

Anthropogenic Sources of Arsenic and Copper to Sediments in a Suburban Lake, Northern Virginia

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Mass balances of total arsenic and copper for a suburban lake in densely populated northern Virginia were calculated using data collected during 1998. Mass-balance terms were precipitation; stream inflow, including road runoff; stream outflow; and contributions from leaching of pressure-treated lumber. More mass of arsenic and copper was input to the lake than was output; the 1998 lake-retention rates were 70% for arsenic and 20% for copper. The arsenic mass balance compared well with a calculated annual mass accumulation in the top 1 cm of the lake sediments; however, the calculated contribution of copper to the lake was insufficient to account for the amount of copper in this zone. Leaching experiments were conducted on lumber treated with chromated copper arsenate (CCA) to quantify approximate amounts of arsenic and copper contributed by this source. Sources to lake sediments included leaching of CCA-treated lumber (arsenic, 50%; copper, 4%), streamwater (arsenic, 50%; copper, 90%), and atmospheric deposition (arsenic, 1%; copper, 3%). Results of this study suggest that CCA-treated lumber and road runoff could be significant nonpoint sources of arsenic and copper, respectively, in suburban catchments.

Introduction

Arsenic (As) and copper (Cu) are priority pollutants (1). Arsenic is a human carcinogen (2), and Cu is an essential element to biota and humans but is toxic in excess (3–5). Because of potential health effects to biota and humans, the presence of these elements in terrestrial and aquatic environments is of concern. For example, accumulation of As and Cu from industrial sources in lake and reservoir sediments has been observed (6–8). Anthropogenic impacts to industrial environments have received more study than suburban environments because point sources of contaminants are easier to quantify than are nonpoint sources. To explore the potential of similar impacts in a suburban environment (i.e., an enrichment of As and Cu in lake sediments over background concentrations), we developed a mass-balance model for a suburban catchment. A mass balance quantifies the natural as well as anthropogenic inputs and outputs of a given constituent for a catchment. If the

catchment contains a lake, input of x to the lake – export of x from the lake = contribution of x to the lake sediments, where storage of x in lake water remains constant.

Potential sources of As and Cu to a suburban catchment include atmospheric deposition, geologic contribution, roadways, and pressure-treated lumber. Combustion of As- and Cu-containing coal and weathering of As- and Cu-containing bedrock provide a source of these elements in atmospheric deposition and streamwater, respectively. Copper can be deposited on roadways by motor-vehicle brake-component wear (9, 10); combustion of gasoline, diesel, and lubricating oils (11); and tire wear (10). Rainfall can wash the Cu from impervious surfaces directly into streams and lakes. Chromated copper arsenate (CCA) is a wood preservative used in pressure-treated lumber, with insecticidal properties provided by As and fungicidal properties provided by Cu (12). Typically, concentrations of As and Cu in the wood range from 1000 to 5000 mg/kg (13). Decks constructed of CCA-treated wood release As and Cu to the underlying soils: up to 350 mg/kg As and 410 mg/kg Cu accumulated in soils beneath decks, as compared to an average of 17 mg/kg As and 4 mg/kg Cu in control soils (13). Contact of lake water or rainwater with CCA-treated wood potentially can release As and Cu to the environment. Although leaching of As and Cu from bulkheads and dock pilings and accumulation in adjacent sediments have been observed in estuarine environments (e.g., refs 14–16), no previous studies are known to have documented trace-element leaching from CCA-treated lumber in a freshwater environment. Environmental managers are becoming aware of the potential of release of As from CCA-treated wood as indicated by the U.S. EPA's announcement of the phase-out of As in treated wood for residential use by December 31, 2003 (17). This study provides an example for environmental managers of the effect that CCA-treated wood can have on the environment.

Study Site. Our case study was of Lake Anne, a recreational, 10.9-ha lake created in 1964 in suburban northern Virginia (Figure 1). The 235-ha catchment is densely populated (1116 people/km²) and heavily developed, with dwellings surrounding the lake shoreline. Two tributaries contribute to the lake, but one drains another dammed lake within the catchment (Figure 1). Although excessive algal growth is common in Lake Anne, it does not serve as a drinking-water supply and has not been treated with CuSO₄·5H₂O. CCA-treated wood is used in the construction of decks and docks in and near Lake Anne and as a bank stabilization material along approximately 75% of the shoreline.

