Accumulation of deicing salts in soils in an urban environment

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Abstract Examining rates of deicing salt accumulation and leaching in urban soils is important for understanding the distribution and movement of salt in the environment. We examined autumn concentrations of deicing salts in soils in a moderately dense urban landscape in eastern New York State. The study area contrasted to the isolated, rural highways examined in previous studies. While NaCl was the most abundantly applied salt, Mg^{2+} (apparently from $MgCl_2$, a secondary deicing salt) was the most abundant salt cation in soils. Moderate Na^+ levels, and equivalent concentrations at depth and in surface samples, indicate that leaching of Na^+ is rapid in this system. Leaching may ameliorate toxicity for land plants but accelerate inputs to aquatic systems. In contrast to rural highway studies, where salt levels declined rapidly with distance to pavement, Na^+ remained elevated at the maximum distance measured. Airborne salt dispersal and dense networks of pavement likely contribute to widespread elevated salt levels. This semi-urban setting had salt levels high enough to be toxic to terrestrial plants and soil protozoa. Even moderate levels of development can have dramatic effects on salt inputs into soils and aquatic systems.

Keywords Anti-icing salt · Deicing salt · Impervious surfaces · Sodium ·
Magnesium · Urban soils

Introduction

Concerns about the extent and severity of environmental impacts of road deicing salt (also known as anti-icing salt, because most applications are done before ice accumulates) have concerned environmental scientists for decades (Hawkins 1971). While salt effects remain poorly understood, it is clear that salt levels have increased in recent years (Jackson and

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Jobbagy 2005; Kaushall et al. 2005). While abundant evidence exists that salts accumulate in aquatic systems (Siver et al. 1996; Mason et al. 1999; Rosenberry et al. 1999; Trombulak and Frissell 2000; Godwin et al. 2003; Jackson and Jobbagy 2005; Kaushall et al. 2005), damage terrestrial vegetation (Viskari and Karenlampi 2000; Bryson and Barker 2002), and can alter the composition of plant communities (Brauer and Geber 2002; DiTommaso 2004; Miklovic and Galatowitsch 2005), much remains unknown about how far salt disperses overland from roads, how much is retained in soils before passing on to plants and waterways, and which mechanisms (e.g., meltwater runoff or airborne spray and dust) are most important in salt dispersal (Fig. 1). Details on the distribution and concentrations of deicing salt in soils would help indicate both the geographic extent of deicing salt impacts and the relative importance of different pathways, including storm drains, leaching through soil, and airborne dust movement, in moving salts from paved surfaces.

Studies of salt storage in soils are especially needed in urban environments. Urban areas are responsible for a large portion of deicing salt use. As cities and suburbs expand in many regions, that use is growing. Most previous studies of the effects of deicing salts have been on remote, rural highways (e.g., Viskari and Karenlampi 2000; Norrström and Bergstedt 2001; Bryson and Barker 2002). On a remote highway, salt effects are easily attributed to as single source (a road), and salt effects are clearly distinguishable in an otherwise uncontaminated environment. It is unclear how well results of rural studies apply to urban areas, where road and sidewalk density is high, total impervious cover is high, and where a greater variety of relatively expensive deicers (such as calcium magnesium acetate or $MgCl_2$) is used. These compounds may act differently than the cheap and ubiquitous NaCl, and they may alter the storage and transport of Na^+, Cl^-, metals, and organic matter in the environment (Amrhein et al. 1992).

Studies of deicing salts in urban areas are also needed to determine the pattern of environmental responses to road salt applications as densities of urban land uses increase. For example, it is understood that areas with little development have low levels of salt accumulation and dense urban areas have high levels. Between these two points, much
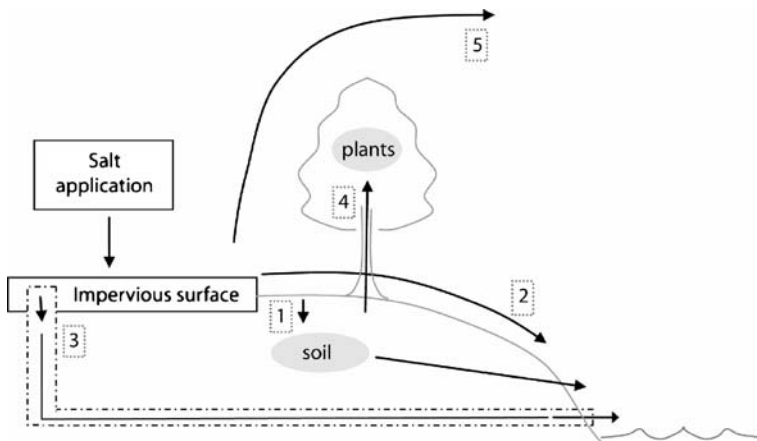

