Abstract. Lake Murray is a large reservoir in central South Carolina, USA, that was constructed by impounding the Saluda River in the 1920’s and 1930’s. In the 1990’s a determination was made that the earthen dam was potentially vulnerable to failure in the event of a large earthquake, and to mitigate this risk a backup dam was constructed between 2003 and 2005. This required the drawdown of Lake Murray water levels until the dam construction was complete. In 2003 sterile grass carp (Ctenopharyngodon idella Valenciennes) were introduced to Lake Murray as a control for aquatic invasive plants that, combined with the drawdown, proved to be effective. Also in 2003 higher precipitation was recorded in South Carolina after several years of drought. During this period the South Carolina Department of Health and Environmental Control’s (SCDHEC) fish tissue-monitoring program collected tissue data in Lake Murray, which provided a unique opportunity to assess the effects of these multiple events on fish tissue mercury (Hg). Two sites were monitored annually over these years with one being close to the dam (S-273), and the other was further up the reservoir (S-223) in Newberry County. In 2004 tissue Hg in largemouth bass (Micropterus salmoides Lacepede) at S-223 was elevated relative to previous years, while at S-273 levels were depressed. In 2005 tissue Hg levels fell at S-223 while they were found to increase at S-273. The two locations on Lake Murray are very different with regard to their physical, chemical, and biological makeup, all of which likely contributed to the patterns observed in fish tissue Hg levels over the study period.

INTRODUCTION

Environmental mercury (Hg) contamination has become a global concern because of its ability to be transported in the atmosphere and, once deposited, to bioaccumulate in aquatic organisms (Mergle et al., 2007). Once atmospheric Hg is deposited, biological processes in the soil and water can cause the formation of methylmercury (MeHg); it is this form that can be biomagnified in fish and other organisms. The South Carolina Department of Health and Environmental Control (SCDHEC) began collecting and analyzing fish for tissue contaminants in 1976. Over the years total Hg has been found in many fish species and waterbodies across the State at levels that have triggered fish consumption advisories (SCDHEC, 2010). This comprehensive statewide monitoring program has been used primarily to survey the waters of the State and issue annual fish consumption advisories. But, as is the case with many large environmental monitoring datasets, insight can be gained into complex issues when data taken for other purposes are linked and evaluated together. Examples of this approach include Glover et al. (2010) in South Carolina, Sackett et al. (2009) in North Carolina, Kammen et al. (2005) in the northeastern USA and southeastern Canada, and Ryple (2010) in Wisconsin, where large datasets were used to evaluate the effects of landscape characteristics or water chemistry data on fish tissue Hg levels.

Numerous studies have shown that the methylation efficiency of waterbodies and surrounding landscapes is extremely important to the amount of Hg that enters the food chain (Munthe et al., 2007). Various physical,
chemical, and biological characteristics of a waterbody and the surrounding landscape have been shown to play critical roles in the amount of Hg that can potentially be found in the flesh of aquatic creatures. The location of the waterbody within a particular ecoregion and the type and size of the aquatic system (e.g., reservoirs versus rivers) can be a good predictor of the amount of Hg within fish tissue in South Carolina (Glover et al., 2010). Furthermore, the drying and rewetting of soils has been shown to increase the methylation efficiency of aquatic systems (Evers et al., 2007). Gilmour et al. (2004) demonstrated that the oxidation of reduced sulfur is stimulated when dried soils are reinundated, causing a flux of methylmercury formation. Glover et al. (2010) added support to this hypothesis by showing an increase in fish tissue Hg after the 2002-2003 drought in South Carolina.