Components of the mass balance of total As and Cu for Lake Anne for 1998 were atmospheric deposition; streamwater, which included runoff over impervious surfaces and constituents derived from weathering of the schist, gneiss, and phyllite bedrock (18); and inputs to the lake through leaching of CCA-treated lumber by both rainwater and lake water. A laboratory leaching experiment was conducted on CCA-treated samples to quantify the approximate mass of As and Cu delivered to the lake from leaching CCA-treated lumber. For the water balance, the contribution of groundwater inflow to the lake and discharge from the lake was assumed to be equal, and the annual change in storage of lake water was assumed to be negligible.

The calculated fluxes were compared to the annual mass accumulations at the top of three lake-sediment cores that were collected from Lake Anne in 1996 (box and gravity cores) and 1997 (box core). Dry-weight concentrations at the top of the sediment cores ranged from 18 to 28 µg/g As and from 105 to 137 µg/g Cu, which are up to 23 and 5 times higher

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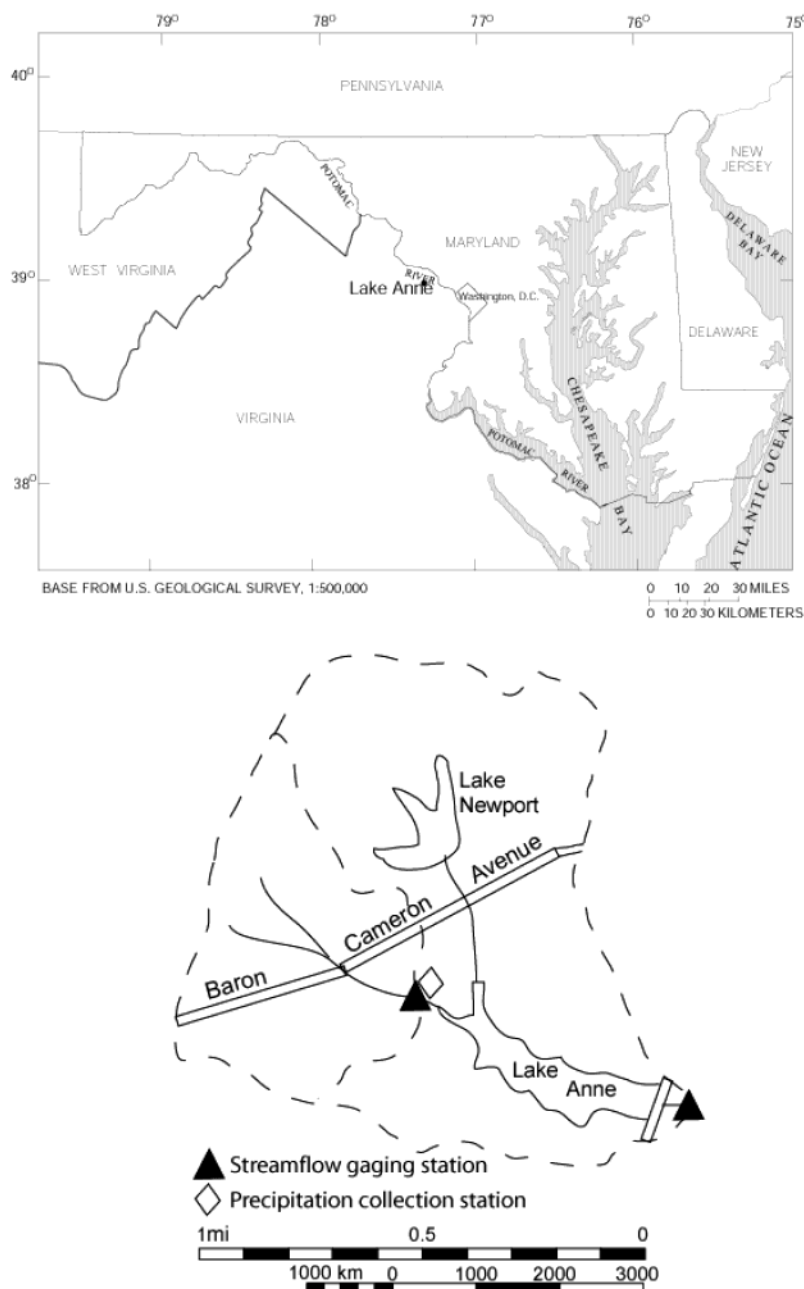


FIGURE 1. Location of study area.

than As and Cu background concentrations, respectively (E. Callender, unpublished data). Inputs measured during 1998 were assumed equivalent to those in 1996 and 1997, as recorded in the top 1 cm of the lake sediment because the average sedimentation rate is approximately 0.6 cm/yr.

