Increased salinization of fresh water in the northeastern United States

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Chloride concentrations are increasing at a rate that threatens the availability of fresh water in the northeastern United States. Increases in roadways and deicer use are now salinizing fresh waters, degrading habitat for aquatic organisms, and impacting large supplies of drinking water for humans throughout the region. We observed chloride concentrations of up to 25% of the concentration of seawater in streams of Maryland, New York, and New Hampshire during winters, and chloride concentrations remaining up to 100 times greater than unimpacted forest streams during summers. Mean annual chloride concentration increased as a function of impervious surface and exceeded tolerance for freshwater life in suburban and urban watersheds. Our analysis shows that if salinity were to continue to increase at its present rate due to changes in impervious surface coverage and current management practices, many surface waters in the northeastern United States would not be potable for human consumption and would become toxic to freshwater life within the next century.

impervious surfaces | land use change

For many years, salinization of fresh water related to agricultural practices has been recognized as an environmental problem in arid and semiarid environments throughout the world (1). Long-term salinization of surface waters associated with increasing coverage by roadways and suburban and urban development has been less considered, although previous research has documented sharp increases in concentrations of sodium and chloride in aquatic systems of the rural northeastern United States over decades due to the use of road salt (2–5). Our analysis shows that baseline salinity is now increasing at a regional scale in the northeastern United States toward thresholds beyond which significant changes in ecological communities and ecosystem functions can be expected.

Salinization refers to an increase in the concentration of total dissolved solids in water and can often be detected by an increase in chloride, an important anion of many salts. In the northeastern United States, chloride derived from salt is commonly associated with runoff from roads at latitudes above ≈39°N, particularly during winter. Concentrations of chloride in soils as low as 30 mg/liter have been found to damage land plants, which typically occur in close proximity to roads (6). Increased chloride concentrations in surface waters, however, can be propagated a substantial distance from roadways, leading to more widespread effects on water quality. Increases in salinity up to 1,000 mg/liter can have lethal and sublethal effects on aquatic plants and invertebrates (7), and chronic concentrations of chloride as low as 250 mg/liter have been recognized as harmful to freshwater life and not potable for human consumption (6, 8). Water with chloride concentrations >250 mg/liter can impart a salty taste and also contain elevated concentrations of sodium and toxic impurities from road salt (9), which are of concern to human health. Road salt is currently not regulated as a primary contaminant to fresh waters of the United States, although a recommended limit exists (8). Regulation of road salt was recently considered by the Canadian government after much controversy (6).

Relatively little is known regarding the relationship between widespread increases in suburban and urban development and long-term changes in baseline salinity across regions of the United States. Impervious surfaces now cover >112,610 km^2 in the United States, an area equivalent to the state of Ohio (10). The amount of impervious surface coverage within the United States is expected to increase sharply with >16,093 km of new roads and 1 million single-family homes being created during the present decade (10). The rate of land-use change may be particularly high in segments of watersheds near surface waters such as streams, rivers, and lakes. As coverage by impervious surfaces increases, aquatic systems can receive increased and pulsed applications of salt, which can accumulate to unsafe levels in ground and surface waters over time (6).

Methods

Rural Sites. We investigated the rate of salinization and increases in the baseline concentration of chloride in inland waters by using long-term data from streams and rivers draining rural watersheds in three locations of the northeastern United States: Baltimore County (Maryland), the Hudson River Valley (New York), and the White Mountains (New Hampshire). Rural sites in these areas have experienced relatively small changes in population growth but contain a low density of roads within their watersheds. The sites in Maryland drain into drinking-water supply reservoirs for Baltimore City and have been monitored over the decades by the municipal government. The sites in the Hudson River Valley have been monitored by the Institute of Ecosystem Studies and the U.S. Geological Survey (2), and the sites in New Hampshire are part of the Hubbard Brook Ecosystem Study (3, 11).

Baltimore Metropolitan Area. Within the Baltimore metropolitan area, we explored long-term changes in chloride concentrations across a broader gradient of land use to determine an empirical relationship between salinization and increasing coverage by impervious surface. The Baltimore metropolitan watersheds drain into the Chesapeake Bay and represent one of the most rapidly developing areas of the northeastern United States. In this region, coverage by impervious surface increased by ≈39% from 1986 to 2000 (12). Streams draining forest, agricultural, suburban, and urban watersheds were sampled as part of the National Science Foundation-supported Baltimore Long Term Ecological Research (LTER) project. Samples were collected weekly from 1998 to 2003 without regard to flow conditions (no

Abbreviation: LTER, Long Term Ecological Research.