Lake Murray is a 50,000-acre reservoir in central South Carolina that was constructed in the late 1920s and early 1930s as a hydroelectric and flood control project (SCE&G, 2010a). During the first decade of the 21st century, two events occurred that resulted in seasonal water manipulation that deviated from the traditional operation of the dam. Normal operation of the project has traditionally resulted in a drawdown in the winter with a minimum level at 350 feet and refill in spring with a maximum level of 358 feet generally achieved in May (SCE&G, 2010b). Between 2003 and 2005 water levels were lowered to allow for the construction of a backup dam. In 2006 water levels were lowered again to allow for repairs of the primary dam, but drought in the Saluda watershed prevented lake levels from returning to their normal summer levels. In 2003 the sterile grass carp (Ctenopharyngodon idella Valenciennes) was introduced to Lake Murray to control invasive aquatic plants such as hydrilla [Hydrilla verticillata (LF) Royl]. By 2005 hydrilla’s coverage of the lake was significantly reduced (SCDNR, 2010a). Also in 2003 precipitation in South Carolina increased significantly after several years of severe drought (SCDNR, 2010b). The SCDHEC collects fish tissue data across South Carolina including 2 sites on

Figure 1. Fish tissue and water quality sampling locations on Lake Murray, SC.
Lake Murray. In addition multiple surface water quality samples are taken routinely as part of the SCDHEC statewide monitoring network. The combination of these datasets during the time period of dam construction, drought, and grass carp introduction in Lake Murray along with water elevation data provided us a unique opportunity to examine the effects of these multiple factors on fish tissue Hg in a large southeastern USA reservoir.

MATERIALS AND METHODS

Like most other large human-made reservoirs, Lake Murray is not a lake but rather a river. For the purpose of this paper we distinguish the Saluda River arm of Lake Murray from the lotic portion by standards set forth by the National Hydrography Dataset (USEPA, 2010). Fish were collected from two distinct locations on Lake Murray (Figure 1). Site S-223 is situated at the upper reaches of the reservoir where SC Highway 391 crosses on the Saluda River arm. Here the reservoir is narrowed and shallow relative to the location near the dam. The banks are gently sloping in most of the upper portions of the reservoir with small coves and pockets holding the majority of the established vegetation and habitat. Largemouth bass (Micropterus salmoides Lacepede) were collected from several of these areas in close proximity. Site S-273 is located near the dam on the lower end of Lake Murray. In contrast to S-223, this latter site is on the pool end of the reservoir. Large coves and long sloping points characterize the lower lake pool, all with deep water relatively close to serve as a deep-water refuge during the hot summer months. The predominant habitat consists of human-made boat docks and piers and some established vegetation in the backs of coves and points. The largemouth bass were collected from docks and piers during the referenced time period.

Field and laboratory methods for collecting and processing fish tissue for Hg are given in Glover et al. (2010). Briefly, freshwater fish were collected using a Smith Root Electroshocking boat. For each sampling station 5 largemouth bass were collected. Specimens were transported to the laboratory and individual specimens were processed with a skin-on fillet taken from the right side of each fish. A Hg reading was obtained for each fish specimen. For consumption advisories SCDHEC follows the weight of evidence approach described by Anderson et al. (1993), using a three-year moving arithmetic mean Hg value from each fish species to estimate the central tendency for a specific water body. This Hg value is evaluated against criteria given in Glover et al. (2010) for the issuance of No Restrictions, One Meal Per Week, One Meal Per Month, or Do Not Eat Any consumption advisories.

Water chemistry data were obtained from both S-223 and S-273 by the SCDHEC ambient water quality monitoring program (SCDHEC, 2008). Lake elevation data were obtained from the United States Geological Survey’s National Water Information System (USGS, 2010).

Data analyses were performed with SAS Institutes SAS/STAT Software v.9.1.3 (SAS, 2002). For this analysis we used largemouth bass collected between 2002 and 2008 to examine spatial and temporal trends. To account for the relatively large number of values below the laboratory analytical detection limits (24 out of 70 specimens or 34%), we used censored regression statistical techniques to estimate means. Statistical techniques to account for censored data have been available since the Tobit Model was developed by economist James Tobin (1958). While the use of censored regression techniques are common in disciplines such as medical research and economics a common method in environmental science has been to assign an arbitrary number, such as one half the detection limit, to account for unknown values (Helsel, 2005). Data below analytical detection limits are often treated as left censored data, but Helsel (2005) recommended using interval censoring techniques, which we refer here to as Interval Censored Regression (ICR). This is because conceptually a contaminant reading cannot be less than zero, as left censoring will allow, but with ICR the interval is set between zero and the detection limit.