Experimental Section

Field. During 1998, 2-week integrated precipitation samples were collected in precleaned, 2-L Teflon-coated wide-mouth low-density polyethylene (LDPE) bottles in the wet side of an Aerochem Metrics model 301 automatic wet-dry precipitation collector, which had been modified for trace-element collection by encasing the lid in plastic and covering the support arms with Teflon tape. Rainfall amounts were recorded with a tipping-bucket rain gage connected to a Campbell Scientific, Inc. CR10X data logger and summed on a daily basis. Discharge data for the major inflow and outflow streams were recorded at 15-min intervals by use of USGS streamflow-gaging stations and techniques (19). Streamwater

samples were collected as grab samples from the deepest and fastest flowing portion of the stream, 5–10 m upstream of each of the gages (Figure 1), over a wide range of discharges, from baseflow to stormflow. Road runoff samples were collected as grab samples of water flowing over impervious surfaces during storms. Lake-water samples were collected as surface grab samples from a rowboat: one at the head of the lake, one approximately at the midpoint of the lake, and a third at the lower end of the lake. Ultraclean techniques were used for all sample collection, including the use of vinyl gloves and precleaned LDPE bottles.

Laboratory. Unfiltered precipitation samples were acidified with Fisher Optima double-distilled HNO_3 to 0.5% v/v, which was sufficient to lower the pH to <2 to prevent adsorption of the elements to the container walls. This treatment is considered a method for analysis of total trace elements in precipitation (20). Precipitation samples were analyzed by inductively coupled plasma-mass spectrometry. Unfiltered road runoff, streamwater, and lake-water samples

were allowed to evaporate at 40 °C from 500 mL to approximately 10 mL, the evaporative residue was digested, and As and Cu concentrations were determined by graphite furnace atomic absorption spectrophotometry (GFAAS). Sediment collected from the top 1-cm sections of the three lake cores was digested and analyzed for As (GFAAS) and Cu (inductively coupled plasma-atomic emission spectrometry; ICP-AES). Quality assurance of all field and laboratory procedures was provided by analysis of several types of blanks, triplicate samples, and certified reference materials (21).

The leaching experiment was designed to give an estimation of the amounts of As and Cu that could be released from the CCA-treated decks and docks surrounding the lake. The experiment simulated a flushing-type environment where the leaching solution could disperse the leached elements into the surrounding water. The leaching solution was unfiltered lake water, collected as a surface grab sample from Lake Anne, with a pH of 6.70 and stored at 4 °C.

Eight samples of different types of lumber and weathering histories were leached with lake water. Two samples were non-CCA-treated wood (yellow pine and cedar) used as controls. A ninth sample was leached in unfiltered rainwater with a pH of 4.70. The samples were cut and rinsed to remove any surface contaminants contributed by shipping, storage, and handling and were dried and weighed. Each sample was placed in precleaned 2-L lidded polyethylene containers, which were filled with 500.0 g of the leaching solution and left undisturbed at approximately 21 °C. After 1 week of leaching, the sample was removed, and the solution was transferred to a LDPE bottle and acidified with Fisher Optima double-distilled HNO₃ to a concentration of 0.5% v/v. The containers and samples were briefly rinsed with deionized water but not redried, and a fresh 500.0-g aliquot of leaching solution was added. This procedure was followed weekly for 14 weeks. Preleach and weekly postleaching solutions were analyzed by ICP-AES. High concentrations and thus accurate quantification were assured by the small volume of water relative to the surface area of the sample. Concentrations were normalized by the mass of each sample to obtain micrograms per gram.

Mass Balance. Volume-weighted wet-deposition fluxes of As and Cu were calculated for 1998. Previous studies in the Chesapeake Bay catchment indicate that dry deposition of As and Cu is at least as important as wet deposition (22). Accordingly, the wet-deposition fluxes were doubled to give an approximation of contributions by wet plus dry (total) deposition. Total atmospheric deposition fluxes of As and Cu were multiplied by the surface area of the lake to determine the mass of each element delivered directly to the lake via atmospheric deposition.

