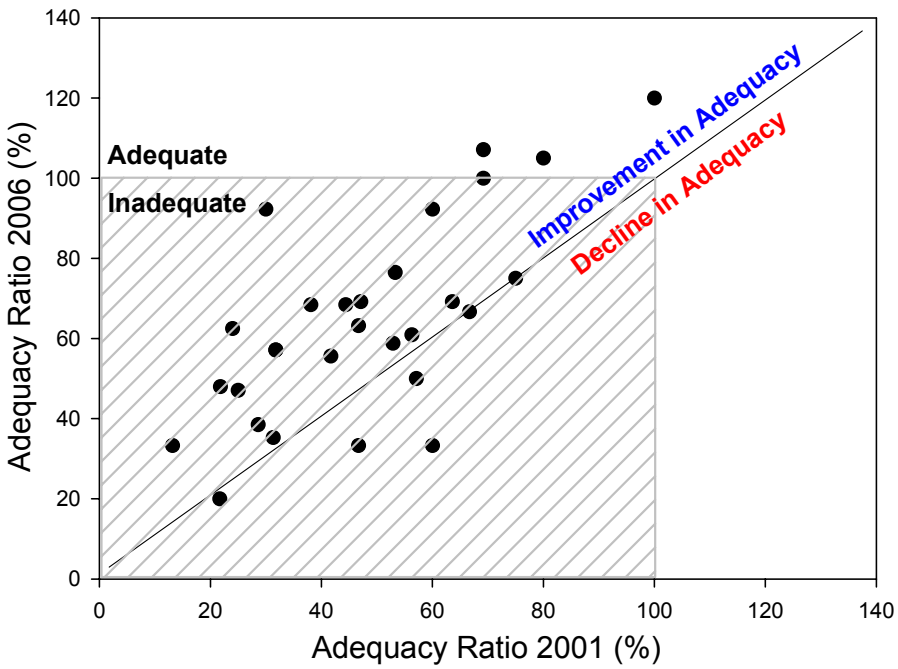

Figure 4. Change in statistical power (adequacy) of arctic biota Hg datasets between 2001 and 2006.

Future prediction of improvements in power based on the 2006 data show that with another 5 years of annual sampling, 27 of 91 time-series (30%) will attain adequate levels of statistical power to detect trends in Hg (Fig. 5). This assumes that all other factors such as intra- and inter-annual variability remain the same.

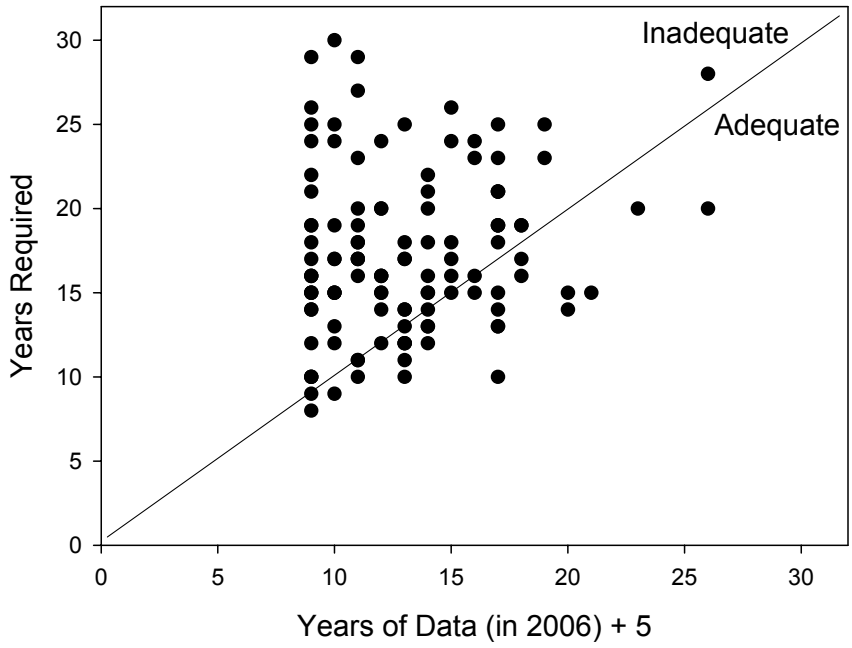

Figure 5. Predicted change in statistical power (adequacy) of arctic biota Hg datasets given an additional five years of data.

5.4 Results of Trend Analyses using Linear Mixed Models for Example Datasets

A linear mixed model (see 4.4 for general methodological information) was used to evaluate temporal trends in mercury concentrations while simultaneously accounting for multiple covariates, specifically, age/length, sex, and stable isotopes of nitrogen and carbon ($\delta^{15}N$ and $\delta^{13}C$). Fifteen Canadian timeseries where data for several of these covariates was available for multiple years were assessed using the linear mixed model. The output of this model included an indication of linear or non-linear trend significance ($p<0.05$) and identified the significant covariates.

In the initial analysis of covariates we assessed each potential covariate one at time, whereas the mixed model allows for the assessment of covariates all at once. In quite a number of cases the results differed between the two methods (Table 1). For example, $\delta^{13}C$ was often a significant covariate when assessed on its own (5 time series), however, when the linear mixed model was applied, $\delta^{13}C$ was only a weakly significant ($p=0.03$) covariate in one time series. This was not the case for $\delta^{15}N$, which was identified consistently as a significant covariate by both methods and in seven time-series assessed using the linear mixed model. Age/length were also frequently identified as significant covariates using the mixed model and in most cases this was consistent with the individual covariate analysis. Detailed output from the linear mixed model (not shown) indicated that there was little random between year variation in the relationship between mercury concentration and age/length or isotope covariates, which is inconsistent with other results for contaminant related datasets in fish (see e.g. Fryer & Nicholson, 2002, and the references cited therein).

In the case of six time series, the linear mixed model produced a different result with respect to the significance of the temporal trend than did the initial trend analysis (Annex 7, Table A and Table B). Among these six time series, there were four instances where the mixed model reported a significant trend and the initial trend analysis did not. It must be noted, however, that these comparisons are a bit spurious given that the two methods were often applied to different sets of data for the same time series. Since the mixed model was working with more variables, it was often only a subset of the original dataset that was complete for all covariants, for example stable isotope data may not have been available for all individuals and/or time points.

Despite only having applied the mixed model to a limited number of relatively complete data sets during the Stockholm workshop, the results presented here demonstrate the successful application of such models to evaluate contaminant time-series while accounting for multiple covariates. The use of a mixed model to evaluate temporal trends, while accounting for the confounding influence of multiple covariates, may be particularly appropriate for the analysis of general contaminant trends using biological indicators. Closer examination of the individual covariates may provide some valuable insight into ecosystem dynamics that could be influencing contaminant concentrations in the indicator species.

Table 1. Summary of trend analysis results generated using a linear mixed model and included the covariates age or length, sex, $\delta^{15}N$ and $\delta^{13}C$.

ID	**species**	**Significant covariates (assessed individually)**	**tissue**	**Trend analysis (From Annex 7) Tables A and B**		**mixed model (age / isotope adjusted)**		
				p	**method**	**p linear**	**p non-linear**	**significant covariates**
CAN-001	black-legged kittiwake	none	eggs	0.432	unadjusted linear	ns	ns	
CAN-002	northern fulmar	none	eggs	0.071	unadjusted linear	**0.015**	ns	
CAN-003	thick-billed murre	none	eggs	**0.005**	unadjusted **linear**	**0.004**	ns	
CAN-011	beluga	age, $\delta^{15}N$, $\delta^{13}C$	liver	**0.04**	age adjusted **linear** (age = Q25%)	ns	ns	age, $\delta^{15}N$
CAN-012	beluga	age, $\delta^{15}N$, $\delta^{13}C$	muscle	0.073[b]	age adjusted linear (age = Q50%)	**0.008**	**0.046**	age, sex, $\delta^{15}N$
CAN-018	Arctic char	length	muscle	**0.010**	length adjusted non-linear (length = Q50%)	ns	**0.014**	length, $\delta^{15}N$
CAN-019	ringed seal	age, $\delta^{15}N$	liver	0.315	age adjusted linear (age = Q50%)	ns	ns	$\delta^{15}N$
CAN-039	burbot	none	muscle	0.845	length adjusted linear (length = Q50%)	ns	ns	
CAN-040	burbot	length	liver	0.267	length adjusted linear (length = Q50%)	ns	ns	length
CAN-041	lake trout	length	muscle	0.434	length adjusted linear (length = Q50%)	ns	ns	length, $\delta^{15}N$, $\delta^{13}C$

ID	species	Significant covariates (assessed individually)	tissue	Trend analysis (From Annex 7) Tables A and B		mixed model (age / isotope adjusted)		
				p	method	p linear	p non-linear	significant covariates
CAN-042	lake trout	length*, $\delta^{15}N$ *	muscle	0.637	length adjusted linear (length = Q25%)	ns	ns	length
CAN-043	burbot	length, $\delta^{15}N$, $\delta^{13}C$	muscle	0.091[b]	length adjusted linear (length = Q50%)	**0.028**	ns	$\delta^{15}N$
CAN-044	burbot	length, $\delta^{15}N$	muscle	0.376	length adjusted linear (length = Q50%)	**0.012**	ns	$\delta^{15}N$
CAN-060	northern pike	length, $\delta^{15}N$, $\delta^{13}C$	muscle	0.457	length adjusted linear (length = Q50%)	ns	ns	length
CAN-061	northern pike	length, $\delta^{15}N$, $\delta^{13}C$ **	muscle	**0.022**	length adjusted non-linear (length = Q50%)	ns	ns	length

b. Unadjusted total mercury concentrations demonstrated a statistically significant temporal trend, however, when total mercury concentrations were adjusted for age, length or weight, the trend was no longer found to be significant.

