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Is it appropriate to composite fish samples for mercury trend monitoring and consumption advisories?

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ABSTRACT

Monitoring mercury levels in fish can be costly because variation by space, time, and fish type/size needs to be captured. Here, we explored if compositing fish samples to decrease analytical costs would reduce the effectiveness of the monitoring objectives. Six compositing methods were evaluated by applying them to an existing extensive dataset, and examining their performance in reproducing the fish consumption advisories and temporal trends. The methods resulted in varying amount (average 34–72%) of reductions in samples, but all (except one) reproduced advisories very well (96–97% of the advisories did not change or were one category more restrictive compared to analysis of individual samples). Similarly, the methods performed reasonably well in recreating temporal trends, especially when longer-term and frequent measurements were considered. The results indicate that compositing samples within 5 cm fish size bins or retaining the largest/smallest individuals and compositing in-between samples in batches of 5 with decreasing fish size would be the best approaches. Based on the literature, the findings from this study are applicable to fillet, muscle plug and whole fish mercury monitoring studies. The compositing methods may also be suitable for monitoring Persistent Organic Pollutants (POPs) in fish. Overall, compositing fish samples for mercury monitoring could result in a substantial savings (approximately 60% of the analytical cost) and should be considered in fish mercury monitoring, especially in long-term programs or when study cost is a concern.

1. Introduction

Mercury is a contaminant of global concern (UNEP, 2013a). Virtually every fish in North America, and possibly worldwide, contains mercury (Stahl et al., 2009; Depew et al., 2013; Evers et al., 2013). Consumption of fish is generally a dominant route of human exposure to mercury (UNEP/WHO, 2008). Mercury is responsible for the most number of restrictive fish consumption advisories, at least in North America (e.g., USEPA, 2013a,b; OMOECC, 2015). Due to spatial variation in fish mercury levels, location-specific advisories are typically provided (e.g., USEPA, 2013a; OMOECC, 2015). Since mercury levels vary by fish species and size (Gewurtz et al., 2011b), monitoring efforts to issue fish consumption advisories and track long-term changes require collection and analysis of a variety of fish spanning their natural size range (USEPA, 2013b). As a result, the total number of annual samples required to adequately monitor fish mercury levels for numerous locations can range from hundreds to tens of thousands.

Due to analytical costs, most contaminant studies limit sample size by reducing the fish species monitored, replication of samples, sampling frequency and/or study period; however, these options are generally not suitable for agencies that rely on the data for long-term trend monitoring and issuing of fish consumption advisories aimed at protecting human health (Gewurtz et al., 2011a). Further, Article 19 of the recently formulated Minamata Convention on Mercury requires parties to develop and improve geographically representative mercury monitoring in environmental media, including fish (UNEP, 2013b). In less than a decade, monitoring data will be called upon to assist in the implementation and evaluation of the convention, which emphasizes the importance of improving monitoring efforts to optimize both the quality of the programs as well as costs.

To decrease program costs, combining multiple temporally or spatially discrete samples, widely known as composites, has been suggested as an effective alternative to chemical analysis on individual samples (USEPA, 2002; Gewurtz et al., 2011a). In addition to substantially reducing analytical cost, the data collected through compositing samples can provide wider temporal and spatial coverage without increasing the sample count. The analysis of data may give more representative estimates of mean concentrations than can the same number of

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discrete samples, albeit at the cost of variability in the observations (USEPA, 2002).

There are several potential approaches to compositing fish contaminant monitoring samples that incorporate different dimensions of the study, such as time (within/across years), location, fish species, and fish size. The optimal compositing approach would be one that reduces the total number of samples for analysis without compromising the objectives of the monitoring program. In addition, the composite method chosen should follow assumptions that correspond to the statistical analysis that is ultimately applied to the data. Several studies have used compositing as a part of their designs for both organic and inorganic contaminants in all media including biota (Rajagopal and Williams, 1989; Turle and Collins, 1992; Blomqvist, 2001; Braune and Noble, 2009; Gewurtz et al., 2011a). However, to our knowledge, a comprehensive study investigating the effectiveness of various compositing approaches for monitoring mercury in fish is lacking in the literature, especially for programs designed to generate fish consumption advice, where variability and the presence of outliers can affect overall risk (Gewurtz et al., 2011a).

In this study, we evaluate six methods of compositing fish samples by examining their performance if they would have been utilized instead of collecting >220,000 individual mercury measurements for >3000 locations by the Province of Ontario, Canada over nearly 50 years. The effectiveness of the composite methods was evaluated by comparing the fish consumption advisories and temporal trends from individual measurements (current sampling design) with those from estimated composite values, calculated by averaging the individual measurements included in each composite. The findings of the study determine whether a compositing method can effectively minimize costs for regular, long term, large scale monitoring programs and set advisories for fish consumption.