Fig. 1 Pathways by which salt ions may move after deicing salt applications. Pathways include (1) movement through soil to groundwater or surface water, with temporary or long-term storage in soils; (2) surface runoff with rain or snowmelt; (3) removal from terrestrial system by to waterways via storm drains; (4) uptake by plants, with temporary or long-term storage in plant tissues; (5) airborne movement of dry dust or liquid droplets. The present study focuses on soil storage (1), but storm drains (3) and airborne movement (5) can be important in urban environments with abundant traffic and dense road networks
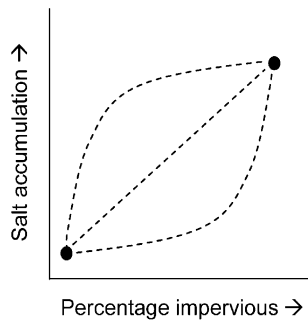
Fig. 2 Salt accumulation in soils and in the environment is known to be low in rural areas and high in urban areas (*solid dots*). Rates of increase in salt accumulation between these two endpoints are uncertain. Three possible scenarios are linear increase in salt storage with increasing impervious cover in a landscape, rapid salt accumulation at low levels of impervious cover, or gradual accumulation up to a threshold, after which salt accumulation rapidly approaches urban levels

remains unknown about the effects of increasing road density and salt accumulation. At intermediate levels of urbanization, does salt accumulation in the environment increase linearly with increasing urban density (Fig. 2)? If salt accumulation in soils increases nonlinearly, does it increase rapidly at low levels of urbanization, or does it accumulate slowly up to some threshold of urban density? These questions are important for understanding the influence of diffuse urbanization, including urban sprawl and exurban development, on the environmental effects of road salt use.

Salt use on roadways

Sodium chloride is the most commonly applied roadway deicer. The USA applies 8–12 million tons of NaCl to paved surfaces annually, at a cost of $240–$360 million dollars per year (Fischel 2001). New York State used an average of 19.6 tons salt/lane km (amounting to 1.1 million tons salt/year) in 2002–2006. Dutchess County, the location of this study, used an average of 12.2 tons salt/lane km in 2002–2006 (J. Ribakove, Public Information Officer, New York State Department of Transportation Region 8, 2007, personal communication) On the Vassar College campus, approximately 31 to 37 tons salt/lane km is used[1]. (J. Horst, Vassar College Director of Buildings and Grounds, 2006, personal communication) This high level of use results from concerns about pedestrian safety and from the risk of lawsuits for a private institution. Most salt used is NaCl, but the college also applies $MgCl_2$ near buildings because $MgCl_2$ causes less corrosion of mortar, bricks, and concrete than does NaCl. Costs of damage to infrastructure have not been quantified, but they include the replacement of stonework and brick masonry, road maintenance to repair salt-induced potholes, annual replacement of turf along paved surfaces, and some damage to landscape vegetation.

The United States Environmental Protection Agency (US EPA) does not consider road salt a contaminant and therefore does not regulate its application (Kaushall et al. 2005). Consequently there is no federal policy regarding the amount of road salt that can be used.

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[1]This value is based on a total usage of 250–300 tons of salt per year. We calculated the area of surfaces as follows: we digitized lengths of sidewalks, roads, and parking lots, assuming that roads have two lanes and sidewalks are 0.5 lanes wide.

Salting policies are driven by risk and liability, rather than environmental considerations. However, colleges are working nationwide to reduce their environmental impacts, and baseline data are needed to establish whether salt applications are leading to critical concentrations in the environment, and if so, to help inform policies regarding deicer use.

Environmental costs of deicing salts

Previous studies of deicing salts have found significant effects of deicing salt on soils, terrestrial vegetation, and aquatic fauna. Environmental effects of deicing salts have been reviewed by Ramakrishna and Viraraghavan (2005). Effects on soils can include displacement of nutrient cations, as well as reduction of soil permeability as Na^+ disperses soil colloids. Greater overland flow and soil erosion can result from lost soil permeability (Ramakrishna and Viraraghavan 2005). In addition, salt cations can displace and mobilize metal ions, including heavy metals, in soils (Amrhein et al. 1992; Bäckström et al. 2004). Retention of cations varies with soil type and is greatest in clay-rich soils because of the negatively charged surfaces of clay particles. Thus, Na^+ and Mg^{2+} accumulate differently, and have different degrees of impacts, depending on soil texture, clay content, and types of clay minerals present (Amrhein et al. 1992; Granato et al. 1995; Bäckström et al. 2004). Magnesium and calcium, which may be introduced in salts or in calcium magnesium acetate deicers, are more reactive than Na^+ within soil, so while Mg^{2+} and Ca^{2+} are less toxic than Na^+, they may displace nutrients and mobilize metals more effectively (Howard and Haynes 1993; Ramakrishna and Viraraghavan 2005).

Most studies of salt effects in soils have focused on damage to vegetation. In terrestrial and aquatic vegetation, both Na^+ and Cl^- can inhibit plant vigor and reproduction. Strongest effects on roadside vegetation have been observed within a few meters of salted surfaces (Richburg et al. 2001; Bryson and Barker 2002). Effects of NaCl differ among plant species, but in general detrimental impacts are a reduction in plant biomass, chlorosis, and the death of the plant (Viskari and Karenlampi 2000). In urban soils with heightened salinity, plant tissue have shown an increase in sodium and chloride ions and a decrease in potassium, magnesium, and calcium ions (Czerniawska-Kusza et al. 2004). The displacement of naturally occurring ions by Na^+ and Cl^- reduces the osmotic potential of the plant (Bryson and Barker 2002). Plant species composition may also be affected as salt tolerant invasive species replace salt-intolerant native species (Richburg et al. 2001; Miklovic and Galatowitsch 2005). Faunal effects include toxicity to aquatic invertebrates (Blasius and Merritt 2002; Benbow and Merritt 2004), and toxicity to larval stages of aquatic vertebrates such as salamanders (Turtle 2000; Sanzo and Hecnar 2006).