* analysis of covariates revealed a significant interaction with both Hg concentration and year of collection

** analysis of covariates revealed a significant interaction with year of collection but not Hg concentration

5.5 Use of Stable Isotopes as Covariates

Data on stable isotopes were available for only a few time series, and in several cases not available for the entire series (all years). These measurements can provide information on changes in feeding habits and contaminant exposure that may explain mercury concentrations in (individual) animals. In addition, stable isotopes and mercury are not always analysed in the same sample matrix, and thus the relationship between the two may reflect a short-term response as opposed to a change in life-time exposure.

Consequently, stable isotopes were not used as a standard part of the procedure to normalise concentration values prior to application of the trend detection methods in the same way as was done for biological covariables. Rather, it was considered that stable isotope information should be used after (unadjusted or biological-parameter-adjusted) trend analysis as a possible aid to interpreting or explaining observed trends. In this way, important information on changes in ecosystem structure caused by, e.g. climate change, is also available for investigation and not simply ‘eliminated’ by the normalisation procedure.

From an examination of the available datasets that included stable isotopes, in the case of marine birds (see example in Figure 6) and beluga from Arctic Canada, no clear relationship was observed between changes in stable isotope levels and increases in mercury concentrations. However, in ringed seals from Central West Greenland during the period 1999 to 2004 Hg concentrations in liver increased significantly. However, after adjusting for stable nitrogen isotope determined in muscle, the increase become less and non-significant (Figure 7; Riget et al. 2006). In this case, the data on stable isotopes helped determine that the rise in Hg was likely a reflection of diet rather than atmospheric deposition Hg.

As datasets for stable isotopes become more complete, and the relationships between stable isotopes and contaminant concentrations are better defined, it may be more appropriate to apply stable isotope normalization.

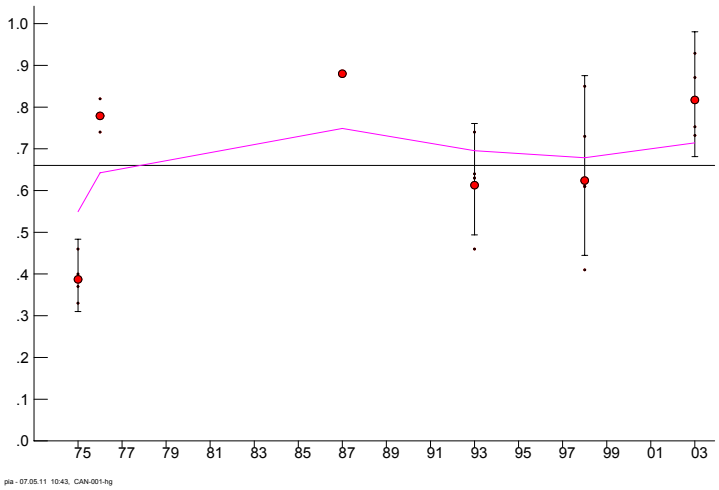

Figure 6a. Temporal trend of mercury (µg/g dw) in black-legged kittiwake (Rissa tridactyla) eggs from Prince Leopold Island, Canada.

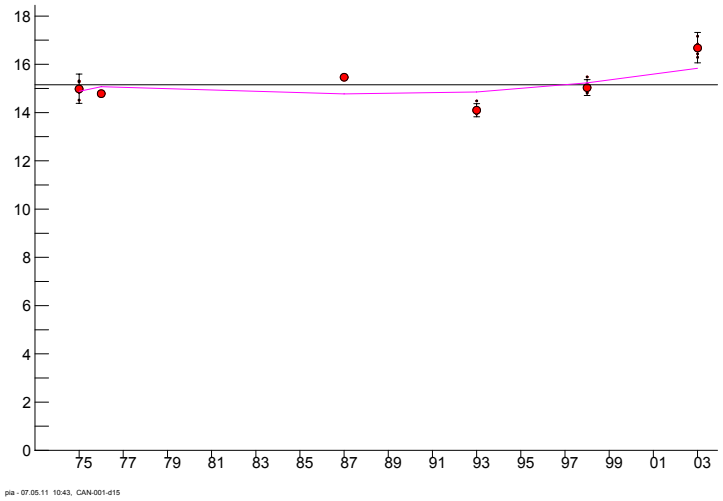

Figure 6b. Temporal trend of δ15N concentrations in black-legged kittiwake eggs from Prince Leopold Island, Canada.

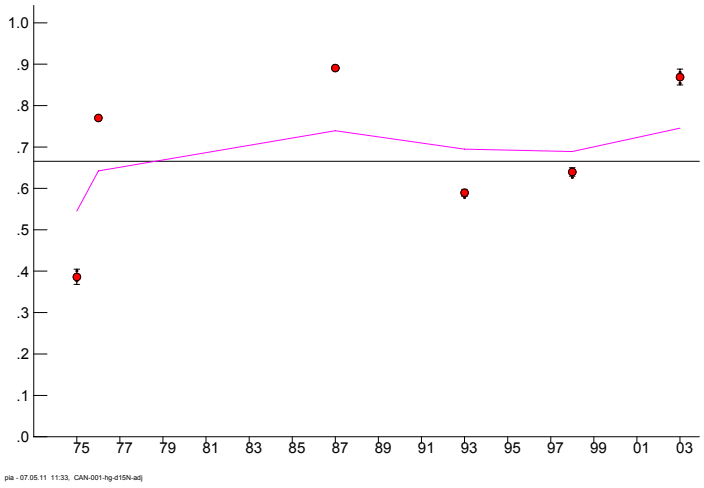

Figure 6c. Temporal trend of $\delta^{15}N$ adjusted mercury concentrations in black-legged kittiwake eggs from Prince Leopold Island, Canada. Trend results are unchanged; however adjustment has slightly increased the index concentration values in 1999 and 2003, relative to Figure 6a (reflecting the upward tendency in $\delta^{15}N$ values towards the end of the time series).

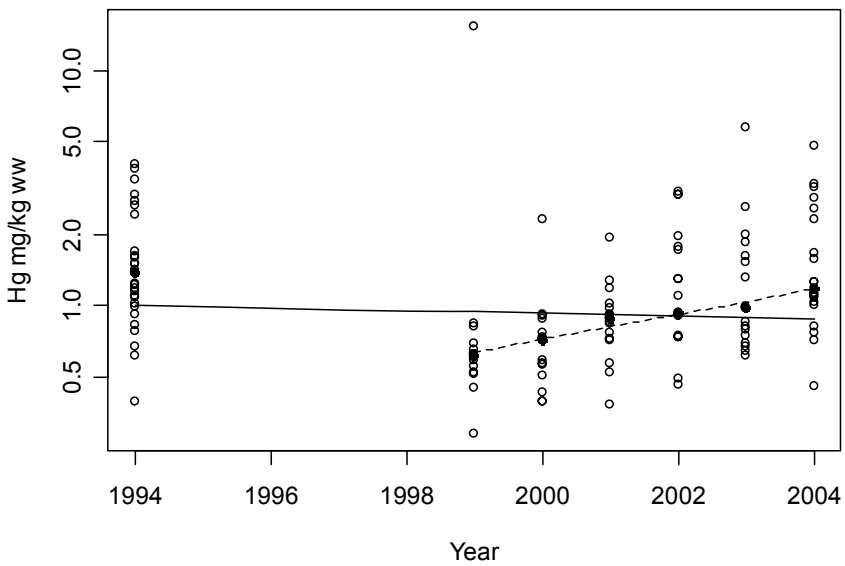

Figure 7a. Temporal trend of unadjusted mercury concentrations in ringed seals from Central West Greenland.

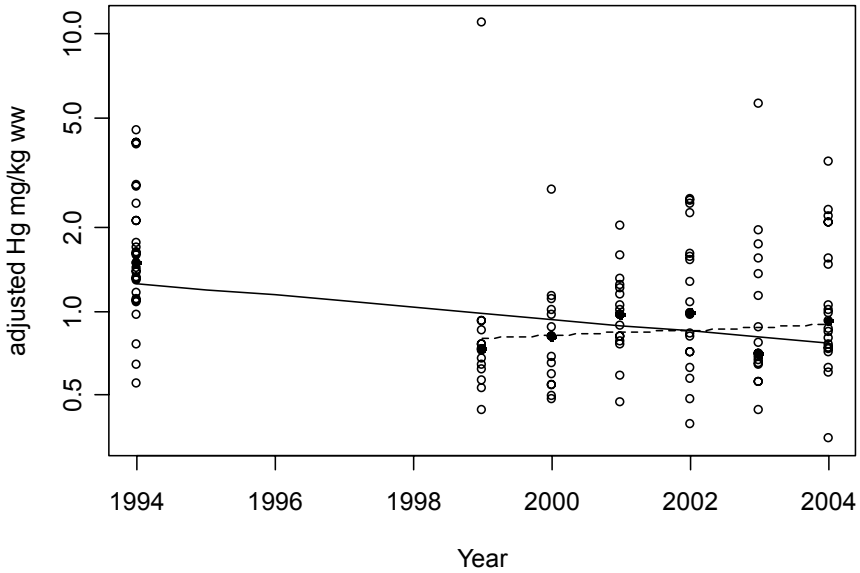

Figure 7b. Temporal trend of $\delta^{15}N$ adjusted mercury concentrations in ringed seals from Central West Greenland. (taken from Riget et al. 2006. The statistical method used was similar but not identical to the method used in the data analysis performed during the Stockholm workshop).

