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Temporal change estimation of mercury concentrations in northern pike (*Esox lucius* L.) in Swedish lakes

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ABSTRACT

Adequate temporal trend analysis of mercury (Hg) in freshwater ecosystems is critical to evaluate if actions from the human society have affected Hg concentrations ([Hg]) in fresh water biota. This study examined temporal change in [Hg] in Northern pike (*Esox lucius* L.) in Swedish freshwater lakes between 1994 and 2006. To achieve this were lake-specific, multiple-linear-regression models used to estimate pike [Hg], including indicator variables representing time and fish weight and their interactions. This approach permitted estimation of the direction and magnitude of temporal changes in 25 lakes selected from the Swedish national database on Hg in freshwater biota. A significant increase was found in 36% of the studied lakes with an average increase in pike [Hg] of 3.7 ± 6.7% per year that was found to be positively correlated with total organic carbon. For lakes with a significant temporal change the dataset was based on a mean of 30 fish, while for lakes with no temporal change it was based on a mean of 13 fish.

1. Introduction

Mercury (Hg) exposure from consumption of freshwater piscivorous fish is considered to be an increasing health problem, as this constitutes the primary route of methyl-Hg exposure in humans and wildlife (Mergler et al., 2007). Environmental and health advisory limits (0.2–0.5 mg kg^{-1}) (UNEP, 2002) are exceeded in thousands or tens of thousands of lakes of many countries and regions, e.g. Scandinavia (Håkansson et al., 1988; Lindqvist et al., 1991; Johansson et al., 2001; Munthe et al., 2007), Canada (Evans et al., 2005b) and the USA (Bahnick et al., 1994; Hinck et al., 2009; Scudder et al., 2009). Using data from a Swedish environmental monitoring program, we examined temporal changes in [Hg] in freshwater northern pike (*Esox lucius* L.) from the late 1990s into the early 21st century.

1.1. Temporal variation in the past

Temporal variation from the 1970s onwards in fish [Hg] from inland freshwaters have been examined in local (Hrabik and Watras, 2002; Suchanek et al., 2008; Gantner et al., 2009; Azim et al., 2011), regional (Kamman et al., 2005; Madsen and Stern, 2007; Levinton and Pochron, 2008; Hinck et al., 2009; Monson, 2009; Bhavsar et al., 2010) and national datasets from Scandinavia (Paasivirta and Linko, 1980; Håkansson et al., 1988; Johansson et al., 2001) and North America (Schmitt and Brumbaugh, 1990; Johnston et al., 2003; Muir et al., 2005; Chalmers et al., 2011). At the end of the 1970s, no significant temporal change in fish [Hg] was detected in Finnish (Paasivirta and Linko, 1980) and US (Schmitt and Brumbaugh, 1990) national monitoring programmes. However, data obtained from Swedish monitoring programmes for Northern pike revealed increasing [Hg] from 1967 to 1985 (Håkansson et al., 1988). The increase found in fish [Hg] was followed by an average decrease of about 20% from the early 1980s to the late 1990s (Johansson et al., 2001). A similar decreasing change as that found in Swedish lakes was also reported in other studies of long-term fish [Hg] changes in the USA (Hrabik and Watras, 2002; Madsen and Stern, 2007; Levinton and Pochron, 2008) and Canada (Johnston et al., 2003; Hinck et al., 2009) during the same period. These decreasing changes have mainly been ascribed to reductions in anthropogenic Hg emissions (Johansson et al., 2001; French et al., 2006; Rasmussen et al., 2007; Monson, 2009). However, temporal changes in more recent data (2000–2010) indicate a reversal, with increasing [Hg] reported in species in certain cases of North America (Muir et al., 2005; French et al., 2006; Monson, 2009), although decreases have also been noted between 1992–1993 and 2005–2006 in lakes within the Adirondack region (Dittman and Driscoll, 2009). A bayesian modelling framework identified an increasing trend of [Hg] in lake Erie fish becoming particularly evident after the mid-1990s (Azim et al., 2011).

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1.2. Fish [Hg]-weight dependence

The main problem in temporal change analysis of [Hg] in freshwater fish is separating the time-dependent variation in [Hg] from nontime-dependent variation (Wente, 2004; Harris et al., 2007). The latter most often constitutes the major variation and operates both within and between lakes. Hg is biomagnified due to trophic transfer and bioaccumulates over life of the fish thus creating variation in [Hg] that will be connected with the trophic position and size of the fish (Meili, 1991; Chasar et al., 2009). The correlation between fish [Hg] and fish size is often so great that most temporal change are masked if this size-dependent variation is not accounted for in the data analysis (Watras et al., 1998; Wente, 2004; Suchanek et al., 2008; Chasar et al., 2009). Consideration to an apparent correlation between fish size and [Hg] can be done *a priori*, separately from the temporal change analysis, by normalisation of fish Hg data to a standard fish size (Johansson et al., 2001; Hrabik and Watras, 2002; Evans et al., 2005a; Suchanek et al., 2008; Monson, 2009). Correction of fish [Hg] is done mainly to (1) reduce the variability in standard [Hg], (2) reduce the error of the normalised mean, and (3) facilitate comparison between fish populations contaminated to a different extent (Stöcker, 1993). The procedure assumes a linear relationship between size and the logarithmic [Hg]. A number of studies have not adjusted the data for covariation between fish size and [Hg] in temporal change analysis, reasoning that no obvious gain is obtained by size adjustment of data (Bignert et al., 2004) or that these relationships are not consistent across years (Sharma et al., 2008; Gantner et al., 2009), or not taking possible covariation into consideration in any part of the analysis (Schmitt and Brumbaugh, 1990).