We examined the distribution of deicing salt cations (Na^+ and Mg^{2+}) in soils in a moderate-density urban environment, comprising 30% roads and sidewalks, in order to evaluate distribution and accumulation of these cations. We compared cation levels on roadside transects and at randomly located sampling points on a college campus in eastern New York State, as well as in presumably uncontaminated sites from a forest near the campus. We sampled at the end of the summer season, to evaluate levels of salt cations in soils at the "cleanest" time of year, which reflect minimum chronic levels of exposure for terrestrial flora and soil fauna. In addition to indicating the magnitude of salt accumulation and distribution of salts, observed distributions of salt in soils can provide insights into transport mechanisms. Greater accumulation adjacent to paved surfaces or downhill from pavement, relative to other locations, would indicate the importance of salt movement with soil water and overland flow. Elevated cation levels at tens of meters from salted surfaces, and not downslope of those surfaces, would indicate that airborne spray or dust contributes

to the distribution of salt in the environment. Greater accumulation at depth than at the surface would imply that leaching through the soil column was removing salt cations from the soil surface at a rate comparable to the rate of accumulation.

The mixed landscape of a college campus provides a useful contrast to previous studies that have focused on isolated rural highways (e.g. Bryson and Barker 2002; Godwin et al. 2003; Bäckström et al. 2004) or densely urbanized environments (Czerniawska-Kusza et al. 2004; Kaushall et al. 2005). In the northeastern USA, one of the most rapid types of land use change is diffuse urbanization on the margins of cities (Hasse and Lathrop 2003); thus a landscape of mixed impervious cover and lawns composes an increasingly important component of the regional environment. In addition, a semi-urban landscape adds data to a knowledge gap regarding the rate of salt accumulation in response to increasing impervious cover (Fig. 2). A college campus provides a moderately developed, mixed landscape of buildings, pavement, and vegetative cover, but unlike most residential and suburban areas, a college has the advantage that soil sampling can be done freely, without regard to property boundaries. In addition, campus landscape and road management are carried out centrally, so that records of salt use exist.

To examine the distribution of salt accumulation in soils, and to gain insights into the movement of salt from pavement into the environment, we tested two hypotheses: (1) salt cations (Na^+ and Mg^{2+}) would accumulate near impervious surfaces and decrease to near zero with distance; (2) salt cation concentrations would be greater at surface than at depth, reflecting application rates in excess of leaching rates.

Methods

Study area

The study area is a college campus in a suburban setting in Poughkeepsie, a city of 30,000 in eastern New York State, USA (41.7°N, 73.9°W). Poughkeepsie's urban core is ringed by expansive suburbs, ranging from fully urban to wooded, low-density residential areas, with an additional population of approximately 40,000. Vassar College, with a student population of 2,400, was built in 1852 on farmlands 3 km from the city center. The town has expanded around the campus in the past century, so that the college now sits within the suburbs at the edge of the dense urban core. The 76 ha of the campus comprises a mixture of mowed grass, trees, and shrubs, and buildings, as well as 22.5 ha of roads and parking lots, 3.6 ha of sidewalks, and 11.5 ha of buildings (Fig. 3). Salted impervious surfaces (roads, parking lots, and sidewalks) make up 34% of the campus area. Buildings contribute to impervious surface in the study area, but because they are not salted surfaces, they were excluded from analysis of salt applications. East of campus is a deciduous forest, interspersed with residential neighborhoods. This area was historically used for orchards and pasture and is currently wooded, dominantly with oak-maple deciduous trees between 50 and 100 years old. Reference "off-campus" samples were taken from this wooded area because it is adjacent to campus, with similar soils, but has a lower density of salted impervious surfaces and less disturbance on the landscape.

The region is humid and temperate, with an average precipitation of 1,270 mm annually and a snow season that normally lasts from approximately mid-December until late February or March (New England Integrated Science and Assessment 2006). Precipitation is distributed evenly through the year. In the 6 months preceding sampling, the area received 680 mm of summer precipitation (National Weather Service 2006).
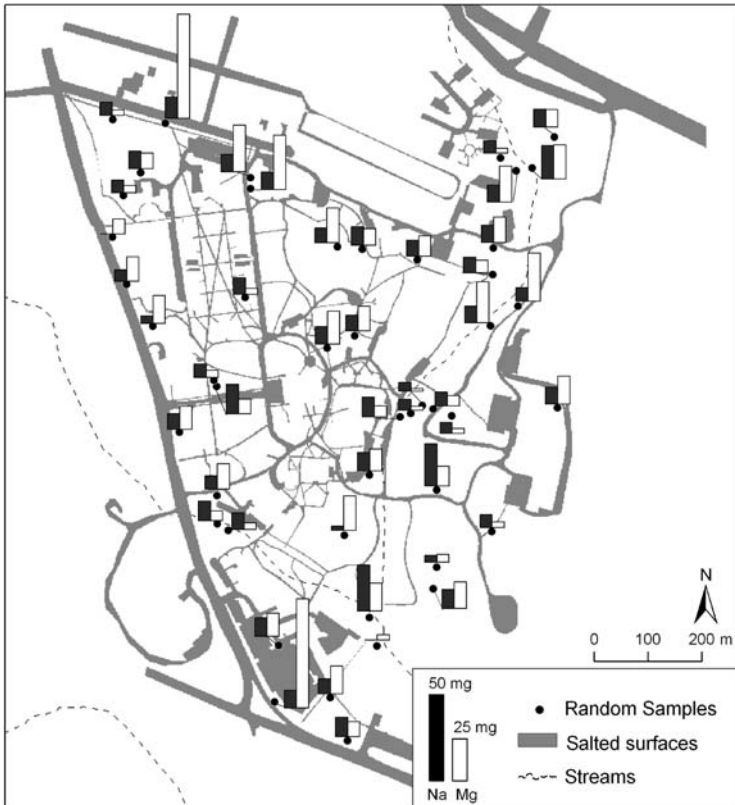
Fig. 3 Concentrations of Na$^+$ and Mg^{2+} for random samples at 12–14 cm depth. Values in bar graphs represent cation concentrations (mg/100 g soil) of cations in three pooled samples at each site