5.6 Influence of Sampling Matrix on Time Series Results

A number of monitoring programs collected several different tissues as part of their respective sampling regimes. In some cases mercury was analyzed in two or more tissues which has allowed us to look at the influence of sampling matrix on our ability to measure temporal trends. For some fish monitoring programs time series mercury data were submitted for muscle and liver. In several marine mammal time series mercury data was submitted for liver and kidney, and/or muktuk (skin and fat), and/or hair. Data was also submitted for liver and kidneys of moose and caribou. Unfortunately it is difficult to conduct a meaningful comparison between the different sampling matrices due to differences in completeness between time series of different tissues. Based solely on the statistical

parameters reported here (Annex 7, Table A and B) there does not appear to be any clear advantage to using one tissue over another, notwithstanding any argument that might be made on a biological basis. Temporal patterns in mercury concentrations displayed by different tissues are generally similar (Figure 8), which supports the notion that apportionment between the various organs of an organism is relatively consistent for a given species.

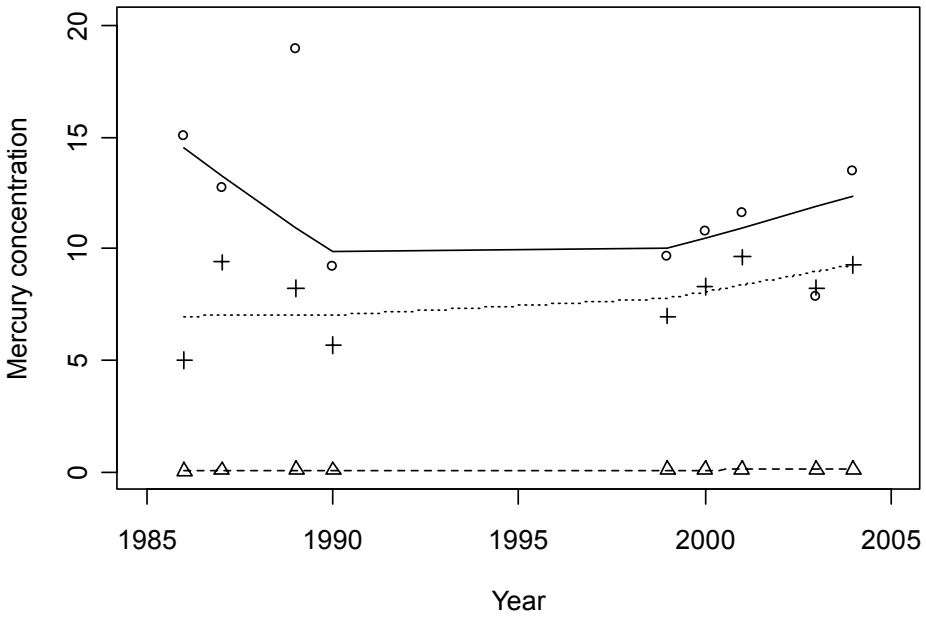

Figure 8. Geometric annual means of Hg in kidney (o), liver (+) and muscle (Δ) tissue of polar bears from East Greenland. Only bears with data on all three tissues are included. The lines shown are from the loess smoother. Some correlated time trends (1999-2001) between tissues are apparent, but this is not obvious.

6 Conclusions and Recommendations

The results of the trend analyses conducted during the workshop support and confirm the picture that was presented in the AMAP 2002 heavy metals assessment, that recent temporal trends in mercury west of Greenland differ from those found east of Greenland (see Figure 1). In the Arctic regions between the Yukon and West Greenland, mercury levels in biota appear to have increased during recent decades, in particular in marine and freshwater biota in the Canadian High Arctic. Some of these trends are dominated by large increases during the early part of the time-series, since when mercury levels have been relatively constant. To the east of Greenland, in the marine environment around Iceland and in the (subarctic) terrestrial and freshwater environments of Scandinavia, a general decrease in mercury levels in biota is observed.

It is clear from the results of co-variable analysis that some co-variables such as age (or an age surrogate such as length or weight), and diet (represented by $\delta^{15}N$) significantly affect measured Hg concentrations in the majority of Arctic biota for which data were available. Furthermore, the effect of these co-variables on Hg in a given species was predictable, i.e. it was rarely subject to significant year-to-year changes. Conversely, other co-variables such as sex and $\delta^{13}C$ are not influential in most cases. On this basis, we recommend that:

Hg monitoring programmes should concurrently make measurements of all of the important co-variables on each individual animal for which Hg data are generated.

The question of which co-variables need to be monitored must be decided for each species on the basis of empirical data (i.e. where there is a significant effect), and should be applied across all monitoring sites where that species is found, even if there is some evidence of site-to-site difference in the effect. Sampling schemes which target specific age/length/weight classes of animals (as opposed to random sampling followed by measurement of these parameters) is also a suitable approach to reducing the effect of these co-variables. Age, length and weight are traditionally viewed as important co-variables for Hg in biota. However, $\delta^{15}N$ (dietary variations) was almost as important as the former three parameters (significant in 72% of cases where data were available). Given that most monitoring programmes do not routinely collect $\delta^{15}N$ data, and given that unrecognised trends in diet could alter the conclusions of a Hg trend analysis, it is recommended that:

Special efforts should be made to concurrently collect $\delta^{15}N$ data on all monitored species, unless there is empirical evidence that it is not an important factor in a particular species.

Although there have been marked improvements since 2001 in the adequacy of most datasets to detect Hg changes in Arctic biota because of the additional years of data provided, most datasets are still not adequate to detect the required changes in Hg concentrations (average of 5% annually) with the acceptable standards of statistical power. This is predominantly due to the length of the datasets being insufficient, given the amount of intra- and inter-year variability that occurs in Hg concentrations. Some of this variability appears to be inherent, i.e. some remains even when known significant co-variables are taken into account. However, in other cases, co-variable data were not available, and so the overall variability in Hg can not be statistically reduced or accounted for. Nonetheless, additional years of data have produced adequacy improvements in most cases. Projections are that another 5 years of monitoring will place 29% of the biotic datasets in the "adequate" category. Therefore:

Existing monitoring programmes which have displayed an improvement in adequacy since 2001 should be continued at the same level of sampling intensity for a further 5 years; and

Datasets which have already achieved the required level of adequacy or which show a significantly increasing or decreasing trend should be continued, because these constitute the best evidence available about changing Hg status in various regions of the Arctic environment and in human food chains.

If cost-savings for reinvestment in other parts of the monitoring programme are required, it is recommended:

Those few datasets which have shown declines in adequacy since 2001, and also show no significant trends in Hg, should be evaluated as to whether they should continue.

The influence of climate change on biotic Hg levels could not be statistically evaluated, however, given the theoretical potential for effects to be mediated through changing prey species availability, Hg speciation and target animal distribution, **further attempts should be made to explore and test associations between climatic indicators such as the North Atlantic Oscillation Index (NAO) and Arctic Oscillation Index (AOI) and Hg trends.**

6.1 Recommendations for future AMAP contaminant trend assessments

On the basis of experience at this Workshop, the following recommendations are provided as practical suggestions for future statistical trend assessment meetings concerning other contaminants:

- Datasets should be confirmed well before the assessment meeting to allow data normalization and preliminary screening of significant co-variables to take place.
- A standard template (table) for presentation of trend results should be agreed on and filled in as far as possible on the basis of preliminary runs.
- Batch processing options should be exploited to automate standard runs (and will be pursued with PIAW)
- Preparation of regular (possibly every 2nd year) updates – data reports for as many trend datasets as required for public information from AMAP and for use by external parties such as UNEP, UN-ECE.
- Before conducting time trend analysis, the dataset should be tested and subsequently normalised for covariance of key biological variables such as the age or length. If neither of these are available the weight should be used.
- Distribute final PIAW to interested groups with instructions/manual for further testing by non-statisticians
- Develop new statistical utilities based on CTSM code

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*meeting co-chairs (co-chairs of the AMAP Mercury Expert Group)

Annex 2. Workshop Agenda

AMAP Workshop on Temporal Trends of Mercury in Biota
Stockholm 30 October - 3 November

1) Presentation of the goals of the workshop
2) Introduction of participants
3) Overview of the available short-term datasets, and the workshop data policy (Simon Wilson)
4) Presentation of the statistical programmes used for preliminary analyses/available at the meeting (Anders Bignert, with possible additional input from Rob Fryer)
5) Introduction to possible normalization procedures, with examples for datasets received (Frank Riget)
6) Presentation of the preliminary results including discussion on lacking data, data problems, normalising possibilities (Simon Wilson and Anders Bignert), with comments from data providers.