Stream fluxes were calculated as the annual sum of the products of instantaneous discharge and predicted instantaneous concentration, divided by the catchment area, where the catchment area for the inflow stream is 70.3 ha and that of the outflow stream is 235 ha. Predicted instantaneous concentrations were determined by the best-fit relation between concentration and discharge. For inflow to the lake via the ungaged tributary, runoff and concentrations of As and Cu per unit area were assumed equal to those in the gaged inflow stream, and the ungaged inflow was scaled-up proportionally. The streamwater flux was considered a conservative estimate because the gaged catchment was somewhat less developed, with 24% impervious surfaces as compared to the ungaged catchment with 27% impervious surfaces.

Contributions of As and Cu to the lake from leaching of CCA-treated lumber were calculated as the sum of (i) lake water in contact with dock pilings and retaining walls and (ii) rainwater falling on decks and docks, which drips directly

into the lake. Fluxes of As and Cu from leaching of CCA-treated lumber by lake water were calculated by use of the average of the concentrations of six samples at week 14 of the leaching experiment:

$$C \text{ (mg L}^{-1}\text{)} \times V \text{ (L wk}^{-1}\text{)} \div A \text{ (m}^2\text{)} \times S \text{ (m}^2\text{)} \times 52 \text{ wk yr}^{-1} \quad (1)$$

where *C*, the average concentration (2.0 mg/L As; 0.27 mg/L Cu) times *V*, the weekly volume of water used in the experiment (0.50 L), divided by *A*, the surface area of the sample (0.042 m²), is the mass leaching flux (in mg m⁻² wk⁻¹). The total mass flow to the lake was obtained by multiplying by *S*, the surface area of wood in contact with lake water (approximately 900 m²). *S* was determined by field reconnaissance and aerial photography analysis.

Contributions of As and Cu leached from decks and docks on the lake by rainwater also were calculated by use of the concentrations obtained at week 14, the assumed steady-state concentration. Equation 1 was used except that the sum of all the days of rainfall greater than 1.27 mm during 1998, expressed as weeks (11.4 weeks), and 2200 m² of wood were used. The 2200 m² of wood was estimated from the approximate dimensions of docks, overhanging decks, and bridges on Lake Anne determined by field reconnaissance and aerial photography analysis. A rainfall depth of 1.27 mm is considered the minimum amount required to sufficiently wet the wood surface to flush any dried salts and create drip to the lake below, an environment similar to the leaching experiment. The assumed effective storage capacity of rainfall of 1.27 mm is conservative because studies in Europe indicate that the amount of rainfall required to produce road runoff ranges from 0.13 to 1.5 mm (23).

To compare the mass balances of As and Cu to the concentrations measured at the top of the lake cores, an annual mass accumulation of each element was calculated as follows:

$$J \text{ (}\mu\text{g g}^{-1}\text{)} \times \text{MAR (g cm}^{-2}\text{ yr}^{-1}\text{)} \times K \text{ (m}^2\text{)} \quad (2)$$

where *J* is the measured concentration of As or Cu in the upper 1 cm of the lake core, MAR is the mass accumulation rate, and *K* is the surface area of the lake. The MAR for the two 1996 cores is 0.19 and 0.15 g cm⁻² yr⁻¹ and for the 1997 core is 0.20 g cm⁻² yr⁻¹ (24). Equation 2 was applied to the three lake cores collected from Lake Anne; results were compared to the values obtained by the mass balance.

Results

Arsenic concentrations were highest in road runoff, followed by lake water, stream inflow, outflow, and precipitation (Figure 2a). Copper concentrations were highest in road runoff, followed by stream inflow, lake water, stream outflow, and precipitation (Figure 2b).

In the leaching experiment, the control samples had minor release of As and Cu (Table 1). The median As and Cu concentrations leached from both new and weathered CCA-treated samples were at least 5 and up to 71 times greater than the controls. Total concentrations leached were more than 30 (As) and 7 (Cu) times that of the controls (Table 1). Temporal changes were observed during the experiment. Concentrations of As and Cu were greatly elevated above initial solution concentrations after the first week of leaching and dropped off sharply for the next 2–3 weeks. After week 8, concentrations leveled off and approached steady state. At steady state, concentrations of Cu were approximately 0.3 mg/L, and concentrations of As were at least 1.8 mg/L in both solutions.