Soils in the area vary at a fine scale in texture, organic content, drainage, and parent material, as a result of the region's irregular topography, mixed land use, and glacial history. Soils in both on-campus and off-campus locations were silt loams in the Cardigan, Nassau, and Wayland series (Natural Resources Conservation Service 2002). All these series have the same reported ranges of pH (4.5 to 6.0) and cation exchange capacity (0 to 7.9 meq/100 g). Mineral content in soils derives from local shale, limestone, and dolomite, as well as from glacial till and clays. Glacial material originates from regions north of the study area with similar rock types. Dominant cation contributions from these materials would be Ca^{2+} (from calcareous rocks) and Al$^+$ (from shales) with slight amount of Mg^{2+} and K$^+$. Thus all soils sampled may contain some Mg^{2+} and Ca^{2+} deriving from local dolomite and limestone, but Na$^+$ is unlikely to originate in soil parent material. In addition, campus soils—like many urban soils—have been heavily modified over two centuries of land use and building. Thus soils are similar at the scale of soil survey units, but they vary at finer scales.

The College attempts to keep walks and roads clear during the snow season. On sidewalks, salt is applied in crystalline form from hoppers on small utility trucks, which broadcast a swath about 1 m wide, or it is scattered by hand. On roads and parking lots, salt is distributed mainly in crystalline form from trucks. These uses are similar to distribution methods used in cities and towns in the region, where homeowners distribute salt by hand and trucks dispense crystalline salt on roads.

Field methods

To establish the pattern of salt accumulation in the study area, we took soil samples at 50 random points in September and October 2006, before winter salt use. Fall samples allowed us to examine baseline conditions in the study area. Random points were generated in ArcMap (ESRI 2004) using Hawth's tools (Beyer 2006). In the field, we located the random points using a global positioning system (GPS) unit and high-resolution aerial photographs. In addition to random locations, samples were taken on short transects perpendicular to salted pavement. Transects were used to evaluate changes in salt concentrations in immediate proximity to salted surfaces. Three 3-m long transects were placed adjacent to salted surfaces, at locations in which high salt use had been observed in previous winters. One transect was taken adjacent to a curbed road, one adjacent to an uncurbed road, and one adjacent to a sidewalk.

In addition to random and transect samples on campus, we took five off-campus samples from the wooded area adjacent to campus, to represent areas uncontaminated by nearby salted surfaces. Off-campus sample sites were uphill from any paved or salted surfaces.

At each random and off-campus site, three soil cores were extracted within 50 cm of each other. On transects, three cores were taken in a row perpendicular to the transect line, at 50 cm intervals from 0 to 3 m on t line perpendicular to one edge of the paved surface. Cores were extracted using a 14-cm long, 5-cm diameter soil core extractor. For each sample site, the bottom 2 cm of all three samples was combined in a plastic bag (Bryson and Barker 2002; Czerniawska-Kusza et al. 2004). On transects and off-campus sites, surface samples (0–2 cm) were also collected to allow evaluation of leaching, or movement of cations with gravity from upper layers to lower layers. Although this approach provided an incomplete assessment of leaching, in the absence of known salt inputs, previous salt levels, drainage rates, and amounts of water applied, higher cation concentrations in deep samples than in shallow samples would indicate that leaching was removing cations from the surface soils at a rate equivalent to or greater than the rate of application per year.

Laboratory methods

Soil samples were allowed to air dry in the open bags before being mixed and loosened by hand while still in bags (Amrhein et al. 1992; Czerniawska-Kusza et al. 2004). After mixing, samples were sieved using a 1 mm sieve (Missouri Agricultural Experiment Station 1998; Czerniawska-Kusza et al. 2004). Samples were then placed on coffee filters and dried in a ventilated lab oven at 40°C for at least 24 h to remove remaining moisture. To extract ions from soil particles, 0.5 g of dry soil was mixed with 25 ml 0.1 M acetic acid. Acetic acid was used to mobilize all ions from the sample, including those ionically bonded to soil clays and organic matter (Missouri Agricultural Experiment Station 1998). Solutions were stirred for 15 min using a magnetic stirring plate, then vacuum filtered using folded 10 cm Whatman grade 1 paper filters. The filtrate was placed in 50 ml capped centrifuge tubes and stored at 4°C until use. Because filters contained small quantities of cations, we estimated the effect of filter blank contamination by passing 25 ml acetic acid through ten clean filters; the average value was subsequently subtracted from all samples before analysis. The minimum and maximum blank filter contributions were also subtracted from transect data, where we examined trends in most detail, in order to estimate the error in rates of distance decay in proximity to pavement.