   Canada

   Greenland/Faroe Islands

   Finland

   Iceland

   Norway

   Sweden

   Other
7) Introduction and presentation of results of some available long term datasets (Peter Outridge and Rune Dietz)
8) Re-analysis of selected datasets (sub-groups of statisticians and data providers)
9) Presentation of the revised results (Simon Wilson, Rob Fryer and Anders Bignert)

   Canada

   Greenland/Faroe Islands

   Finland

   Iceland

   Norway

   Sweden

   Other
10) Preparation of data summaries for the workshop report and possible products for possible external use
11) Presentation of Conclusions of trend analyses (Simon Wilson, Peter Outridge, Jason Stow, Rune Dietz)

12) Discussion of content of Workshop report and drafting arrangements

13) Development of Recommendations Including possible adjustments to the AMAP monitoring programme and procedures for future handling of mercury trend datasets (Jason Stow, Simon Wilson, Peter Outridge, Rune Dietz)

14) Future plans

    Continuation of short term sampling programmes (country by country plans) IPY Hg Long Term Time Series (Peter Outridge, Birgit Braune and Rune Dietz). Experiences gained and relevance to AMAP POPs trend assessment work Plans for next comprehensive mercury assessment, and possible related planning meeting in 2007.

15) Report preparation

    Production of a relatively short document that will include an overall summary from the workshop of the trend interpretations and findings, and include tables summarizing the statistical outputs. This document will be distributed to AMAP/Arctic Council and to various external agencies, after review by all data providers. To be drafted during the Workshop and finalized by Simon, Jason, Peter and Rune and with input from the participants and others.

Annex 3. Workshop Data Policy Statement

The main goal of the workshop is to consider trend results collectively rather than individually. A statistical methodology will be applied that is designed to be robust with respect to data with varying characteristics and, while not necessarily constituting the optimal approach for any particular dataset, it is appropriate to the goals of the workshop.

Data policy will be addressed through the following general principles:

1. All workshop participants will be asked to sign the AMAP data confidentiality agreement. This agreement has been used in previous AMAP activities to safeguard scientists rights when they contribute their unpublished data for use by AMAP in its assessments and related work.

2. No individual dataset or products derived from an individual dataset will be presented as part of a report for public dissemination without the express agreement of the owner of the data concerned.

3. The Workshop Report will serve as a record of the workshop discussions and conclusions, and is essentially intended for AMAP internal use. While such reports are generally made available to interested parties, they are not widely disseminated (i.e. do not constitute an AMAP assessment report); they should, however, be viewed as 'publicly available'.

The workshop report would have the same status as, e.g. an NCP annual report. This type of 'grey literature' does not constitute a formal publication and therefore should not prevent scientists from publishing their own papers in the scientific literature at some later point.

The workshop report will not include plots or tables of specific trend data except for illustrative purposes, and then only with the data owners permission.

4. Electronic copies of raw data will only be available to those workshop participants responsible for collecting and compiling national data (AMAP Secretariat and national focal points) and statisticians (4-5 workshop participants) who will conduct some preliminary statistical runs. Other workshop participants will only see paper or screen copies for discussion purposes;

5. All data owners will be given the opportunity to comment on the Workshop Report before it is finalized.

Product	Audience	Policy
Raw datasets	Sub-group of workshop participants (statisticians and dataset compilers responsible for preliminary analyses)	All workshop participants will be asked to sign the AMAP data confidentiality agreement.
Trend analysis summary sheets *(statistical analyses results for individual datasets)*	Workshop Participants	All workshop participants will be asked to sign the AMAP data confidentiality agreement.
	AMAP products in the public domain	Data sheets will only be made available with express permission of the owners of the data concerned,
	AMAP products for external organizations (e.g. UNEP / UN ECE - for possible use in evaluations of the effectiveness and sufficiency of international agreements, etc.)	Data sheets will only be made available with express permission of the owners of the data concerned,
Summarised data products *(overview table(s) summarizing trend results for (all) received datasets)*	Workshop Participants	All workshop participants will be asked to sign the AMAP data policy agreement (see above)
	AMAP products in the public domain	If any data owners object to their results being included in this type of summary, they can ask that entries relating to their datasets be omitted from any tables that may be included in public domain products.
Workshop report	AMAP products in the public domain	The workshop report will not include plots or tables of specific trend data except for illustrative purposes and only with the data owners permission. All data owners will be given the opportunity to comment on the Workshop Report before it is finalized.
AMAP Assessment Reports *AMAP update reports* NOTE: It is <u>not</u> envisaged that any external product resembling an 'AMAP assessment report' will be produced on the basis of this workshop alone. The next full AMAP mercury assessment is planned for 2010.	AMAP products in the public domain	Data owners are invited to contribute their information, and any contributions would be fully acknowledged and appropriately referenced, etc. Significant contributors would be invited to be co-authors of an eventual review article.
Scientific paper / review article	Scientific community, general public	Standard practices (i.e. direct requests to data owners) would be followed for obtaining permission to use/refer to other peoples data. Significant contributors would be invited to be co-authors of an eventual review article. All contributions would be fully acknowledged and referenced.

Annex 4. PIA

The basic statistical methodology applied when running PIA is a robust regression-based analysis to detect trends in time-series datasets (Nicholson et al., 1998). The procedures for time-trend detection, and power calculations follow those described in Bignert (2001).

The standard run in PIA calculates annual geometric means from the input data and uses these as the annual 'index' values that are tested for linear trends; by default a 3-year running-mean smoother is also fitted and compared with the (log) linear regression in the tests for non-linear trend components.

Input data comprise un-normalized contaminant concentration values, with multiple values allowed in each year.

Options can be set to modify the standard analysis so that, for example, medians are calculated instead of geometric means as the annual index values. The 95% confidence interval for the median is estimated by a method described in Dixon and Massey (1969). Similarly, PIA can be directed to apply a 'loess' smoother (Cleveland, 1979) (with a window defined by the user), instead of the 3-year running-mean smoother.

Finally, PIA can be directed to use the 'third' value in the input data records as an 'adjustment' variable. Data values are then adjusted to a specified value of the adjustment variable, either using the best-fit (linear) regression relationship between the data values relationship, or a forced-fit, where the regression 'slope' for the adjustment variable, is specified.

Important features in the statistical output are based on the concept of 'statistical power', i.e., the chance to detect a true trend. The 'statistical power' depends on the inherent random (unexplained) variation in a dataset. The potential of any dataset to show a statistically significant change over time depends on its statistical power; a low variation implies higher statistical power, and that smaller changes can be detected, or that the time period required to detect a given change will be shorter.

Users should refer to the methodological descriptions that are available (including descriptions aimed at non-statistical audiences) for further information about the statistical method applied by PIA.

A 'PIA distribution package' (a zip file containing the program, documentation and examples) has been posted on the public area of the AMAP website. It can be found in a folder under online publications' on the AMAP website (www.amap.no) or directly via the following link:

http://amap.no/documents/index.cfm?dirsub=%2FPIA%20Trend%20Analysis%20Package

Bignert, A., 2001. Comments concerning the National Swedish Contaminant Monitoring Programme in Marine Biota. Annual report to the Swedish EPA.

Cleveland, W. S. 1979. Robust locally-weighted regression and smoothing scatterplots. Journal of the American Statistical Association. 74. 829-836.

Dixon, J.W.,F.J. Massey. 1969. Introduction to Statistical Analysis. 3rd ed. McGraw-Hill, New York. 638p.

Nicholson M.D., R. Fryer and J.R.Larsen. 1998. A Robust Method for Analysing Contaminant Trend Monitoring Data. Techniques in Marine Environmental Sciences. ICES.

Annex 5. CTSM

The basic statistical methodology applied when running CTSM is a robust regression-based analysis to detect trends in time-series datasets (Nicholson et al., 1995, 1998). This approach is adapted according to the statistical methodology described in Fryer and Nicholson, 1999.

CTSM comprises a series of statistical routines, developed in the 'R' or 'S' statistical languages, that fit an annual contaminant time series model to the specified input dataset. The basic input dataset consists of a pair of vectors (of equal length), one giving the year of each observation (without duplicates), and the second the corresponding contaminant index value in each year. Index values used are normally the log median contaminant concentration values.

Options allow CTSM to perform a weighted fit using weighting values supplied in a third vector. This is typically used to weight values based on (analytical) quality assurance considerations. By default an un-weighted fit is produced.

CTSM fits a loess smoother to the data, with (by default) a 7 year window. The window can be specified. CTSM can also be instructed to handle missing values in different ways.

Fryer R.J., and M.D.Nicholson, 1999. Using smoothers for comprehensive assessments of contaminant time series in marine biota. ICES Journal of Marine Science, 56: 779-790.

ANNEX 6. Summary of results of analysis for covariate effects

The ANCOVA/ANOVA analysis for covariate effects were conducted primarily for the covariates age, length or weight. Age is the main biological covariate of interest, with length and weight typically being used as a surrogate for age, and generally being easier to determine. Sex was also examined as a biological covariate. In addition, for some time series, data were available for the stable isotopes **δ^{15}N** and **δ^{13}C**, which respectively provide information on trophic level and feeding habit of the animals concerned (i.e., feeding environment: freshwater vs marine vs terrestrial, and composition: benthic vs pelagic in the marine environment).