We constructed two ICR models to calculate trends in largemouth bass tissue Hg levels. Model one was used to estimate the overall mean of largemouth bass tissue Hg levels normalized to a length of 37.1 cm (the mean length of all largemouth bass in the dataset) from Lake Murray. Model two evaluated tissue Hg levels by station, fish length, and year, to allow for non-linear time effects, and station-year interactive effects.

Stepwise multiple linear regression was used to evaluate the relationship of length normalized tissue Hg levels (from ICR Model 2) and mean monthly lake elevations at each station for a given year and the mean monthly December-minus-May lake level reading for each year. During the historic operation of the dam, the December lake elevations were lower than the May lake levels. By subtracting the May lake elevations from the December lake elevations we were able to evaluated the potential effects on tissue Hg of the anomalous years of 2003, 2004, and 2006. The Wilcoxon rank-sum test was used to test for differences in water chemistry readings between the two sampling locations.
RESULTS AND DISCUSSION

Figure 2 shows mean monthly lake elevations from 2002 through 2008. This figure illustrates that in 2004 and 2006, the December lake levels were higher than in May of the same calendar year. In 2003 December and May levels were similarly low because of the secondary dam construction. This deviated from the historic operation.

Many studies have shown tissue Hg levels to be correlated with size or age of most fish species (Munthe et al., 2007) and length here was positively correlated for both models (Model 1 Chi Square = 42.88 p<0.0001; Model 2 Chi Square= 34.63 p<0.0001). The overall mean predicted Hg value for Lake Murray largemouth bass normalized to a length of 37.1 cm was 0.22 ppm. This relatively low value is consistent with findings by Glover et al. (2010) and Rypel et al. (2008) who found that southeastern US large reservoirs had fish with low levels of tissue Hg relative to flowing, unregulated rivers.

Nevertheless, there are interesting temporal trends in tissue Hg exhibited at both sites. The results of the ICR model 2, which accounted for length, station, year, and station-year interactions, are shown in Figure 3. As shown, a marked increase in tissue Hg occurred in year 2004 at site S-223, while it declined at S-273 near the dam. In the years that followed, fish tissue Hg levels were gradually observed to increase at S-273 but decrease at S-223.

Figure 2. Mean monthly lake elevations for Lake Murray, South Carolina.
Results of the stepwise multiple regression analysis indicated that the mean lake elevations were not statistically significantly related to Hg levels in fish for either station. However, the difference in the December and May lake levels in the same calendar year revealed a statistically significant relationship with tissue Hg at S-273 (R square = 0.78, p<0.05) and nearly so at S-223 (R square = 0.56, p=0.05). The parameter estimates (negative for S-273 and positive for S-223), along with Figures 3 and 4, show that an apparent response of tissue Hg to changing lake levels is opposite for the station nearest the dam and the station in the upper end of Lake Murray. The response at S-273 was similar to that reported by Glover et al. (2010) for statewide trends. In their study the cause of the drying and rewetting of soil occurred because of a statewide drought followed by a year of heavy rains. For Lake Murray the cause of the drying and rewetting was human water manipulation but the physical and chemical processes resulting in changes in tissue Hg are likely the same as for natural hydrological variability. The response at S-223 however was in the opposite direction than what would have been predicted from previous studies.

Munthe et al. (2007) reviewed the importance of spatial, geochemical, and biological controls of Hg methylation. Table 1 shows the medians along with the results of a Wilcoxon rank-sum test for various water chemistry variables from each station. Iron, Alkalinity, Turbidity, and various nitrogen measurements were higher at S-223 than at S-273 and were statistically significantly different. Glover et al. (2010) found a similar pattern when comparing regulated rivers and reservoirs. Site S-223, while still considered part of Lake Murray, is more riverine than S-273 so these patterns were not unexpected. While a variety of studies have shown that the above and other water chemical parameters can be correlated with fish tissue Hg levels, how they contributed to the patterns seen in Lake Murray fish is unclear. For example, iron has been shown to be positively correlated with Hg in fish tissue (Munthe et al., 2007) and was much higher at S-223 than S-273 (Table 1). However, as indicated from ICR Model 2 the length normalized tissue Hg levels were not significantly different at these two stations (Chi Square = 0.23 p = 0.63) but, the stations-year interactive effects were statistically significant (Chi Square = 55.78 p < 0.0001), demonstrating a difference in the temporal but not the

Figure 3. Length normalized mercury levels for largemouth bass calculated using interval censored regression from two locations on Lake Murray, SC.

