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Lead Contamination and Mobility in Surface Water at Trap and Skeet Ranges

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Large quantities of spent lead shot are deposited at trap and skeet ranges, where shot densities of 10^8 pellets ha^{-1} and total Pb concentrations exceeding 10^4 $\mu g\ g^{-1}$ have been reported in surface soils and sediments (ERCO 1986; Ma 1989; Roscoe et al. 1989).

The ingestion of spent shot from trap and skeet shooting has caused lead poisoning in cattle that consumed shot-contaminated silage (Howard and Braum 1980; Frape and Pringle 1984; Rice et al. 1987) and in waterfowl that fed in a shot-contaminated tidal meadow (Roscoe et al. 1989). Ma (1989) reported strongly elevated Pb concentrations in the kidney, liver and bone tissue of small mammals captured at an abandoned shooting range in an area with sandy, acidic soils. Shrews (Sorex araneus) and bank voles (Clethrionomys glareolus) from the site had higher kidney-to-body-weight ratios than controls, a possible indication of lead poisoning.

Relatively little is known about the potential impacts of lead shot deposition in aquatic environments. Elevated lead concentrations have been reported in ribbed mussels (Modiolus demissus) in an estuary contaminated with lead shot from a shooting range (ERCO 1986). Lead was also elevated in the interstitial water of the shot-contaminated sediments, but not in the overlying water column.

In this paper we report the results of a survey of lead contamination at shooting ranges where shot has been deposited in open water or wetland areas. We then looked for lead contamination in a lake adjacent to the range that had the greatest amount of shot deposition.

MATERIALS AND METHODS

Eight shooting ranges were selected for sampling based on the presence of surface water in the shot fall zone. We define the shot fall zone as the area

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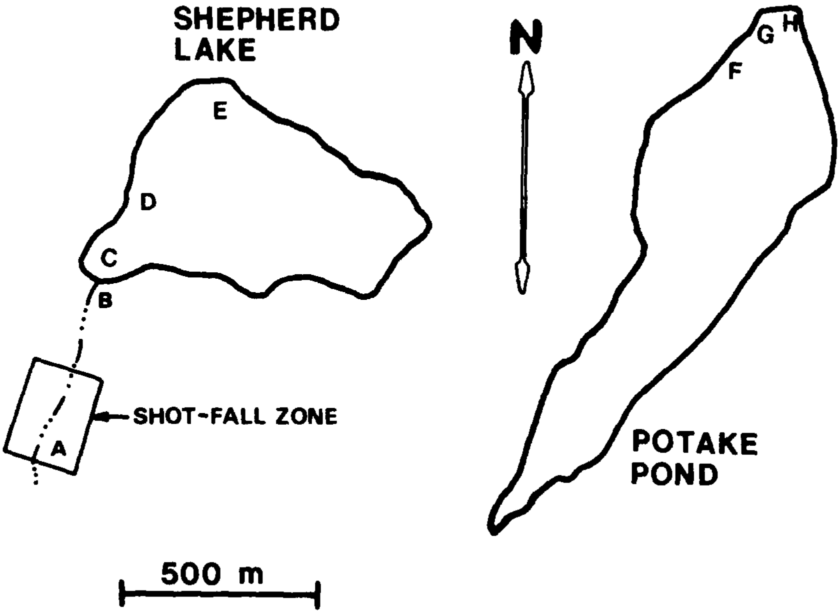

Figure 1. Water sampling locations at the Thunder Mountain Trap and Skeet Range, Shepherd Lake and Potake Pond (control site).

directly in front of the shooting positions, from the first visible concentration of broken clay birds out to a distance of 200 m. The number of shot in the upper 7.5 cm of soils or sediments in the water-covered areas in the shot fall zone was determined at each range. Twenty five core samples were collected by the method of Quist and Kirby (1978), the pellets were counted and the number of shot ha^{-1} was calculated by extrapolation from the surface area of the core samples.

At each range, we collected three water samples from the shot fall zone and a nearby control area for total lead analysis. Various types of surface water were sampled, including marshes, ponds, streams and seasonal pools. The samples were collected in acid-washed polyethylene bottles, acidified with nitric acid and analyzed by flameless atomic absorption spectrophotometry using the method of standard additions. The method detection limit was 1 $\mu g\ L^{-1}$. Total alkalinity (USEPA 1979) and pH (Orion SA 230 pH meter) were determined at a single location in the shot fall zone and the control area.