We analyzed the Na^+, Mg^{2+}, and Ca^{2+} content of samples using a Spectro Genesis inductively coupled plasma optical emission spectrometer (ICP-OES). We report Ca^{2+}

because it was the most abundant cation measured, even though it occurs only as a trace element in the salts used on campus (Cargill Deicing Technology 2006). We tested for Cl^- levels by titrating using Mohr's method (Denny 1927), but Cl^- concentrations were below detection levels in both shallow and deep samples. Low levels of Cl^- can be attributed to relatively rapid leaching of negatively charged ions, in contrast to cations, which are attracted to negatively charged soil particles. We subsequently focused on cations in all analysis.

Spatial and statistical analysis

Salted surfaces were digitized using ArcGIS 9.2 (ESRI 2004) from high resolution (16 cm cell size) orthorectified aerial photographs taken in 2001. Digitized polygons of salted surfaces (sidewalks, roads, and parking lots) were converted to raster format at 1 m cell size for analysis. Three raster layers representing proximity to impervious surface were then calculated: (1) distance salted surfaces, (2) percentage of salted surfaces within 30 m of sample points, and (3) downhill distance (a variable that considers steepness of slopes in calculating the shortest downhill distance from pavement to a sampling point; calculated using the "cost distance" utility with inverse slope as the cost factor in ArcGIS). Cation concentrations at sample points were mapped using ArcGIS 9.2. Values from each of the three layers were extracted for each mapped sampling point, producing a table that contained cation concentrations and three explanatory variables for each sample point. We regressed cation levels against these three explanatory factors to evaluate the direction and approximate strength of trends. All statistical analysis was done using JMP 6.0 (SAS Institute 2006). To assess whether cations were leaching from the surface or accumulating at the surface, we compared shallow and deep samples using a non-parametric Kruskal–Wallis test. We also compared on-campus and off-campus samples using a Kruskal–Wallis test.

Results

Soil concentrations of Na^+ and Mg^{2+} varied widely across the study area (Fig. 3). Highest concentrations of cations occurred adjacent to roadways or downhill from parking lots; however, some sites adjacent to paved surfaces had low cation concentrations. Most abundant cations measured, were, in order, Ca^{2+}, Mg^{2+}, and Na^+. For all samples taken together, Ca^{2+} averaged 153 mg/100 g soil; Mg^{2+} concentrations averaged 30 mg/100 g soil; Na^+ averaged 13 mg/100 g soil, with a maximum of 57 mg Na^+/100 g soil. The average Na^+ level in deep samples (12–14 cm depth) on transects was 18 mg Na^+/100 g soil; random samples averaged 12 mg Na^+/100 g soil; off-campus samples averaged 9 mg Na^+/100 g soil. Magnesium levels in transect samples averaged 31 mg Mg^{2+}/100 g soil; random samples averaged 19 mg Mg^{2+}/100 g soil; off-campus samples averaged 2 mg Mg^{2+}/100 g soil. Levels of Mg^{2+} and Ca^{2+} were strongly correlated (Spearman's ρ correlation coefficient=0.90), but these cations were not strongly correlated with Na^+ concentrations (Spearman's ρ=0.47 for correlation between Na^+ and Mg^{2+}; ρ=0.38 for Na^+ and Ca^{2+}).

Transects had higher cation concentrations of Mg^{2+} than did random or off-campus samples, but ranges of Na^+ differed only slightly (Fig. 4). Aggregated on-campus samples (random+transect) versus off-campus samples, however, showed significantly lower concentrations of both Na^+ and Mg^{2+} in off-campus samples. For Na^+, Kruskal–Wallis χ^2=8.0 (p=0.004); for Mg^{2+}, χ^2=8.2 (p=0.005).

Among the three explanatory variables, distance to salted surfaces provided the best explanation for cation concentrations. For random points, distances from sample points to

Fig. 4 Comparison of cation concentrations among samples taken at 12–14 cm depth. Groups are: randomly distributed samples (*N*=50), Transects (curbed road, uncurbed road, and sidewalk transects, *N*=7 for each transect), and Off-campus samples (*N*=5). *Boxes* show 25th and 75th percentiles and median values; *whiskers* show 10th and 90th percentiles; outliers are also shown. *Y*-axes are stretched to fit data ranges, to show patterns within graphs
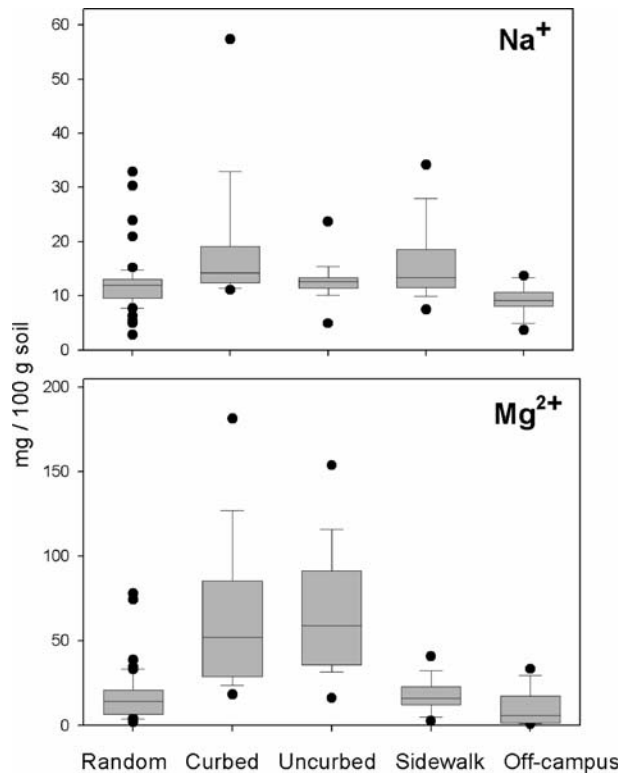
salted surfaces on campus ranged from 0 to 75 m. For off-campus samples, the distances ranged from 49 to 184 m. Among all deep samples, cation concentrations declined with distance, with the sharpest declines near to paved surfaces (Fig. 5). Regression using an exponential decay model described Mg^{2+} reasonably well, despite the fact that the model disregarded influential factors such as topography and local variability in soil characteristics. The exponential decay model described Na^+ poorly, as did other regression models; however Na^+ did generally decline with distance to paved surfaces.