The mass-balance calculations indicated that 70% of the As and 20% of the Cu input to the lake was retained (Table

TABLE 1. Results of Leaching Experiment

wood type	water type	As (μg/g)				Cu (μg/g)			
		min	median	max	total	min	median	max	total
Controls									
yellow pine	lake	0.18	0.29	0.6	2.8	<0.1	0.09	0.23	0.83
cedar	lake	<0.2	0.24	0.5	2.0	<0.1	0.20	0.47	1.6
New Wood									
fence board	lake	10	15	43	230	1.5	4.4	15	62
fence board	rain	8.7	15	96	270	1.5	5.8	130	230
landscape timber	lake	3.6	14	22	100	0.52	1.9	2.8	14
landscape timber, 10 yr in storage	lake	3.8	12	21	99	1.0	2.6	4.0	21
Weathered Wood									
fence board, 4 months	lake ^a	7.5	17	30	84	1.3	2.6	4.0	13
landscape timber, 3 yr	lake	3.8	11	20	92	0.49	1.3	1.8	11
decking timber, 5 yr	lake	2.9	10	20	85	0.76	2.4	4.5	20

^a All boards leached for 14 weeks, except for the fence board weathered for 4 months, which was leached for 10 weeks.

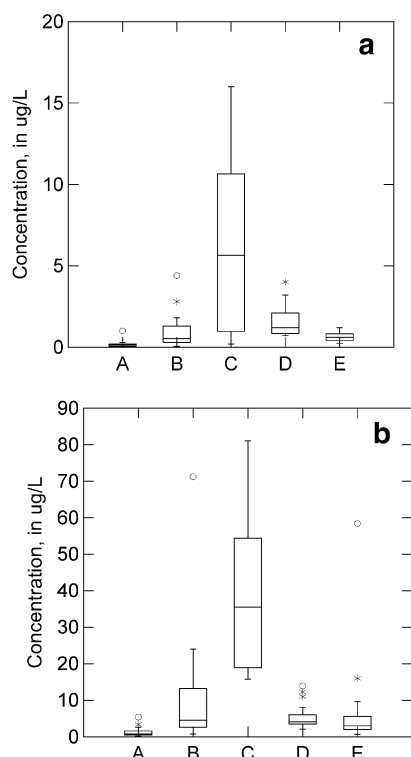


FIGURE 2. Concentrations of (a) arsenic and (b) copper in precipitation, stream inflow, road runoff, lake water, and stream outflow.

TABLE 2. Results of Mass-Balance Calculations of Total Arsenic and Copper Inputs to and Outputs from Lake Anne during 1998

source	As (mg)	Cu (mg)
atmospheric deposition	2.2×10^4	1.5×10^5
stream inflow (173 ha)	1.4×10^6	5.5×10^6
leaching (steady state)	1.6×10^6	2.5×10^5
total input	3.0×10^6	5.9×10^6
stream outflow (235 ha)	9.1×10^5	4.8×10^6
total (input - output) = contribution to lake sediments	2.1×10^6	1.1×10^6
retention in the lake (%)	70	20

2). Results of the calculation of annual mass accumulation (eq 2) at the top of the three lake cores ranged from 4.0×10^6 to 5.2×10^6 for As with a median of 4.6×10^6 mg/yr and from 1.9×10^7 to 3.0×10^7 for Cu with a median of 2.2×10^7 mg/yr. These results indicate that the mass-balance calcu-

tion for As is about 2 times less than the median annual mass accumulation in the sediment, whereas that of Cu is 20 times too low.

Discussion

Precipitation samples had the lowest As and Cu concentrations of all the waters sampled (Figure 2), indicating that rainfall serves to mobilize contaminants internal to the catchment rather than to directly contribute As and Cu to the lake. Low concentrations in baseflow samples collected from the inflow stream indicated that geologic contributions of As and of Cu to the lake via groundwater were negligible. Elevated concentrations in stormflow samples indicated mobilization of these elements from near-surface terrestrial sources (e.g., mobilization of As-laden soil from near decks and of Cu from roadways). The increase in streamflow during storms increased the streamwater fluxes of internally derived As and Cu to the lake. The majority of streamflow during storms is provided by road runoff via stormwater drains. In-lake sorption (25–27) and sedimentation processes caused As and Cu concentrations to be lower in the outflow stream than in the inflow stream or lake water (Figure 2).