Table 1. *Overview of the log-Hg covariance results related to animal* ***age*** *conducted for the available Arctic time trend series with more than 4 years of data Cells values are counts in the respective category.*

Species Group	age			age*			Total
	N.S.	Signif.	(blank)	N.S.	Signif.	(blank)	
Freshwater fish	-	-	33	-	-	33	33
Terrestrial mammals	2	3	12	2	3	12	17
Marine fish	-	-	17	-	-	17	17
Marine mammals	2	29	14	24	7	14	45
Seabirds	-	-	6	-	-	6	6
Total	4	32	82	26	10	82	118

* interaction with year of collection

blank indicates that calculations were not performed for these time series, typically due to incompleteness or lack of availability of covariate data.

Table 2. *Overview of the log-Hg covariance results related to animal* ***length*** *conducted for the available Arctic time trend series with more than 4 years of data. Cells values are counts in the respective category.*

Species Group	length			length*			Total
	N.S.	Signif.	(blank)	N.S.	Signif.	(blank)	
Freshwater fish	2	16	15	16	2	15	33
Terrestrial mammals	-	-	17	-	-	17	17
Marine fish	-	7	10	4	3	10	17
Marine mammals	1	5	39	5	1	39	45
Seabirds	-	-	6	-	-	6	6
Total	3	28	87	25	6	87	118

* interaction with year of collection

blank indicates that calculations were not performed for these time series, typically due to incompleteness or lack of availability of covariate data.

Table 3. *Overview of the log Hg covariance results related to animal* ***weight*** *conducted for the available Arctic time trend series with more than 4 years of data. Cells values are counts in the respective category.*

Species Group	weight			weight*			Total
	N.S.	Signif.	(blank)	N.S.	Signif.	(blank)	
Freshwater fish	5	12	16	16	1	16	33
Terrestrial mammals	-	-	17	-	-	17	17
Marine fish	-	4	13	4	-	13	17
Marine mammals	-	-	45	-	-	45	45
Seabirds	-	-	6	-	-	6	6
Total	5	16	97	20	1	97	118

* interaction with year of collection

blank indicates that calculations were not performed for these time series, typically due to incompleteness or lack of availability of covariate data.

Table 4. *Overview of the log Hg covariance results related to animal* ***sex*** *conducted for the available Arctic time trend series with more than 4 years of data. Cells values are counts in the respective category.*

Species Group	sex			sex*			Total
	N.S.	Signif.	(blank)	N.S.	Signif.	(blank)	
Freshwater fish	1	-	32	1	-	32	33
Terrestrial mammals	-	-	17	-	-	17	17
Marine fish	1	-	16	-	1	16	17
Marine mammals	5	1	39	6	-	39	45
Seabirds	-	-	6	-	-	6	6
Total	7	1	110	7	1	110	118

* interaction with year of collection

blank indicates that calculations were not performed for these time series, typically due to incompleteness or lack of availability of covariate data.

***Table 5**. Overview of the log Hg covariance results related to* **δ^{15}N** *conducted for the available Arctic time trend series with more than 4 years of data. Cells values are counts in the respective category.*

Species Group	deltaN			deltaN*			Total
	N.S.	Signif.	(blank)	N.S.	Signif.	(blank)	
Freshwater fish	3	7	23	9	1	23	33
Terrestrial mammals	-	-	17	-	-	17	17
Marine fish	-	-	17	-	-	17	17
Marine mammals	1	13	31	10	4	31	45
Seabirds	4	-	2	4	-	2	6
Total	8	20	90	23	5	90	118

* interaction with year of collection

blank indicates that calculations were not performed for these time series, typically due to incompleteness or lack of availability of covariate data.

***Table 6.** Overview of the log Hg covariance results related to* **δ^{13}C** *conducted for the available Arctic time trend series with more than 4 years of data. Cells values are counts in the respective category.*

Species Group	deltaC			deltaC*			Total
	N.S.	Signif.	(blank)	N.S.	Signif.	(blank)	
Freshwater fish	8	2	23	9	1	23	33
Terrestrial mammals	-	-	17	-	-	17	17
Marine fish	-	-	17	-	-	17	17
Marine mammals	7	7	31	9	5	31	45
Seabirds	4	-	2	4	-	2	6
Total	19	9	90	22	6	90	118

* interaction with year of collection

blank indicates that calculations were not performed for these time series, typically due to incompleteness or lack of availability of covariate data.

Annex 7. Results of trend analysis

Table A. Significant trend results. Summary of time series that had significant temporal trends with either unadjusted or age/length/weight adjusted concentrations of total mercury.

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. ****
ICE-004A[a]	Blue mussel	soft tissue	64.350	-21.483	Hvítanes, Hvalfjordur	13	1992	2004	13	**0.044**	unadjusted non-linear	1.5 (-14.4, 7.5)	0.27	0.68
ICE-006A[a]	Blue mussel	soft tissue	64.03333	-22.033	Straumur, Straumsvík	12	1992	2004	12	**0.037**	unadjusted non-linear	-1.2 (-8.9, 6.4)	0.33	0.57
ICE-015	Cod	muscle	64.500	-14.000	SE Iceland	28	1990	2003	7	**0.045**	unadjusted non-linear	-7 (-12, -2.5)	0.13	0.58
ICE-016	Cod	muscle	64.500	-23.000	SW Iceland	19	1990	2001	6	**0.004**	unadjusted non-linear	-5.3 (-16, 5.2)	0.15	0.33
NOR-003	Cod	muscle	69.933	29.667	Varangerfjorden (10B)	294	1994	2005	12	**0.022**	length adjusted non-linear (length = Q50%)	-3.2	0.24	0.63
CAN-003	Thick-billed murre	eggs	74.000	-90.000	Prince Leopold Island	35	1975	2005	9	**0.005**	unadjusted **linear**	2.5 (1.1, 3.9)	0.25	0.69
CAN-011.0	Beluga	liver	69.500	-133.586	Hendrickson Island	165	1981	2003	9	**0.04**	age adjusted **linear** (age = Q25%)	3.4	0.28	0.67
CAN-011.1	Beluga	liver	69.500	-133.586	Hendrickson Island	165	1981	2003	9	**0.001**	age adjusted **linear** (age = Q75%)	8.3	0.29	0.67
CAN-018	Arctic char	muscle	74.683	-94.950	Resolute Lake	95	1997	2006	8	**0.010**	length adjusted non-linear (length = Q50%)	2	0.16	0.6
CAN-021.1	Ringed seal	liver	70.65155	-117.726	Holman Island	142	1972	2004	7	**0.043**	age adjusted **linear** (age = Q75%)	2.6	0.25	0.46

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. ****
CAN-027.0	Beluga	liver	66.124	-65.626	Pangnirtung	132	1984	2002	10	**0.004**	age adjusted **linear** (age = Q25%)	7.4	0.24	0.63
CAN-034	Narwhal	kidney	72.800	-77.000	Pond Inlet	159	1978	2004	8	**0.023**	age adjusted **linear** (age = Q50)	1.9	0.15	0.7
CAN-049	Moose	kidney	63.000	-135.000	Yukon	269	1993	2005	12	**0.034**	unadjusted non-linear	2.9 (-7.8, 14)	0.42	0.48
CAN-056	Caribou	kidney	63.000	-135.000	Porcupine herd	393	1991	2005	14	**0.007**	unadjusted **linear**	10 (3.3, 17)	0.43	0.61
CAN-061A	Northern pike	muscle	61.171	-113.672	Great Slave Lake - West Basin	169	1976	2002	10	**0.022**	length adjusted non-linear (length = Q50%)	0.3	0.15	0.4
GRE-001.1	Ringed seal	liver	77.48	-69.36	Avanersuaq	102	1984	2004	5	**0.011**	age adjusted **linear** (age = Q75% = 4 yr old)	3.7		0.21
GRE-002	Ringed seal	liver	69.120	-49.780	Qeqertarsuaq	150	1994	2004	7	**0.005**	age adjusted non-linear (age = Q50% = 1 yr old)	2.3		0.44
GRE-006[a]	Polar bear	hair	70.483	-21.95	Ittoqqoortoormiit	146	1973	2004	10	**0.033**	unadjusted **linear**	-2.1		**0.92**
FAR-002.0[c]	Atlantic cod	muscle	62.000	-7.500	Faroe Islands	792	1979	2005	18	**0.018**	length adjusted non-linear (length = Q25% = 58.5 cm)	-0.7		**0.9**
SWE-002	Arctic char	muscle	68.3	18.65	Lake Abisko	190	1981	2000	19	**0.012**	unadjusted non-linear	-1.2 (-2.6, 0.23)	0.11	**1.73**
SWE-003	Reindeer	muscle	68.3	18.65	Abisko	185	1983	2004	21	**0.001**	unadjusted non-linear	4.3 (-0.64, 9.2)	0.4	0.75
SWE-005	Reindeer	muscle	65.967	16.067	Ammarnäs	65	1996	2004	9	**0.019**	unadjusted non-linear	-3.2 (-18, 11)	0.29	0.41
FIN-004.0[b]	Northern pike	muscle	67.930	27.589	Lokka	328	1971	2006	16	**0.033**	length adjusted **linear** (length = Q25% = 44.8 cm)	-6.4		**1.07**

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. ****
FIN-004.1[b]	Northern pike	muscle	67.930	27.589	Lokka		1971	2006	16	**0.027**	length adjusted **linear** (length = Q75% = 55 cm)	-3.0		**1.07**
FIN-007[b]	Burbot	muscle	68.077	26.599	Porttipahta	184	1980	2006	12	**0.025**	length adjusted **linear** (length = 49.5 cm)	-4.3		0.57
FIN-009[b]	Northern pike	muscle	68.077	26.599	Porttipahta	378	1980	2006	13	**0.014**	weight adjusted non-linear (weight = 860 g)	-2.8		**0.81**
FIN-011.1[b]	Whitefish	muscle	68.077	26.599	Porttipahta	214	1980	2006	11	**0.008**	weight adjusted non-linear (weight = Q75% = 635 g)	-6.1	0.38	0.48

* CAN – Canada; FAR - Faroe Islands; FIN – Finland; GRE – Greenland; ICE – Iceland; NOR – Norway; SWE – Sweden. Addition of '.0' and '.1' to Time series ID is used to distinguish 'small/young' and 'large/old' individuals, respectively, in adjusted runs where the dataset was split into these two categories.