On transects, concentrations of Mg^{2+} declined steeply with increasing distance to the pavement (Fig. 6), especially within the first 1–1.5 m. Sodium concentrations did not decline significantly on transects. Levels of Mg^{2+} varied among transects, with maximum levels ranging from 41 to 182 mg/100 g soil; Na^+ varied less among transects (maximum Na^+ on a transect ranged from 24 to 60 mg/100 g soil). Magnesium levels were lowest on the sidewalk transect and equivalent on the two road transects. Sodium levels were similar among the three transects.

Deep samples had lower levels of Mg^{2+} than did shallow samples, when all sample locations were taken together (χ^2=15.1, $p<0.0001$). Shallow and deep samples had equivalent levels of Na^+.

Discussion

Our results partially supported the first hypothesis that salt cations would show accumulation near impervious surfaces and decline with distance. Among all samples,

Fig. 5 Response of Na$^+$ and Mg^{2+} to distance from salted surfaces, using all deep samples (random, transect, and off-campus). An exponential decay model fits Mg^{2+} reasonably but fits Na$^+$ poorly (as did other regression models). *Y*-axes are stretched to fit data ranges, to show patterns within graphs
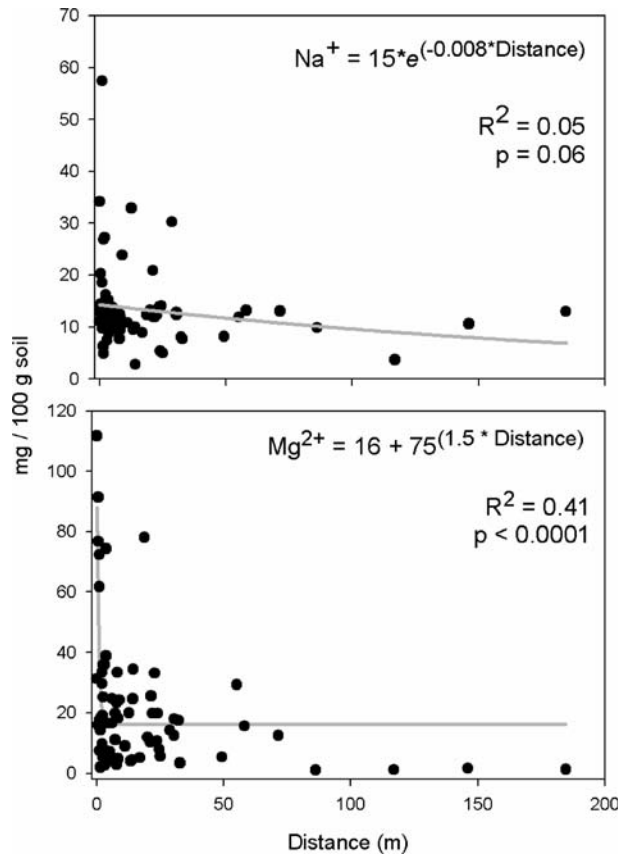
Na$^+$ and Mg^{2+} levels were higher near paved surfaces than far from them (Fig. 5), and for Mg^{2+}, declines continued to the maximum distance measurable on the campus. While this trend was similar for both cations, it was more dramatic (and more significant) for Mg^{2+}, which had very high levels near salted surfaces and very low levels at distance. In addition, levels of Mg^{2+} declined with distance on the 3-m transects (Fig. 6) and in a comparison of groups (Fig. 4). The lack of decline in Na$^+$ on transects does not necessarily contradict this hypothesis, because rapid leaching of cations through soil could account for steady levels of Na$^+$ (see discussion below).