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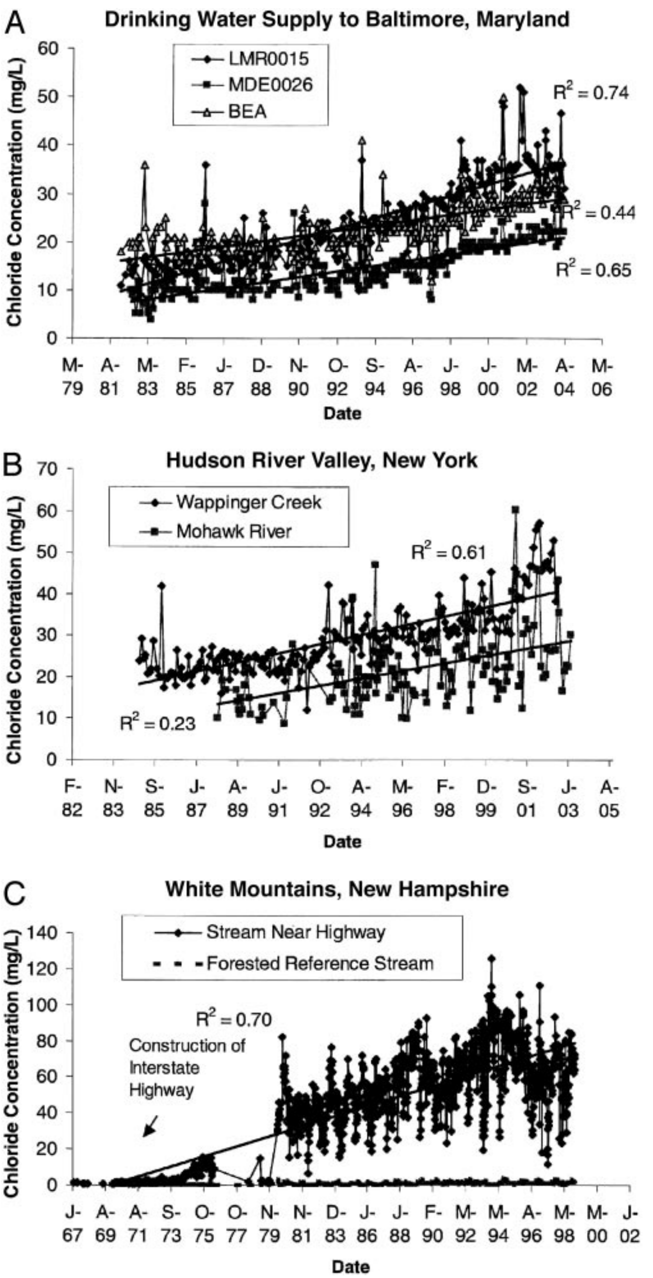

Fig. 1. Examples of significant, long-term increases in baseline concentration of chloride for streams and rivers of the northeastern United States. The R^2 values are given for linear regressions. All streams and rivers are located in rural areas but contain roads within their watersheds. (*A*) LMR0015 (Little Morgan Run), MDE0026 (Middle Run), and BEA (Beaver Run) are sampling stations for tributaries to Liberty Reservoir, a drinking water supply for Baltimore. (*B*) Wappinger Creek and the Mohawk River are tributaries to the Hudson River in the Hudson River Valley. (*C*) The streams in the White Mountains drain into Mirror Lake; one is located near an interstate highway in the Hubbard Brook Valley, and the forested reference stream is watershed 6 of the Hubbard Brook Experimental Forest (10).

bias toward storm flow vs. base flow), filtered in the field (47-μm glass microfiber and 0.45-μm-pore-size nylon filters), and analyzed for chloride by using a Dionex LC20 series ion chromatograph. Detailed site descriptions and sampling protocols are described in ref. 13. Baltimore LTER sites were not downstream of any wastewater treatment plants, which could release chloride. Municipal records indicate that >82,000 metric tons of NaCl were applied to roadways in the city of Baltimore (not including private property and interstate highways) as deicing material during the study period (14).