** p-value and best p method: If p for the non-linear component is significant, the p-value corresponds to the non-linear component and the 'best method' is described as non-linear, otherwise the p-value corresponds to that for the linear component p and 'best method' is described as 'linear'. The best method column also details any adjustments that were applied; Q25% = 25 percentile, Q50% = median, Q75% = 75 percentile value of the adjustment covariate, respectively.

*** Square root of the residual variance around the smoothed line.

**** Adequacy (the number of time points divided by the number of years to detect 5% annual change at 80% power) from unadjusted runs (bold font indicates values > 80%).

a. Mercury concentrations on a dry weight basis.

b. Samples collected from hydroelectric reservoirs, with known mercury contamination associated with releases from sediments during reservoir construction.

c. Before 1994, monitoring was based on salt fish (without head). A factor of 1.3 was applied to convert to fork length.

Table B. Non-significant trends results (cells with yellow highlight indicates sets with significant trends – see table A; cells with grey shading indicate sets with too few years for inclusion in the analyses).

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
ICE-001A[a]	Blue mussel	soft tissue	66.56667	-18.0167	Grimsey	11	1992	2004	11	0.823	unadjusted linear	.52 (-5.2, 6.3)	0.32	N (0.69)
ICE-002A[a]	Blue mussel	soft tissue	64.33333	-21.7167	Eyri, Hvalfjordur	17	1992	2004	13	0.884	unadjusted linear	.26 (-5.5, 6.0)	0.27	N (0.76)
ICE-003A[a]	Blue mussel	soft tissue	64.38333	-21.4333	Hvalstod, Hvalfjordur	13	1992	2004	13	0.888	unadjusted linear	-0.28 (-.6.8, 6.2)	0.33	N (0.68)
ICE-004A														
ICE-005A[a]	Blue mussel	soft tissue	64.01667	-22.15	Hvassahraun	12	1992	2004	12	0.882	unadjusted linear	-0.34 (-7.6, 6.9)	0.38	N (0.57)
ICE-006A														
ICE-007A[a]	Blue mussel	soft tissue	63.43333	-20.3167	Vestmannaeyjar	5	1992	1998	5	0.479	unadjusted non-linear	11 (-32, 54)	0.38	N (0.2)
ICE-008A[a]	Blue mussel	soft tissue	66.05	-23.15	Úlfsá, Skutulsfjordur	6	1997	2004	6	***** (0.609 linear)	unadjusted non-linear	-2.9 (-17, 12)	0.39	N (0.38)
ICE-009A[a]	Blue mussel	soft tissue	65.18333	-14	Mjóifjordur (head)	8	1996	2004	8	0.866	unadjusted linear	0.33 (-5.6, 6.3)	0.18	N (0.67)
ICE-010A[a]	Blue mussel	soft tissue	65.2	-13.8	Mjóifjordur (Hofsá)	9	1995	2004	9	0.809	unadjusted linear	0.71 (-6.6, 8.1)	0.27	N (0.6)
ICE-011A[a]	Blue mussel	soft tissue	65.26667	-13.5667	Mjóifjordur (Dalatangi)	6	1997	2004	6	0.107	unadjusted linear	-6 (-14, 2)	0.14	N (0.55)
ICE-012A[a]	Blue mussel	soft tissue	65.98333	-23.0333	Dvergasteinn, Álftafjordur	8	1996	2004	8	0.907	unadjusted linear	-0.2 (-8, 7.6)	0.19	N (0.57)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
ICE-013A[a]	Blue mussel	soft tissue	66.08333	-23.0333	Arnarnes Isafjardardjup	4	1992	1995	4	***** (0.567 linear)	unadjusted linear	-15 (-99, 78)	.00	N (0.18)
ICE-014	Atlantic cod	muscle	66.5	-13.5	NE Iceland	51	1990	2005	15	0.781	unadjusted linear	0.37 (-2.6, 3.3)	0.23	**Y (1.00)**
ICE-015														
ICE-016														
ICE-017	Atlantic cod	muscle	66.5	-24.5	NW Iceland	49	1990	2005	15	0.056	unadjusted linear	-2.8 (-5.8, 0.11)	0.25	**Y (1.07)**
ICE-018	Dab	muscle	64.5	-14	SE Iceland	15	1990	1999	6	0.115	unadjusted linear	-8.8 (-21, 3.1)	0.33	N (0.33)
ICE-019	Dab	muscle	63.5	-20.5	S Iceland	8	1993	2000	7	0.425	unadjusted linear	4.1 (-7.9, 16)	0.28	N (0.44)
ICE-020	Dab	muscle	64.5	-23	SW Iceland	15	1990	1994	5	***** (0.237 linear)	unadjusted linear	-13 (-40, 15)	0.19	N (0.33)
ICE-021	Dab	muscle	66.5	-24.5	NW Iceland	21	1990	1999	9	0.487	unadjusted linear	-3 (-13, 6.6)	0.26	N (0.5)
NOR-001A	Blue mussel	soft tissue	69.78333	30.18333	11A Brashavn	21	1998	2005	7	0.585	unadjusted linear	-2.3 (-12, 7.7)	0.21	N (0.47)
NOR-002	Blue mussel	soft tissue	70.06667	30.15	10A Skallneset	36	1994	2005	12	0.066	unadjusted linear	-2.1 (-4.5, 0.19)	0.10	**Y (1.2)**
NOR-003														
NOR-004	Atlantic cod	muscle	68.2	14.8	98B+F Lille Molla	244	1993	2004	10	0.937	length adjusted linear (length = Q50%)	0.2	0.35	N (0.59)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
NOR-005	Plaice	muscle	69.91667	29.85	10F Skoger°y	35	1997	2005	8	0.073	length adjusted **linear** (length = Q50%)	-3.6	0.11	N (0.67)
NOR-006	Plaice	muscle	68.2	14.8	98B+F Lille Molla	40	1993	2005	10	0.549	length adjusted **linear** (length = Q50%)	2.3	0.43	N (0.38)
CAN-001	Black-legged kittiwake	eggs	74	-90	Prince Leopold Island	21	1975	2003	6	0.432	unadjusted linear	1.0 (-2.3, 4.4)	0.25	N (0.35)
CAN-002	Northern fulmar	eggs	74	-90	Prince Leopold Island	36	1975	2005	8	0.071	unadjusted linear	1.2 (-0.15, 2.6)	0.618	N (0.67)
CAN-003														
CAN-004	Thick-billed murre	eggs	62.5	-83	Coats Island	20	1993	2005	4	***** (0.332 linear)	unadjusted	-1.6 (-6.8, 3.7)	.00	N (0.44)
CAN-005	Burbot	liver	61.1833	-135.2	Laberge	55	1996	2004	4	0.232	length adjusted linear (length = Q50%)	3.7	0.14	N (0.19)
CAN-006	Burbot	muscle	61.1833	-135.2	Laberge	86	2000	2004	4	0.54	length adjusted linear (length = Q50%)	4.8	0.21	N (0.29)
CAN-007	Lake whitefish	muscle	61.1833	-135.2	Laberge	37	1998	2002	4	0.197	length adjusted linear (length = Q50%)	-5.4	0.08	N (0.29)
CAN-008	Lake trout	muscle	61.1833	-135.2	Laberge	85	1993	2004	9	0.199	length adjusted linear (length = Q50%)	-1.6	0.11	N (0.69)
CAN-009	Lake trout	muscle	60.35	-136.367	Kusawa	77	1993	2005	7	0.299	length adjusted linear (length = Q50%)	-3.1	0.26	N (0.5)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
CAN-010	Lake trout	muscle	61.0833	-133.083	Quiet	53	1992	2003	5	0.158	length adjusted linear (length = Q50%)	-2.3	0.11	N (0.42)
CAN-011.0														
CAN-011.1														
CAN-012	Beluga	muscle	69.500	-133.586	Hendrickson Island	160	1981	2003	9	0.073 [b]	age adjusted linear (age = Q50%)	1.9	0.18	**Y (0.8)**
CAN-013	Beluga	muktuk	69.500	-133.586	Hendrickson Island	131	1993	2003	7	0.154 [b]	age adjusted linear (age = Q50%)	-0.1	0.24	N (0.75)
CAN-014	Beluga	kidney	69.500	-133.586	Hendrickson Island	126	1981	2001	7	0.104 [b]	age adjusted linear (age = Q50%)	4.3	0.35	N (0.47)
CAN-015	Beluga	MeHg - liver	69.49996	-133.586	Hendrickson Island	114	1993	2003	6	0.731	age adjusted linear (age = Q50%)	0.9	0.24	**Y (0.8)**
CAN-016	Beluga	MeHg - muscle	69.49996	-133.586	Hendrickson Island	73	1993	2001	4	0.323	age adjusted linear (age = Q50%)	3.8	0.18	N (0.55)
CAN-017	Beluga	MeHg - muktuk	69.49996	-133.586	Hendrickson Island	62	1993	2001	3	0.528	age adjusted linear (age = Q50%)	2.6	0.18	
CAN-018														
CAN-019	Ringed seal	liver	74.6833	-94.8333	Resolute Bay	54	2000	2005	3 [c]	0.315	age adjusted linear (age = Q50%)	7.7	0.16	
CAN-020	Ringed seal	muscle	70.65155	-117.726	Holman Island	122	1972	2004	6	0.419	age adjusted linear (age = Q50%)	-1.1	0.32	N (0.56)
CAN-021.0	Ringed seal	liver	70.65155	-117.726	Holman Island	142	1972	2004	7	0.087	age adjusted linear (age = Q25%)	2.9	0.36	N (0.46)
CAN-021.1														
CAN-022	Ringed seal	MeHg - muscle	70.65155	-117.726	Holman Island	68	1993	2003	4	0.281	age adjusted linear (age = Q50%)	3.3	0.18	N (0.27)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
CAN-023	Ringed seal	MeHg - liver	70.65155	-117.726	Holman Island	68	1993	2003	4	0.357	age adjusted linear (age = Q50%)	-9	0.63	N (0.22)
CAN-024	Ringed seal	muscle	71.97401	-125.253	Sachs Harbour	119	1987	2004	3 [c]	0.31	age adjusted linear (age = Q50%)	-4.9	0.35	N (0.24)
CAN-025	Ringed seal	liver	71.97401	-125.253	Sachs Harbour	150	1987	2004	4	0.28	age adjusted linear (age = Q50%)	-5.1	0.53	N (0.22)
CAN-026.0	Ringed seal	liver	69.9069	-92.2528	Arviat	116	1992	2003	3 [c]	0.738	age adjusted linear (age = Q25%)	-5.7	1.02	N (0.14)
CAN-026.1	Ringed seal	liver	69.9069	-92.2528	Arviat	116	1992	2003	3 [c]	0.392	age adjusted linear (age = Q75%)	11.2	0.62	N (0.14)
CAN-027.0														
CAN-027.1	Beluga	liver	66.124	-65.626	Pangnirtung	132	1984	2002	10	0.119	age adjusted linear (age = Q75%)	10	0.85	N (0.63)