Levels of Mg^{2+} dropped to near zero in off-campus samples, but Na$^+$ levels remained in the range of 10 mg Na$^+$/100 g soil at the maximum distance sampled (Fig. 5). Declines in Na$^+$ with distance to pavement were not significant when among groups (Fig. 4) or on transects (Fig. 6). The Na$^+$ results contradict our expectations and suggest that movement mechanisms other than direct infiltration and overland flow contribute to salt movement in this environment (see Fig. 1). Elevated salt levels at distances tens of meters from salted surfaces suggest that airborne movement of salt dust and spray are an important mechanism of cation distribution in this study area. Similar conclusions can be drawn from results on transects adjacent to curbed and uncurbed roads: levels of both cations were similar on these two transects, indicating that vectors such as spray and dust can move salt cations over curbs. While storm drains undoubtedly remove much salt from roadways, delivering it directly to stream channels, the elevated cation levels on transects indicates that much salt
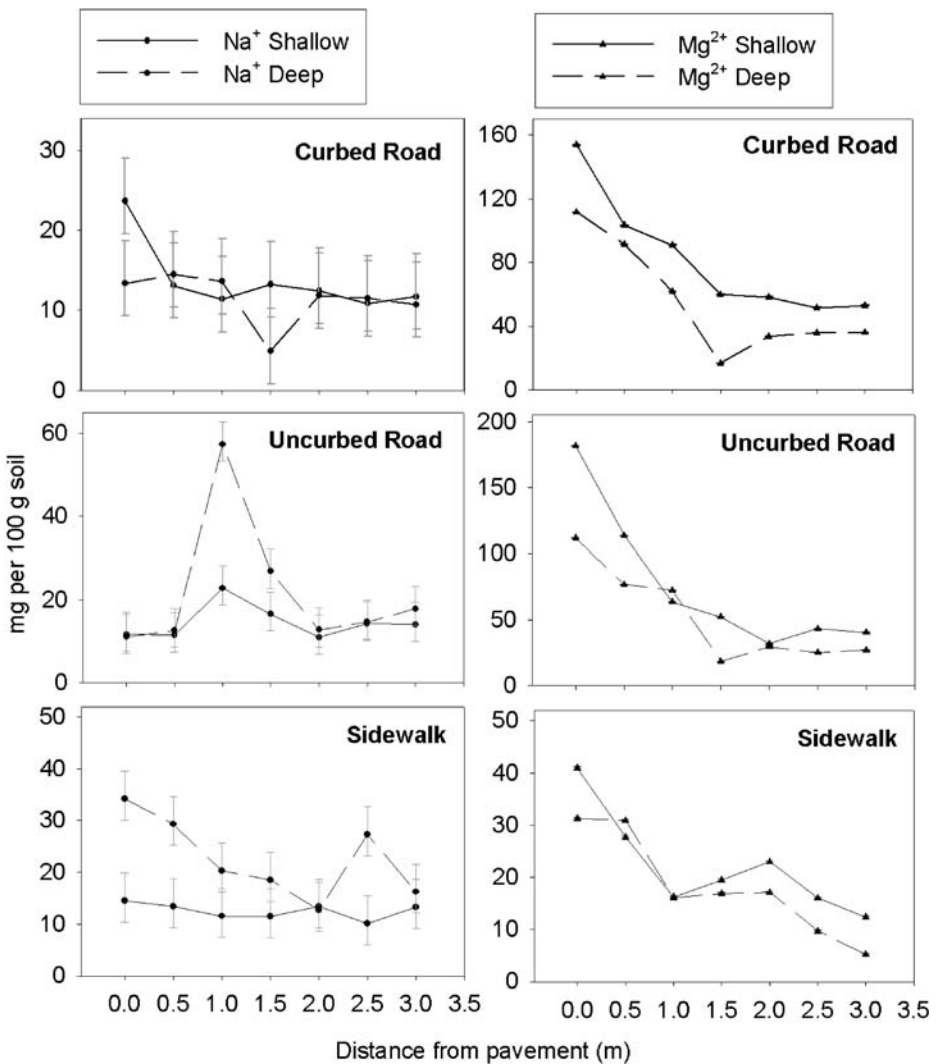
Fig. 6 Concentrations of Na$^+$ (*left*) and Mg^{2+} (*right*) with distance from pavement (*X*-axis) on transects adjacent to three different paved surfaces. Error bars (too small to see for Mg^{2+}) show minimum and maximum estimates of concentrations (see "Methods"). *Y*-axes are stretched to fit data ranges, to show patterns within graphs

stays in the environment despite the presence of storm drains. Surface runoff is undoubtedly an important factor in salt movement, but throughout our study area, salt accumulated in measurable levels despite surface runoff and storm drains. Our data do not directly address surface runoff, but in a study that examined salt removal by overland flow in Toronto, Canada, Howard and Haynes (1993) estimated that only 45% of Cl$^-$ was removed annually by surface runoff in Toronto, Canada, with the rest remaining in soil water or ground water. Thus storage in soils remains an important component in the salt movement and storage, even in a well drained urban environment.

Patterns of Na^+ distribution were consistent with ubiquitous salt sources, as well as airborne distribution of salts. Sodium chloride is applied on roads and sidewalks throughout the region, and the suburban road network is dense enough that no sample in this study was >184 m from a paved surface. The density of road networks in the surrounding area makes it likely that sampling points unaffected by airborne inputs from roads would be difficult to find in the study area. Ubiquitous salt distribution is a serious consideration in this environment because the area's isolated wetlands and vernal pools provide important breeding habitat for wood frogs, salamanders, invertebrates, and other taxa. Many of these groups show increased mortality or stress at even moderate levels of salt (Turtle 2000; Benbow and Merritt 2004; Ramakrishna and Viraraghavan 2005; Sanzo and Hecnar 2006).

Natural sources, especially long-range transport with precipitation, could partially account for widely distributed, moderate levels of Na^+ in sites far from pavement. Junge and Werby (1958) estimated Na^+ contributions from precipitation across the United States. For eastern New York, they reported an annual average of 0.5–1 mg Na^+/l. In a study of medium-range airborne transport, Williams and Stensland (2006) found that wind transported salts hundreds or even thousands of meters from major roads. Because none of our sites were more than 184 m from salted or paved surfaces, we cannot distinguish between atmospheric and local airborne inputs in our data.