2. Methods

2.1. Compositing methods

Fish mercury levels vary by species and size, and can change seasonally as well as over time under the influence of a variety of internal and external factors, such as bioenergetics and ambient water chemistry (Bhavsar et al., 2010; Azim et al., 2011; Gewurtz et al., 2011a; Stern et al., 2012; Greenfield et al., 2013). As such, we opted to group species-specific samples collected during the same sampling event within the composites.

There is a well-known relationship between mercury concentrations and fish size that is typically described by the power-series regression (Gewurtz et al., 2011b). As such, similar sized samples could be considered for creating a composite sample. However, the resultant fish size range (i.e., maximum–minimum fish lengths) would likely be less than the regular, individual measurements. This could result in trimming of a regression at the extreme ends, and thereby loss of advisories for certain fish sizes. Alternatively, if one or two of the largest and/or smallest individuals are retained with all other samples being composited, then the fish size range could be captured, and a power series regression between fish length and composited mercury concentrations might be improved.

Compositing of 3, 5, 7, 10 or more samples have been used in many studies (Hites et al., 2004; Carlson and Swackhamer, 2006; French et al., 2011; Pantazopoulos et al., 2013). Since a collection of about 20 fish samples per species and sampling event over a possible maximum size range is generally considered a preferred method for mercury monitoring (e.g., Gewurtz et al., 2011a), compositing more than 5 samples (i.e., having less than four composites), may not be sufficient for characterizing the fish size/mercury relationships. Alternatively, compositing samples within a narrow size range (e.g., 35–40 cm, 40–45 cm and so on) regardless of the number of samples within that size range may be appropriate as the impact on the fish size/mercury relationship would likely be minimal.

Based on the above notes, we considered six compositing methods: (1) composite samples in batches of five in the order of decreasing fish size (Fig. 1a, b), (2) retain individual samples for the largest and smallest fish and composite samples in between in batches of five in order of decreasing fish size (Fig. 1a, c), (3) retain the two largest and smallest individual samples and composite the samples in between in batches of five in order of decreasing fish size (Fig. 1a, d), (4) retain the largest and smallest individual samples and composite the samples in between in batches of three in order of decreasing fish size (Fig. 1a, e), (5) retain the two largest and smallest individual samples and composite the samples in between in batches of three in the order of decreasing fish size (Fig. 1a, f), and (6) composite samples within a 5 cm size range (Fig. 1a, g).

2.2. Data source

The above described compositing methods were evaluated by simulating composite data from the individual fish measurements, assuming that the same mass of each fish is added to the composite. For this purpose, we used an extensive and consistent fish mercury dataset comprising 223,318 individual, widely varying measurements for skinless, boneless dorsal fillets of >10 cm fish of 66 fish species (Table S1) collected by the Ontario Ministry of the Environment and Climate Change (OMOECC), Canada in partnership with the Ontario Ministry of Natural Resources and Forestry and other agencies over nearly 50 years (1967–2014) from >3000 locations in the Province of Ontario, Canada, that spans 41° to 56° N and 74° to 95° W (Fig. S1). The samples were analyzed for total mercury using acid digestion and cold vapor flameless atomic absorption spectroscopy as described in detail by Bhavsar et al. (2010). The dataset contained 16,900 species/location/year combinations for 6440 sampling events (location/year) and varied widely (1 to 274) in the number of individual samples for a species in a sampling event (species/location/year) (Fig. S2).

2.3. Statistical analysis

The performance of each composite method in comparison to the regular, individual measurements was evaluated based on its accuracy in reproducing the fish consumption advisories as well as the direction and magnitude of the long-term temporal trends. As illustrated in Fig. S3, a power series regression was conducted for each of 16,900 species/location/year-specific sampling events using the regular, individual measurements as well as the composite values calculated using the six methods considered in this study. Using these total 118,300 power series regressions (i.e., 16,900 × 7), fish mercury levels were calculated at 5 cm intervals for the available size range in each species-specific sampling event (Fig. S3). These mercury concentrations were used in calculating fish consumption advisories using the benchmarks for the general population and sensitive population (children and women of child-bearing age), which is the standard method used by the Province of Ontario, Canada (Table S2, Fig. S3). Advisories for each 5 cm interval calculated using the six composite methods were compared with those from the regular, individual measurements (Table S4), and classified into three categories: 1) same, 2) more restrictive, and 3) less restrictive..

For a comparison of temporal trend analyses from the regular and composite methods, rates of changes in fish mercury levels (μg/g decade) were calculated using the slope of the linear relationship between year and mercury concentration standardized to a fish length. Since the purpose is to compare rates from the regular and composite methods, appropriateness of a linear regression is essentially a moot point (Azim et al., 2011). Since a temporal trend analysis is typically conducted on a suitable indicator species with good monitoring data, four species, namely Lake Trout (*Salvelinus namaycush*), Walleye (*Sander*