CAN-028	Beluga	kidney	66.1236	-65.6264	Pangnirtung	122	1984	2002	9	0.177	age adjusted linear (age = Q50%)	2.8	0.24	N (0.63)
CAN-029.0	Beluga	liver	56.5333	-79.2333	Sanikiluaq	98	1994	2004	6	0.863	age adjusted linear (age = Q25%)	0.9	0.49	N (0.26)
CAN-029.1	Beluga	liver	56.5333	-79.2333	Sanikiluaq	98	1994	2004	6	0.297	age adjusted linear (age = Q75%)	-4	0.32	N (0.26)
CAN-030	Beluga	kidney	56.5333	-79.2333	Sanikiluaq	98	1994	2004	6	0.184	age adjusted linear (age = Q50%)	-3.6	0.22	N (0.32)
CAN-031	Beluga	liver	69.9069	-92.2528	Arviat (Eskimo Point)	106	1984	2003	4	0.133	age adjusted linear (age = Q50%)	3.9	0.26	N (0.21)
CAN-032	Beluga	kidney	69.9069	-92.2528	Arviat (Eskimo Point)	98	1984	2003	4	0.06	age adjusted linear (age = Q50%)	2.6	0.11	N (0.33)
CAN-033	Narwhal	liver	72.8	-77	Pond Inlet	144	1978	2004	8	0.661	age adjusted linear (age = Q50%)	0.4	0.23	N (0.44)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
CAN-034														
CAN-035	Narwhal	liver	73.0167	-85.1167	Arctic Bay	73	1983	2004	5	0.473	unadjusted[e] linear	-1.7 (-8.2, 4.9)	0.37	N (0.21)
CAN-036	Narwhal	kidney	73.0167	-85.1167	Arctic Bay	57	1986	2004	4	0.65	length adjusted linear (length = Q50%)	1	0.29	N (0.25)
CAN-037	Narwhal	liver	67.5667	-63.9	Broughton Island	83	1993	2004	5	0.4	length adjusted linear (length = Q50%)	1.8	0.17	N (0.38)
CAN-038.0	Narwhal	kidney	67.5667	-63.9	Broughton Island	81	1993	2004	5	0.599	length adjusted linear (length = Q25%)	-1.8	0.28	N (0.33)
CAN-038.1	Narwhal	kidney	67.5667	-63.9	Broughton Island	81	1993	2004	5	0.658	length adjusted linear (length = Q75%)	-2.3	0.43	N (0.33)
CAN-039	Burbot	muscle	66.25699	-128.635	Fort Good Hope	238	1999	2005	7	0.845	length adjusted linear (length = Q50%)	-0.9	023	N (0.62)
CAN-040	Burbot	liver	66.25699	-128.635	Fort Good Hope	234	1999	2005	7	0.267	length adjusted linear (length = Q50%)	6.5	0.27	N (0.64)
CAN-041	Lake trout	muscle	61.50015	-114.001	Great Slave Lake - East Arm	68	1995	2005	7	0.434	length adjusted linear (length = Q50%)	2.8	0.27	N (0.47)
CAN-042A.0	Lake trout	muscle	61.17099	-113.672	Great Slave Lake - West Basin	152	1979	2005	11	0.637	length adjusted linear (length = Q25%)	0.8	0.44	N (0.44)
CAN-042A.1	Lake trout	muscle	61.17099	-113.672	Great Slave Lake - West Basin	152	1979	2005	11	0.489	length adjusted linear (length = Q75%)	1.1	0.39	N (0.44)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
CAN-043	Burbot	muscle	61.500	-114.001	Great Slave Lake - East Arm	46	1999	2004	5	0.091[b]	length adjusted linear (length = Q50%)	12.1	0.19	N (0.56)
CAN-044A	Burbot	muscle	61.17099	-113.672	Great Slave Lake - West Basin	78	1975	2004	11	0.376	length adjusted linear (length = Q50%)	-0.6	0.19	N (0.6)
CAN-045	Burbot	muscle	60.00015	-111.884	Slave River		2000	2002	3[f]					
CAN-046	Ringed seal	kidney	71.97401	-125.253	Sachs Harbour		1987	2005	4 (only 2 with data)[f]					
CAN-047	Moose	muscle	63	-135	Yukon		1994	2005	6 (only 2 with data)[f]					
CAN-048	Moose	liver	63	-135	Yukon	17	1993	2005	5	***** (0.407 linear)	unadjusted non-linear	-13 (-55, 29)	0.94	N (0.17)
CAN-049														
CAN-050	Caribou	kidney	63	-135	Carcross herd	7	1999	2002	4	***** (0.697 linear)	unadjusted non-linear	4.9 (-43, 52)	0.00	N (0.27)
CAN-051	Caribou	kidney	63	-135	Coal River herd	8	1996	2002	5	***** (0.501 linear)	unadjusted non-linear	5.0 (-16, 26)	0.35	N (0.29)
CAN-052	Caribou	kidney	63	-135	Finlayson herd	61	1992	2001	8	0.436	age adjusted linear (age = Q50%)	6.2	0.62	N (0.32)
CAN-053	Caribou	kidney	63	-135	Hart River herd	11	1996	2000	4	***** (0.704 linear)	unadjusted non-linear	-8.2 (-90, 74)	0.00	N (0.17)
CAN-054	Caribou	kidney	63	-135	Nahanni herd	8	1993	2003	6	0.631	unadjusted linear	4.7 (-20, 30)	0.76	N (0.21)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
CAN-055	Caribou	kidney	63	-135	Pelly herd	6	1996	2003	4	***** (0.429 linear)	unadjusted non-linear	11 (-37, 59)	0.00	N (0.16)
CAN-056														
CAN-057	Caribou	kidney	63	-135	Redstone herd	7	1993	2003	4	***** (0.164 linear)	unadjusted non-linear	19 (-18, 55)	0.00	N (0.15)
CAN-058.0	Caribou	kidney	63	-135	Tay herd	33	1993	2004	5	0.126	age adjusted linear (age = Q25%)	-9.7	0.41	N (0.43)
CAN-058.1	Caribou	kidney	63	-135	Tay herd	33	1993	2004	5	0.076	age adjusted linear (age = Q75%)	-7.2	0.24	N (0.43)
CAN-059	Caribou	kidney	63	-135	Wolf Lake herd	8	1993	2003	5	0.001	unadjusted non-linear	7.7 (-8.3, 24)	0.5	N (0.26)
CAN-060	Northern pike	muscle	61.50015	-114.001	Great Slave Lake - East Arm	40	1999	2002	4	0.457	length adjusted linear (length = Q50%)	1.9	0.05	N (0.5)
CAN-061A														
CAN-062	Walrus	liver	61.17099	-113.672	Igloolik	60	1982	1996	5	0.496	age adjusted linear (age = Q50%)	2.1	0.34	N (0.4)
CAN-063	Walrus	kidney	61.17099	-113.672	Igloolik	61	1982	1996	5	0.755	age adjusted linear (age = Q50%)	0.5	0.17	N (0.4)
GRE-001.0	Ringed seal	liver	77.48	-69.36	Avanersuaq	102	1984	2004	5	0.106	age adjusted **linear** (age= Q25% = 1 yr old)	8.0	0.21	N (0.21)
GRE-001.1														
GRE-002														
GRE-017	Ringed seal	liver	70.483	-21.95	Ittoqqortoormiit	182	1986	2004	7	0.252	age adjusted **linear** (age = 4 yr old)	3.5	0.35	N (0.35)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
GRE-003.0	Polar bear	muscle	70.483	-21.95	Ittoqqortoormiit	149	1986	2004	9	0.296	age adjusted non-linear (age = Q25% = 3 yr old)	1.5	0.43	N (0.56)
GRE-003.1	Polar bear	muscle	70.483	-21.95	Ittoqqortoormiit		1986	2004	9	0.109	age adjusted **linear** (age = Q75% = 7 yr old)	3.0	0.50	N (0.56)
GRE-004	Polar bear	liver	70.483	-21.95	Ittoqqortoormiit	164	1983	2004	12	0.156	age adjusted non-linear (age = 4.5 yr old)	0.8	1.09	**Y (0.92)**
GRE-005	Polar bear	kidney	70.483	-21.95	Ittoqqortoormiit	161	1983	2004	12	0.077	age adjusted linear (age = 4.5 yr old)	-2.7	0.67	N (0.63)
GRE-006														
GRE-007	Polar bear	muscle	77.48	-69.36	Avanersuaq				3 [f]					
GRE-008	Polar bear	liver	77.48	-69.36	Avanersuaq				3 [f]					
GRE-009	Polar bear	kidney	77.48	-69.36	Avanersuaq				3 [f]					
GRE-010	Polar bear	hair	77.48	-69.36	Avanersuaq				3 [f]					
GRE-011	Guillemot	liver	69.12	-49.78	Qeqartarsuaq				5 [g]					N (0.35)
GRE-012	Arctic char	muscle	65.53	-38.97	Qaqortoq / Isortoq	89	1994	2004	4	0.319	length adjusted non-linear (length = 39 cm)	0.5	0.44	N (0.27)
GRE-013	Shorthorn sculpin	liver	70.483	-21.95	Ittoqqortoormiit	85	1985	2002	5	0.128	unadjusted linear	3.9	0.36	N (0.33)
GRE-014.0	Shorthorn sculpin	liver	77.48	-69.36	Avanersuaq	71	1987	2004	4	0.500	length adjusted linear (length = Q25% = 24 cm)	13.2	<0.13	N (0.25)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
GRE-014.1	Shorthorn sculpin	liver	77.48	-69.36	Avanersuaq		1987	2004	4	0.367	length adjusted linear (length = Q75% = 32 cm)	5.5	0.15	N (0.25)
GRE-015.0	Shorthorn sculpin	liver	69.12	-49.78	Qeqertarsuaq	145	1994	2004	7	0.520	length adjusted linear (length = Q25% = 24.8 cm)	-4.1	0.32	N (0.29)
GRE-015.1	Shorthorn sculpin	liver	69.12	-49.78	Qeqertarsuaq		1994	2004	7	0.701	length adjusted non-linear (length = Q75% = 32.5 cm)	<0.01	0.27	N (0.29)
GRE-016	Walrus	liver	77.48	-69.36	Avanersuaq	61	1977	2003	7	0.151	unadjusted linear	<0.01	0.41	N (0.35)
FAR-001A	Pilot whale (immature)	muscle	62	-7.5	Faroe Island	78	1997	2002	6	0.132	unadjusted non-linear	-3.5	0.75	N (0.3)
FAR-001B	Pilot whale (females)	muscle	62	-7.5	Faroe Island	123	1997	2002	6	0.247	unadjusted linear	-1.5	0.33	N (0.3)
FAR-001C	Pilot whale (males)	muscle	62	-7.5	Faroe Island	54	1997	2002	6	0.162	unadjusted non-linear	-2.2	0.40	N (0.3)
FAR-002.0														
FAR-002.1	Atlantic cod	muscle	62	-7.5	Faroe Island	792	1979	1989	11	0.222	length adjusted linear (length = Q75% = 85.8 cm)	-3.7	0.69	**Y (0.9)**
FAR-003	Shorthorn sculpin	liver	62.0645	-6.905	Faroe Islands - Kaldbaksfjordur	55	1999	2004	4	0.001	length adjusted non-linear (length = 26.5 cm)	-21	0.00	N (0.11)
FAR-004	Guillemot	eggs	61.76833	-6.80167	Faroe Islands - Skuvoy/Koltor	74	1999	2004	4	0.116	unadjusted non-linear	0.2	0.25	N (0.25)
SWE-001	Northern pike	muscle	65.7	17.133	Lake Storvindeln	316	1968	1999	29	0.001 [b]	weight adjusted non-linear (weight = Q25% = 945 g)	-.18% (-.96, .61)	0.12	**Y (2.23)**