1.2.1. Statistical tools

The introduction of fish size within statistical test, such as analysis of covariation (ANCOVA) or multiple linear regression (MLR) techniques, excludes any normalisation of [Hg] to fish size. In a study on the correlation between [Hg] and length of Arctic char (*Salvenius alpinus* L.) by ANCOVA revealed significant length $*$ year interaction, and adjustment of [Hg] by length was considered unsuitable for temporal comparison for this reason (Gantner et al., 2009). An approach with MLR models was applied by Levinton and Pochron (2008). Their regression models included variables for sampling location, time, and fish size. The use of fish size as a continuous variable in the statistical analysis assumes that conditions for linear regression between fish size and [Hg] is fulfilled (Fox, 1997) and that there actually exist such an correlation. Instead of using fish size as a continuous variable Johnston et al. (2003) used least square means for each of three fish length classes in recent and historic periods to determine whether the observed temporal changes were influenced by the size of the fish. Indicator variables for defined fish size classes in MLR statistics is not error prone in subsequent tests of different lakes and offset the variation in fish weight in the temporal change analysis.

1.3. Hypothesis

The hypothesis for the present study was that [Hg] in freshwater fish generally changed in lakes from the end of the last decade until 2005 and this temporal change was context dependent and differ depending on lake characteristics. Due to the magnitude of between-lake variation in mean fish [Hg] lake-specific temporal change analysis is performed. Preliminary analysis of the data revealed inconsistencies in the covariation between fish size and [Hg]. Therefore the fish size was included using three defined fish size classes in the temporal change analysis. To test our hypothesis, datasets on pike taken from the Swedish environmental monitoring program were examined by MLR. Relationships between the annual change in pike [Hg] and lake water chemistry variables (pH and total organic carbon (TOC)) were also investigated.

2. Materials and methods

2.1. Description of datasets

All data were taken from the national Swedish database on Hg in freshwater biota, retrievable online at www.ivl.se (IVL Swedish Environmental Research Institute). The initial dataset obtained included data on lake name, geographical coordinates, sampling date (year–month–day), pike size (weight (kg) and length (cm)) and total [Hg] (mg kg^{-1}, wet weight), covering the period 1993–2008. Only Hg data from muscle tissue were accepted for inclusion in the dataset. The initial dataset consisted of 4959 with fish data entries from 815 lakes. Water chemistry characteristics from national monitoring stations (http://info1.ma.slu.se/db.html; 2010-10-20) in the lakes analysed, covering the period examined for temporal changes, were used to further examine relationships between temporal changes in fish [Hg] and variation in pH, water colour (mg Pt L^{-1}) and TOC (mg L^{-1}) between lakes.

2.2. Data quality assessment and data selection

Data entered into the original database were not restricted by any quality protocol for collecting the fish samples or Hg analysis. Data with arbitrary or unclear assignment in length and weight were corrected, if possible, or excluded before further analysis. Correction or exclusion of data outliers was only carried out after validation of the dataset by linear regression analysis of weight–length or Hg-size relationships of the whole dataset, in individual lakes when possible. No data were rejected solely on the suspicion that systematic errors had occurred in the dataset because of different analytical methods used or errors during collection of the fish.

For data from a particular lake to qualify for inclusion in the analysis, two criteria had to be met: (1) The lake had to be sampled at least once during each of two 5-year periods and (2) within each 5-year period at least three pike had to be sampled. These criteria resulted in nine possible combinations with respect to 5-year periods and number of fish samples for the periods 1993–2000 and 2001–2008 (Table 1). The overall model results were not sensitive for the exact combination of periods chosen for comparison of the fish [Hg] (data not shown). The sign of the model coefficients was the same for the different model combinations, but the overall significance of model coefficient increased with increasing distance between the periods chosen. The final dataset, consisting of 25 lakes and 498 fish samples, made it possible to compare a recent period (2001–2005) with an earlier period (1994–1998) separated by at least 3 years, a time lapse also used by Håkansson et al. (1988).

2.3. Statistical analysis

All calculations were carried out using JMP 8.0.1 (SAS Institute Inc., Cary, NC, USA). The distribution of pike [Hg] was assessed according to the Shapiro–Wilks test (Shapiro and Wilk, 1965),

Table 1
Number of lakes and number of pike data (within brackets) available according to the selection criteria described in Section 2.2, in combinations of periods covering 5 years before and after the year 2000.

	2001–2005	2002–2006	2003–2007
1994–1998	25 (498)	17 (366)	8 (262)
1995–1999	23 (490)	15 (373)	8 (274)
1996–2000	26 (548)	19 (439)	11 (318)