We conducted additional sampling at the range with the highest shot density to look for movement of lead off-site to a nearby recreational and water supply lake (Shepherd Lake) via a small (width $\cong$ 1m) intermittent stream that flows through the shot fall zone (Figure 1). Water samples were collected from the shot fall zone, Shepherd Lake, the tributary stream and a nearby control lake (Potake Pond) that does not receive runoff from the range. In addition to the parameters previously mentioned, water samples were filtered through 0.45 μm membrane filters prior to acidification for determination of filterable lead. Three sediment samples were collected from each lake, dried at 103°C, digested in nitric acid (USEPA 1979) and analyzed for lead by graphite furnace AA. Sediment samples from Shepherd Lake were collected near the stream inlet, while those from Potake Pond were collected in the same locations as were the water samples (Figure 1).

Largemouth bass (Micropterus salmoides) and pumpkinseed sunfish (Lepomis gibbosus) were collected from both lakes by electrofishing and stored frozen prior to lead analysis. The fish were thawed, total length was determined and samples of muscle and liver tissue were obtained using stainless steel implements. The tissue samples were dried overnight at 103°C, charred at 250°C and dry ashed at 450°C. The residue was dissolved in nitric acid, diluted to volume with distilled-deionized water and analyzed by graphite furnace AA.

Field blanks, duplicate samples and EPA QA/QC samples (U. S. Environmental Protection Agency, Cincinnati, Ohio) were analyzed along with the water samples and EPA Trace Metals in Fish QA/QC samples were processed and analyzed along with the fish tissues. Analysis of variance tests were performed on log transformed lead data using the GLM procedure in Statistical Analysis System Software for personal computers (SAS 1987).

RESULTS AND DISCUSSION

Shot densities in the upper 7.5 cm of soil and sediments in the water-covered areas in the shot fall zones ranged from 4.15×10^6 to 3.70×10^9 pellets ha^{-1} (Table 1). Total lead concentrations in water samples collected within the shot fall zones were elevated relative to those in nearby control areas. Lead results are not reported for two of the sites because of salinity-related matrix interferences encountered using the graphite furnace AA method. Mean total lead concentrations were not correlated with H^+ concentration ($P > 0.10$), alkalinity ($P > 0.10$) or shot density ($0.05 < P < 0.10$). The influence of these variables on lead concentrations may have been masked by differences in the total volume and/or flow rate of surface water at the various ranges, as well as the spatial variability in shot density and total lead concentrations in water in the shot fall zones.

Follow-up sampling at the Thunder Mountain Trap and Skeet Range confirmed our initial finding of high total lead concentrations in a marshy area in the shot fall zone (Table 2). Approximately 94% of the waterborne lead was present in the non-filterable fraction. Jorgensen and Willems (1987) reported that lead pellets deposited in the soil become encrusted with $Pb_3(CO_3)_2(OH)_2$ and $PbCO_3$, which are among the least soluble lead compounds commonly found in oxidizing environments (Hem and Durum 1973). Our findings of large

Table 1. Shot density (pellets ha^{-1}) in the upper 7.5 cm of soil/sediment in the shot fall zones, and pH, total alkalinity (mg L^{-1} as $CaCO_3$) and total lead concentration ($\mu g\ L^{-1}$) in surface water in the shot fall zones and nearby control areas.

Site	pH	Total Alk.	Total Pb x (range); n=3	Shot Density
Thunder Mt.	6.5	6.8	581 (127 - 838)	3.70×10^9
Control	6.4	NR[a]	2.9 (2.2 - 3.2)	
Fairview	9.0	47.4	118 (16.1 - 273)	1.00×10^9
Control	7.2	65.7	4.2 (3.3 - 5.5)	
Chester	6.7	21.9	78.8 (1.4 - 168)	1.49×10^8
Control	6.8	NR	1.1 (<1 - 1.8)	
Troy Meadows	9.7	73.1	56.3 (41.9 - 69.7)	2.80×10^9
Control	8.3	105	4.5 (4.0 - 5.5)	
Union County	7.4	98.9	14.1 (8.9 - 23.0)	5.08×10^8
Control	7.5	91.6	5.5 (3.9 - 6.8)	
Etna	6.8	82.5	11.9 (7.4 - 18.3)	2.17×10^8
Control	6.9	42.5	6.3 (5.6 - 7.4)	
Delaware Bay (Abandoned)	7.3	NR	ND[b]	4.15×10^6
Atlantic City (Abandoned)	7.2	NR	ND	1.32×10^6

[a] Not recorded
[b] Not determined due to salinity-related matrix interferences.

amounts of non-filterable lead could be explained by the suspension of this crust material in the water column. Lead concentrations downstream from the shot fall zone were similar to those measured previously in the control area. Non-filterable lead appears to settle out of the water column within a relatively short distance from the shot fall zone. In our earlier sampling, total lead concentrations in the stream declined from 838 $\mu g\ L^{-1}$ midway through the shot fall zone to 127 $\mu g\ L^{-1}$ near the downstream edge of the zone.