Figure 4. Regression analysis showing the relationship of length normalized tissue mercury levels and December minus May mean monthly lake elevations for Lake Murray, SC.

| Table 1. Median surface water chemistry parameters from Lake Murray. Test statistic Wilcoxon rank-sum test. |
|---------------------------------|------------|------------|------------------|
| Variable                        | S-223      | S-273      | p value          |
| Alkalinity (mg/l)               | 24.0       | 21.0       | <0.0001          |
| Total Organic Carbon (mg/l)     | 3.20       | 3.25       | 0.79             |
| Dissolved Oxygen (mg/l)         | 8.9        | 8.5        | 0.35             |
| Iron (ug/l)                     | 415        | 51         | <0.0001          |
| Ammonia (mg/l)                  | 0.07       | 0*         | NA*              |
| Total Kjeldahl Nitrogen (mg/l)  | 0.37       | 0.24       | <0.0001          |
| Nitrate + Nitrite (mg/l)        | 0.20       | 0*         | NA*              |
| pH                              | 7.40       | 7.6        | 0.25             |
| Phosphorous (mg/l)              | 0.04       | 0*         | NA*              |
| Turbidity                       | 8.85       | 1.25       | <0.0001          |

* NA=not applicable. Most values below detection limit.
spatial magnitude of tissue Hg levels. Whether iron or any other chemical parameter was influential in the diverging patterns in tissue Hg at these two sites is unknown.

Likely it is the combination of chemical, physical and biological differences in S-223 and S-273 that played a role in the diverging response of fish tissue Hg to the multiple events that occurred over the study period. Evers et al. (2007) noted that fish tissue Hg from reservoirs with steep sided organic-poor substrates are less likely to be effected by water level manipulation than reservoirs with shallower, gently sloping banks. This is because the littoral zone of the latter has a large surface area allowing for an increased methylation efficiency. The dewatering and inundation that occurs in this littoral zone because of water level manipulation results in a transitioning reduction-oxidation cycle, which favors the sulfur reducing bacteria that is important in the methylation process. Several studies demonstrate that the drying and rewetting of soils, either by natural causes or water manipulation in reservoirs, contributes to a flux in Hg methylation (Munthe et al., 2007; Evers et al., 2007).

Site S-223 is more analogous to a shallow and narrow reservoir with gently sloping banks, while S-273 is more analogous to a large open-water reservoir with abundant deep-water habitat. The greatest response in tissue Hg levels at both sites was in 2004 (Figure 3). These specimens were collected in April of that year after the prolonged drawdown for dam construction (Figure 2). Without the annual drawdown and refill that generally occurs under historic reservoir management, it would be reasonable to predict that fish tissue Hg would decrease, which it did at site S-273. Fish tissue Hg at S-273 increased beginning in 2005 after reservoir refill. This pattern is similar to that found statewide in South Carolina at the end of a prolonged drought (Glover et al., 2010). However in 2004 fish tissue Hg levels were higher at S-223 than previous years and decreased sharply in 2005 (Figure 3). Because of its relatively smaller channel size, this location likely retains more connectivity with the surrounding landscape. Thus the increased precipitation in 2003 after several years of drought may have played a more important role in bioaccumulation here than at S-273, where the much larger volume of water could have mitigated the influence of MeHg load from tributaries.