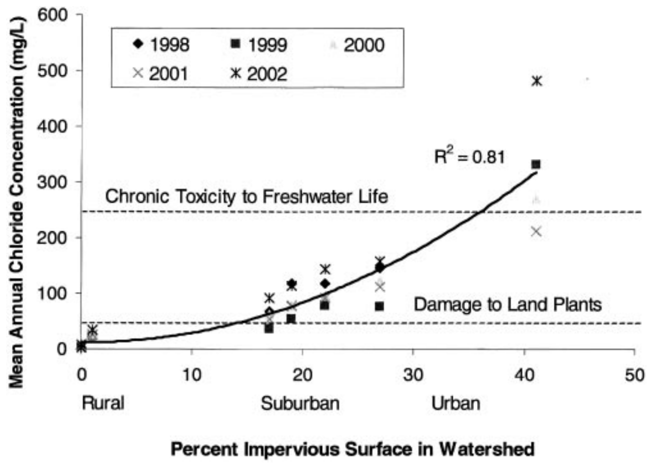

Fig. 2. Relationship between impervious surface and mean annual concentration of chloride in streams of the Baltimore LTER site during a 5-year period ($R^2 = 0.81$). Sites are located along a gradient of urbanization. Dashed lines indicate thresholds for damage to some land plants and for chronic toxicity to sensitive freshwater life (6, 8).

Results and Discussion

Rising Salinity in Rural Streams. Despite temporal fluctuations in precipitation in the northeastern United States throughout the study (11), we observed strong increases in the baseline concentration of chloride in rural watersheds with low density of roadways in Maryland, New York, and New Hampshire over the past 30 years (Fig. 1). In the White Mountains, chloride concentrations in some rural streams now exceed 100 mg/liter on a seasonal basis, which is similar to the salt front of the Hudson River estuary. Streams entering the Baltimore drinking water reservoir and streams and rivers of the Hudson River Valley also showed significant increases in concentrations of chloride over the past several decades ($P < 0.05$). We assumed a conservative linear rate of increase, although it may be possible that salinity is increasing at an exponential or logarithmic rate. If salinization were to continue to increase at its present linear rate (assuming no change in rates of road salt application or impervious surface coverage), we estimate that baseline chloride concentrations in many rural streams will exceed 250 mg/liter in the next century, thereby becoming toxic to sensitive freshwater life and not potable for human consumption.

Impervious Surface and Long-Term Salinization. Across the broader land-use gradient in Baltimore, we found that salinization of inland waters was strongly related to the amount of impervious surface coverage, and that chloride concentrations in many suburban and urban streams now already exceed the maximum limit (250 mg/liter) recommended for the protection of freshwater life. The mean annual concentration of chloride in Baltimore LTER streams increased as a logarithmic function as the relative amount of impervious surface increased within watersheds (Fig. 2). In developed areas with >40% impervious surface coverage, mean annual concentrations of chloride exceeded the thresholds of tolerance for sensitive taxa of freshwater life (6, 8). In suburban and urban streams, chloride concentrations remained elevated throughout winters, with peak concentrations of almost 5 g/liter (25% of the concentration of seawater) (Table 1). Interestingly, concentrations of chloride also remained elevated throughout the spring, summer, and

Table 1. Land use and peak chloride concentrations in streams of Baltimore

Station	Land use	Drainage area, ha	Population density, people per ha	Maximum Cl^- concentration, mg/liter			
				Winter	Spring	Summer	Fall
			With roadways				
Baisman Run	Suburban/forest (≈1% impervious surface)	381	1	38–116	19–29	22–37	23–29
Gwynnbrook	Suburbanizing	1,066	16	181–1,051	34–57	30–216	24–33
Glyndon	Suburban	81	9	229–1,509	79–117	96–469	72–606
Villa Nova	Suburban/urban	8,348	12	341–2,458	45–285	38–54	39–55
Dead Run	Suburban/urban	1,414	13	1,786–4,629	249–336	176–211	101–391
Carroll Park	Urban	16,278	20	960–2,085	63–86	44–86	49–66
			Without roadways				
Pond Branch	Forested	32.3	0	3–6	3–8	3–4	2–3
McDonogh	Agriculture	7.8	0	5–8	4–5	4–5	5–7

Range in the maximum concentration of chloride from 1998 through 2003 during winter, spring, summer, and fall in the streams of the Baltimore LTER site.

autumn and were up to 100 times greater than concentrations found in streams draining forest and agricultural watersheds without impervious surfaces. The relative contribution of salt from additional sources, such as septic effluent and discharges from water softeners, appeared to be low, compared with road deicers, which is reflected by the large peaks in chloride concentration during late fall and winter in both the rural streams from Maryland to New Hampshire and the urbanizing streams of the Baltimore LTER site (Fig. 1 and Table 1).