Table 2. Total alkalinity (mg L^{-1} as $CaCO_3$), pH and lead concentrations ($\mu g\ L^{-1}$) in surface water at the Thunder Mountain Trap and Skeet Range, Shepherd Lake and Potake Pond. See Figure 1 for sampling locations.

Location	pH	Total Alkalinity	Lead Concentration: Total	Lead Concentration: Filterable
A - Shot Fall Zone (Marsh)	6.3	20.5	1,270	83
B - Stream	7.0	33.9	1.3	2.1[a]
Shepherd Lake				
C	8.4	17.0	<1.0	<1.0
D	8.2	20.2	14.6	<1.0
E	7.4	19.1	<1.0	<1.0
Potake Pond				
F	7.0	7.9	<1.0	<1.0
G	7.0	8.2	<1.0	<1.0
H	7.0	8.0	<1.0	<1.0

[a] Probable contamination: 2.0 $\mu g\ L^{-1}$ lead was detected in a filterable lead field blank collected the same day.

The filterable lead concentration of 83 $\mu g\ L^{-1}$ in the marsh is consistent with calculations performed by Hem and Durum (1973) indicating that dissolved lead concentrations could approach or exceed 100 $\mu g\ L^{-1}$ in low alkalinity water near pH 6.5. The lead concentration in the stream, however, was much lower than in the marsh. At the higher pH in the stream, increased adsorption of dissolved lead to sediments and settleable particulates and precipitation of lead salts may occur (Hem and Durum 1973; Drever 1988).

Lead was below the detection limit in two of the water samples from Shepherd Lake, but was elevated in one sample collected near a public swimming area adjacent to a parking lot. The higher lead concentration at this site may have been due to the re-suspension of sediments that were enriched with lead from parking lot runoff. Filterable lead was below the detection limit in all of the water samples.

Significant amounts of lead could be transported to Shepherd Lake during brief

Table 3. Lead concentrations ($\mu g\ g^{-1}$ dry weight) in fish tissues and sediments from Shepherd Lake and Potake Pond.

	Lead Concentration Mean (SD); n=3	
Sample	Shepherd Lake	Potake Pond
Pumpkinseed Sunfish[a]		
Liver	1.74 (0.64)	1.13 (0.32)
Muscle	0.07 (0.01)	0.18 (0.05)[c]
Largemouth Bass[b]		
Liver	0.07 (0.01)	0.44 (0.13)[c]
Muscle	0.06 (0.01)	0.05 (0.01)
Sediments	6.50 (2.18)	68.0 (36.5)[c]

[a] Total Lengths: Shepherd Lake 151 - 171 mm
Potake Pond 117 - 142 mm

[b] Total Lengths: Shepherd Lake 332 - 370 mm
Potake Pond 165 - 209 mm

[c] Significantly higher ($P < 0.05$) than concentrations in Shepherd Lake

high flow periods, which were not sampled in this study. However, any lead input to the lake should be reflected in elevated concentrations in sediments and biota, the ultimate lead sinks in the system.

We found no evidence of enhanced uptake of lead by fish in Shepherd Lake, although the power of the statistical comparisons is limited by the small sample sizes (Table 3). Liver lead concentrations in pumpkinseed sunfish from both lakes were similar. Liver lead concentrations in largemouth bass from Potake Pond were higher than concentrations in bass from Shepherd Lake ($P < 0.05$), probably due, at least in part, to differences in body size. Largemouth bass collected from Shepherd Lake were larger than those from Potake Pond, and a decrease in liver lead concentration with increasing length has been reported elsewhere (Pagenkopf and Neuman 1974; Bollingberg and Johanssen 1979). Muscle lead concentrations in largemouth bass were similar in fish from both lakes, but concentrations in pumpkinseed sunfish from Potake Pond were higher than concentrations in pumpkinseed sunfish from Shepherd Lake. In all cases, muscle lead concentrations were well below the limit of 0.3 $\mu g\ g^{-1}$ (fresh weight) recommended by the World Health Organization for human consumption (WHO 1972).

We also found no evidence of excess lead accumulation in Shepherd Lake sediments. In fact, sediment concentrations were higher in Potake Pond. However, these results should be interpreted cautiously because no measurements of particle size or organic content were performed. Both parameters have a strong influence on sediment lead concentrations (Jenne and Zachara 1987), and our results may reflect differences in the adsoption capacity of the sediments. Potake Pond is located in an undeveloped, forested watershed with no obvious sources of lead contamination.

Total lead concentrations were elevated in surface water in the shot fall zones at six trap and skeet ranges, but we found no lead contamination in water, sediments or fish from a lake adjacent to the range with the greatest amount of shot deposition. Our findings suggest that there is little off-site transport of lead lead via surface water at neutral to alkaline pH. However, we found a significant concentration of filterable lead in a slightly acidic (pH 6.3) marsh located in the shot fall zone at one range, which suggests that Pb could be mobilized at lower pH.

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