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
SWE-002														
SWE-003														
SWE-004	Reindeer	liver	68.3	18.65	Abisko	254	1981	2004	21	0.640	unadjusted linear	0.7 (-2.3, 3.7)	0.38	**Y (1.05)**
SWE-005														
SWE-006	Reindeer	liver	65.967	16.067	Ammarnäs	116	1983	2004	9	0.156	unadjusted non-linear	-8.7 (-22, 4.3)	0.34	N (0.45)
FIN-001[d]	Burbot	muscle	67.9296	27.5889	Lokka	172	1979	2006	14	0.144	length adjusted non-linear (length = 44 cm)	-2.3	0.67	N (0.56)
FIN-002[d]	Ide	muscle	67.9296	27.5889	Lokka	27	1979	1985	6	0.542	unadjusted linear	-4.5	0.33	N (0.35)
FIN-003.0[d]	Perch	muscle	67.9296	27.5889	Lokka	108	1980	2006	12	0.300	length adjusted linear (length = Q25% = 22.2 cm)	-2.4	0.46	N (0.52)
FIN-003.1[d]	Perch	muscle	67.9296	27.5889	Lokka		1980	2006	12	0.267	length adjusted linear (length = Q75% = 29cm)	-2.1	0.52	N (0.52)
FIN-004[d]														
FIN-005[d]	Roach	muscle	67.9296	27.5889	Lokka	57	1978	1994	7	0.186	length adjusted linear (length = 26 cm)	4.5	0.37	N (0.47)
FIN-006[d]	Whitefish	muscle	67.9296	27.5889	Lokka	81	1982	2004	8	0.283	unadjusted non-linear	-1.3	0.62	N (0.57)
FIN-007														
FIN-008.0[d]	Pearch	muscle	68.0767	26.5988	Porttipahta	100	1980	2006	10	0.117	weight adjusted linear (weight = Q25% = 149.4 g)	-4.0	0.38	N (0.42)

Time series ID*	Species	Tissue	Lat.	Long.	Location	n	First year	Last year	Number of years with data	p-value**	Best p method**	% annual change (lower CL, upper CL)	SD (sm) ***	Adeq. >80% ****
FIN-008.1[d]	Pearch	muscle	68.0767	26.5988	Porttipahta				10	0.088	weight adjusted linear (weight = Q75% = 370 g)	-3.3	0.45	N (0.42)
FIN-009														
FIN-010 [d]	Roach	muscle	68.0767	26.5988	Porttipahta	38	1980	1985	5	0.659	unadjusted linear	4.3	0.28	N (0.29)
FIN-011.0[d]	Whitefish	muscle	68.0767	26.5988	Porttipahta	214	1980	2004	11	0.068	weight adjusted linear (weight =Q25% = 398 g)	-4.6	0.50	N (0.48)
FIN-011.1 [d]														

* CAN – Canada; FAR - Faroe Islands; FIN – Finland; GRE – Greenland; ICE – Iceland; NOR – Norway; SWE – Sweden.

** p-value and best p method: If p for the non-linear component is significant, the p-value corresponds to the non-linear component and the 'best method' is described as non-linear, otherwise the p-value corresponds to that for the linear component p and 'best method' is described as 'linear'. The best method column also details any adjustments that were applied; Q25% = 25 percentile, Q50% = median, Q75% = 75 percentile value of the adjustment covariate, respectively.

*** Square root of the residual variance around the smoothed line.

**** Adequacy (the number of time points divided by the number of years to detect 5% annual change at 80% power) from unadjusted runs.

***** Smoother residuals worse than total variance.

a. Mercury concentrations on a dry weight basis.

b. Unadjusted total mercury concentrations demonstrated a statistically significant temporal trend, however, when total mercury concentrations were adjusted for age, length or weight, the trend was no longer found to be significant.

c. One or more years of data lacked covariate data, so only 3 years of data remained in the 'covariate adjusted' time-series.

d. Samples collected from hydroelectric reservoirs, with known mercury contamination associated with releases from sediments during reservoir construction.

e. No covariate data available.

f. Less than 3 years with data available; trend analysis not performed.

g. Dataset unbalanced with respect to age; trend analysis not performed.