Leaching Experiment. Concentrations of As in lake water were greater than in stream inflow (Figure 2a), suggesting an in-lake source of As, such as docks and bank stabilization materials. Warner and Solomon (28) found that leaching from weathered wood is similar to that from new wood with a few instances of higher concentrations leached from weathered wood. Low but continuous concentrations of As, Cu, and Cr released from CCA-treated wood have been observed in an estuarine environment (29). These previous results along with our leaching results suggest that submerged CCA-treated lumber can be a long-term, continuous source of these elements to lakes and other bodies of water in the environment.

The length of time needed to deplete all As and Cu from 1 kg of new wood was determined using the steady-state concentrations obtained in the experiment. It would take approximately 40 yr to deplete all of the As in 1 kg of wood leached in lake water, and it would take approximately 45 yr in rainwater. It would take approximately 180 yr to remove all of the Cu in 1 kg of wood in either lake water or rainwater. These calculations indicate that the steady-state concentrations used in the mass-balance calculations are by no means gross overestimates of depletion of these elements from wood, especially given the fact that docks and decks typically are replaced every 1–2 decades.

Even though rainwater is only intermittently in contact with docks and decks, the initial leaching response by rainwater suggests that it could be a more important

mechanism for leaching than lake water in continual contact with submerged pilings and retaining walls. The pH control of leaching of the elements from the wood is particularly important in the mid-Atlantic and northeastern United States, where acid rain is a well-documented environmental problem. During 1998, the volume-weighted H^+ concentration in precipitation collected in the Lake Anne catchment was 81 μM (pH 4.09) (24), among the most acidic rain in the United States (30).

Mass Balance and Sensitivity Analysis. Results of the mass-balance calculations for 1998 indicated that the majority of the As and Cu contributed to Lake Anne was internally derived in the catchment. Leaching of CCA-treated lumber (using the steady-state concentrations) was at least as important a source of As to the lake as streamwater, and atmospheric deposition was a negligible (<1%) source. Atmospheric deposition also was a negligible (<3%) source of Cu; however, by contrast to As, streamwater was the most important source of Cu, with leaching of docks only a slightly larger source than atmospheric deposition. CCA-treated lumber appears to be a significant source of As to Lake Anne, whereas road runoff appears to be the most significant source of Cu.

Despite careful and complete measurements, the mass balances did not match the concentrations of As and Cu at the top of the lake sediment cores. To explore the discrepancies, a sensitivity analysis was performed for each mass-balance term. Precipitation was the least sensitive term to the analysis because the flux was at least an order of magnitude less than any of the other terms (Table 2). Some researchers, however, contend that dry deposition of these elements may be as much as twice that of wet deposition (e.g., ref 31). To test the sensitivity of atmospheric deposition to the mass balance, wet deposition was multiplied by 3 to account for our inability to accurately measure dry deposition. This had little effect on the comparison of the mass balance to the lake sediments: As was still 2 times too low, whereas Cu improved slightly to 19 times too low.

Next, the streamwater flux was examined. The upper 95% confidence intervals for the concentration–discharge relation for the inflow and outflow streams were calculated. For As, there was little effect, the overall mass balance was still 2 times too low, whereas for Cu, the mass balance improved to 9 times too low. Most road runoff within the catchment flows directly into the two inflow streams (maps on file at Fairfax County Maintenance and Stormwater Drainage Division). Fluxes of As and Cu from direct road runoff were calculated as an upper bound on the stream-input term of the mass balances. The Rational Method was used to estimate flow of runoff from impervious surfaces:

$$Q_{pk} = 0.278CIA$$

where Q_{pk} is the peak flow (in m^3/s), C is the dimensionless runoff coefficient, I is the average rainfall intensity (in mm/h), and A is the area of roads (in km^2). In this exercise, C was taken as unity (impervious surfaces), thus the equation can be used to obtain total runoff for storms. The depth of effective precipitation per day (I) was the amount of precipitation needed to generate runoff and mobilize particulates from roadways. Detention storage was accounted for by subtracting 1.27 mm (23) from the daily rainfall data and replacing negative values with zero. Fluxes of As and Cu to the lake via road runoff were obtained by summing $I \times A \times C_o$ (the median concentration of the road-runoff samples) (5.6 $\mu g/L$ As; 36 $\mu g/L$ Cu) over time:

$$\sum_{i=0}^i I \text{ (mm)} \times A \text{ (km}^2\text{)} \times C_o \text{ (mg L}^{-1}\text{)} \quad (3)$$