Ecological Implications. Our observations of long-term increases in chloride concentrations in northern rural areas and in rapidly developing areas located in relatively warm climates, such as Maryland, suggest that chloride pollution may be pervasive across seasons and large geographic areas of the northeastern United States. Over time, a gradual accumulation of chloride in groundwater can lead to elevated concentrations during baseflow conditions in the summer months and can contribute to long-term increases in the baseline salinity of surface waters (3, 5, 15). Related work has shown increasing salinity in the lakes of the midwestern United States due to road salt (16, 17). Even very large bodies of water have experienced increases in salinity, e.g., chloride concentrations in the lower Laurentian Great Lakes have increased to approximately 3 times their original concentration in the 1850s (18).

The concentrations of chloride that we observed in the Baltimore LTER streams are high enough to induce a variety of effects within both aquatic and terrestrial ecosystems. These effects include acidification of streams (19), mobilization of toxic metals through ion exchange or impurities in road salt (9, 20), changes in mortality and reproduction of aquatic plants and animals (21–23), altered community composition of plants in riparian areas and wetlands (23–25), facilitation of invasion of saltwater species into previously freshwater ecosystems (17, 25), and interference with the natural mixing of lakes (15). At relatively lower concentrations, salt also has been shown to alter the structure of microbial communities (26) and inhibit denitrification (27), a process critical for removing nitrate and maintaining water quality in surface waters. The undesirable effects of increasing salinity on particular taxa may influence broader ecosystem processes in aquatic systems related to primary productivity, decomposition, nutrient cycling, and the trophic complexity of food webs.

We also observed very large seasonal fluctuations in chloride concentrations, particularly in suburban and urban streams of the Baltimore LTER (increases in winters to almost 5 g/liter). These fluctuations may be particularly harmful to freshwater life, which may not have adaptations to adjust the osmotic potential of their cells over shorter time scales. Supporting work has shown that chloride associated with sodium can be more toxic than chloride associated with other divalent cations (21), and that minimum tolerance by organisms decreases with increasing temperature (28) and as fluctuations in its concentration increase (21). Thus, the effects of chloride pollution may be particularly pronounced in suburban and urban streams during summer base flow when dilution is minimal. Both elevated ranges and variability in concentrations of salt related to impervious surfaces can be a major influence on aquatic communities, even when other pollutants are absent or present in low amounts (29). Increased salinity should be included as an important ecological variable in explaining the extremely low abundance and diversity of freshwater life observed in inland waters draining rapidly developing landscapes (14, 29).

The increase in mean annual chloride concentration with impervious surface coverage suggests that chloride pollution is an increasing problem not only in rural areas with little population growth, but also in areas experiencing rapid and large changes in land use. Many cities and suburbs of the United States are within the range of impervious surface area reported for sites within the Baltimore metropolitan area. If the construction of roadways and parking lots were to continue to increase at its current rate, there likely would be large changes in baseline salinity across many northern regions of the United States and in other urbanizing areas throughout the world (14, 18, 19, 30). Many large cities receive substantially more snow than the mid-Atlantic region of the United States, which typically only has a mean annual snowfall of 46 cm, and concentrations of chloride presented in this study are not as elevated as those observed in more northern regions (5). Our observations suggest that even small changes in land use have resulted in large changes in the baseline salinity of aquatic systems. Moreover, the accumulation of road salt in aquifers and groundwater has eventually led to increased salinity throughout all seasons and across years in the northeastern United States and may persist for decades, even if use of salt is discontinued (14, 5). Given that land-use change is rapid in many regions of the world (9, 11, 14, 30–32), we suggest that salinization associated with increasing suburban and urbanization deserves attention as one of the most significant threats to the integrity of freshwater ecosystems in the northeastern United States.

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