Another possibility is that the extended drawdown between 2002 and 2004 could have resulted in a trophic shift or other behavioral changes in largemouth bass. Largemouth bass have been shown to change predation tactics in response to habitat changes such as vegetation reduction (Savino and Stein, 1982). Sammons et al. (2003) found that largemouth bass exhibited an increase in movement and moved more during the day following hydrilla removal in an arm of a Georgia reservoir. They speculated that this behavior may have been a result in a shift from the more littoral zone prey base such as bluegill (Lepomis macrochirus Rafinesque) to more pelagic prey such as gizzard shad (Dorosoma cepedianum Lesueur) and threadfin shad (D. petenense Cook). Colle et al. (1989) found that when immersed aquatic vegetation is removed from a reservoir, inshore largemouth bass prefer emergent weedy areas, while fish that stayed off shore preferred human piers (such as those retrieved from S-273). The extended drawdown between 2002 and 2004 effectively eliminated the traditional littoral habitat frequented by largemouth bass in the growing season of 2003, which could have contributed to the higher levels of tissue Hg in 2004 collected at S-223.

Yet another potential factor was the introduction of 64,500 sterile grass carp in 2003 to help control the invasive aquatic plant hydrilla (SCDNR, 2010a). Since late 2005 no appreciable hydrilla was found in lake surveys. It is unclear if the same largemouth bass behavioral changes reported by Sammons et al. (2003) after hydrilla removal has occurred in Lake Murray fish. However it could account for observed patterns of tissue Hg from fishes in this reservoir.

CONCLUSION

That a trophic shift could lead to a change in bioaccumulation of Hg in fish was demonstrated by Kelly et al. (2006). In their work they found that a forest fire increased nutrient levels in a Canadian lake, leading to increased productivity, and causing a shift to increased picivorey by some fish species. They suggested that this cascade of events accounted for the majority of the observed increase Hg levels in fishes post fire. In the Florida everglades a similar increase in fish tissue Hg occurred post fire (Krabbenhoff and Fink, 2001). However the Florida team demonstrated that the most plausible explanation for this increase was the oxidation of sulfur species into sulfate (Gilmour et al., 2004). After rewetting, remobilized soil Hg along with increased sulfate caused a flux of MeHg production.

Our results highlight the complexity of the Hg cycle, and suggest that external events can shape the magnitude and direction of bioaccumulation, even within a single waterbody. Thus physical, chemical, and biological conditions must be considered when attempting to analyze spatial and temporal trends. In Lake Murray it is likely that several factors were responsible for temporal patterns observed in fish tissue Hg at two different
locations. However, the magnitude and direction of the response varied by the location in the lake where fish reside. So while the relative contribution of the drying and rewetting of soil could have been the predominant contributing factor for patterns seen in fish tissue Hg at S-273 it could have been relatively less important at S-223. Here a trophic shift, either because of water level manipulation or aquatic vegetation reduction, could have been the primary drivers. The increased precipitation in 2003 after an extended drought could also have been relatively more important at S-223 than S-273. All of these explanations are plausible and based on concepts derived by researchers over the past 30 years. However how these various factors interacted to contribute to the patterns seen in fish tissue Hg is complex and unclear at this time.

There is mounting evidence that reductions in atmospheric Hg in the USA over the past several decades have lead to a decrease in tissue Hg levels in fish (Glover et al., 2010; Chalmers et al., 2010; Lang 2006; Vokoun and Perkins, 2008). While it may seem intuitive to attribute all spatial and temporal trends in fish tissue Hg to atmospheric deposition, our results suggest that this should be done with caution. Examination of trends at individual locations and over relatively short periods of time can, in our opinion, lead to spurious conclusions if drivers in methylation and bioaccumulation are not carefully considered.

In most instances the collection of large amounts of data at multiple locations is required to fully understand a particular system. This view supports the findings of Paller et al. (2004) on their work on bioaccumulation factors (BAFs) for the Savannah River in South Carolina. A BAF is the ratio of Hg in fish flesh to the Hg in the water (USEPA, 2009) and, from a regulatory perspective, can shape the outcomes of Total Maximum Daily Loads (TMDLs) and predictive models that attempt to forecast effects on fish tissue from point source aerial loads. Thus a representative BAF for specific water bodies is critical for predicting outcomes of reductions or increases of Hg loads to a system. The USEPA (2009) reported default BAFs for various trophic levels of fish in the US but recommend that site specific BAFs be developed for individual waterbodies of interest. Our study adds support to this recommendation but, along with a growing body of literature, suggests that in the development of site specific BAFs large amounts of data must be collected, over space and time, to maximize precision.

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LITERATURE CITED