The independently calculated fluxes of total As and Cu in road runoff (eq 3) were 2.0×10^6 mg of As and 1.3×10^7 mg of Cu. The As value is similar to the stream-inflow value; however, the Cu flux in road runoff is about twice that of the stream flux (Table 2), suggesting that some road runoff enters the lake directly rather than via streamflow. Road runoff was sampled well after storms had begun, i.e., the highest concentration expected during the first flush was not sampled. For example, the median concentration of Cu (36 $\mu g/L$) sampled in road runoff in the Lake Anne catchment was lower than a previously reported value of 78 $\mu g/L$ (32). The insufficient sampling of road runoff may partially explain the disagreement between the mass balance and the annual accumulation. The road runoff analysis indicates that roadways are a more important source of Cu than of As.

Finally, the leaching term was examined. While lake water contact with wood is constant, rainwater contact with wood is intermittent with wetting and drying cycles. We hypothesize that rainwater drying on wood could create a diffusion gradient whereby the elements within the wood could be drawn through pores in the wood to the surface. The freshly diffused elements could dry on the wood surface by evaporation, leaving a salt crust, which would be flushed again by the next rainfall. This hypothesized process is similar to formation of efflorescent salts, which form as a surface encrustation on rocks or soils from evaporation of water brought to the surface by capillary action or by loss of water of crystallization upon exposure to air. Leaching calculated with the first-flush (at the end of week 1) rainwater concentrations yielded 6.6×10^6 mg of As and 7.8×10^6 mg of Cu, which are 4 and 30 times, respectively, greater than the fluxes calculated with the steady-state leaching values (Table 2). If the first-flush concentration for rainwater is used in the mass-balance calculations, the calculated contribution of As to lake sediments changes from 2 times too low to 0.6 times too high; likewise, for Cu, the difference changes from 20 times to 2.5 times too low. These results suggest that the acidity of rainwater and the frequency of rain events are important controls on the leaching of these elements from CCA-treated lumber and that the actual leaching contribution may be a combination of the steady-state and the first-flush values. The rainwater-leaching mechanism needs additional study.

The reasonable agreement of the As but not the Cu mass balance with the median annual mass accumulation in the sediment suggests that the dominant source of Cu may experience more annual variability than that of As. Because a major source of As was leaching of CCA-treated lumber, there should be little interannual variability in the contribution unless numerous new docks were installed in the lake. By contrast, the dominant source of Cu was road runoff, which is affected by annual rainfall. The years that the lake cores were collected, 1996 and 1997, had more days with greater than 1.27 mm precipitation (97 and 86, respectively) than did 1998 (80). This possible extra delivery of Cu during 1996 and 1997 than during 1998 also may help to explain the difference between the calculated flux and the annual mass accumulation.

Results of this study indicate that more mass of As and Cu was input to Lake Anne than was exported during 1998 and that the majority was internally derived in the catchment from anthropogenic activities rather than from geologic sources. Atmospheric deposition was an insignificant source of both As (2.2×10^4 $mg\ yr^{-1}$) and Cu (1.5×10^5 $mg\ yr^{-1}$) as compared to leaching of CCA-treated lumber or road runoff. The major source of Cu to the lake was road runoff ($\geq 5.5 \times 10^6$ $mg\ yr^{-1}$). For As, leaching of CCA-treated lumber (1.6×10^6 $mg\ yr^{-1}$) was at least as important a contributor as stream inflow (1.4×10^6 $mg\ yr^{-1}$). CCA-treated lumber could be an important nonpoint source of As in other suburban catch-

ments given that there are perhaps tens of thousands of such "real-estate lakes" and several hundred millions of cubic meters of CCA-treated wood in use in public (e.g., parks, playgrounds) and residential settings throughout the United States (American Wood Preservers Institute, written communication, 1999).

Acknowledgments

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