Contributions of salt-associated cations to soil from local bedrock appeared to be minimal. Levels of Mg^{2+} in local dolomite should be at trace levels, relative to Ca^{2+} levels. In samples, Mg^{2+} levels were closer to one fifth of Ca^{2+} levels (sample averages were 30 mg Mg^{2+}/100 g soil and 153 mg Ca^{2+}/100 g soil). Moreover, these two cations declined steeply with distance from pavement, suggesting that the pavement was the principal source of these cations and that background contributions from parent material were slight. Local bedrock sources are unlikely to have contributed meaningful levels of Na^+ to samples.

Other factors associated with urbanization may also contribute to cation levels observed in this study. Pouyat et al. (1995) found that urban soils had higher concentrations of a number of metals, including Pb, Cu, Ni, Ca, Mg, and K, and total soluble salts. These contaminants may result from a wide variety of sources and causes in an urban environment (Paul and Meyer 2001). Because deicing salts are applied more abundantly than most other contaminants, it seems unlikely that such sources explain cation levels found in this study, but they may contribute to observed levels.

Evidence of leaching

Our second hypothesis, that cation levels would be greater in shallow than deep samples, was supported by our results for Mg^{2+} but not for Na^+. Levels of Na^+ were not significantly different in shallow and deep samples, indicating that Na^+ has been leached through the soil column at a rate exceeding or equaling application rates. Magnesium, on the other hand, was significantly higher in surface samples. Although NaCl is by far the most abundantly applied salt in the study area, concentrations of Mg^{2+} were much higher than those of Na^+. Thus retention of Mg^{2+} tended to be high, while Na^+ retention was low. Because of its stronger attraction to negatively charged soil particles, Mg^{2+} may accelerate Na^+ leaching by displacing Na^+ in soils. Leaching, in addition to cation displacement, is consistent with the flat trends in Na^+ on transects adjacent to salted surfaces.

In addition to Na^+ and Mg^{2+}, our samples had abundant Ca^{2+}, which was strongly correlated with Mg^{2+}. Because Ca^{2+} occurs only at trace levels in the salts applied on campus, the most likely source of Ca^{2+} is corrosion from concrete and mortar in sidewalks, steps, and building walls. Calcium released from pavement, together with Mg^{2+}, may

exacerbate release of Na^+ to groundwater and to surface aquatic systems. Faster movement of Na^+ through soils should make Na^+ less available to plants but more available to aquatic systems. Other studies have found that cations from deicing salts accelerate the release of heavy metals from soils to groundwater (Amrhein et al. 1992; Bäckström et al. 2004). The interactions of these different cations in releasing both Na^+ and other metals to ecosystems, should be more dramatic in urban than in rural watersheds. Their effects on vegetation and aquatic systems deserve further study.

Implications for understanding urban salt effects

In comparison to previous rural and urban studies, our Na^+ levels were intermediate, but concentrations declined more sharply in rural studies than in ours. Bäckström et al. (2004) found that salt concentrations dropped by an order of magnitude from 5 to 30 m from rural roads in Sweden. On rural highways in Massachusetts, USA, Bryson and Barker (2002) found 10–15 mg Na^+/100 g soil within 5 m, with a decline to 2 mg Na^+/100 g at 10 m from roads. Our Na^+ levels averaged 15 mg Na^+/100 g within 5 m and 11 mg Na^+/100 g soil at >5 m and 9 mg for off-campus samples. On urban roadsides in Opole, Poland (Czerniawska-Kusza et al. 2004), samples adjacent to salted roads had an average of 15 to 31 mg Na^+/100 g soil, but they did not report rates of decline with distance to pavement. Magnesium levels in the present study were much higher than those in Opole, where all samples had <1 mg Mg^{2+}/100 g soil. Our transect samples averaged from 19 to 66 mg Mg^{2+}/100 g. The use of $MgCl_2$ in our study area would account for this difference. The wide distribution of salts suggest that the density of roads and sidewalks, even when 34% of the landscape is composed of salted surfaces, is enough to make salt distributions in this semi-urban environment more similar to an urban setting than to a rural one. This suggests that the most likely response trend in Fig. 2 is the upper nonlinear curve. Additional studies of this type would be useful in further defining the shape of this trend and in identifying thresholds at which increasing road density makes the landscape function like an urban environment.

Levels of Na^+ were in a range that could negatively affect plant and animal life, although levels were not as great as expected given the high rates of application. Czerniawska-Kusza et al. (2004) noted that 26 mg Na^+/100 g soil can have toxic effects on protozoa, and they found necrosis and chlorosis in leaves at levels of 13.2 mg Na^+/100 g of soil. With a median level of 12 mg Na^+/100 g soil for all samples and high values in the range of 60 mg Na^+/100 g soil, Na^+ levels in approximately half our samples were high enough to damage some terrestrial plants. Even moderately salt-tolerant species such as eastern white cedar (*Thuja occidentalis*) are visibly damaged or dead in some parts of the study area in concentrated drainages from roads and parking lots. Rapid leaching, possibly exacerbated by the presence of Mg^{2+} and Ca^{2+}, as well as by the humid local climate, may account for moderate Na^+ levels despite heavy applications. High Mg^{2+} levels suggest that removal by storm drains is unlikely to account for moderate Na^+ levels.

Our best explanatory factor was distance to paved, salted surfaces. This measure was also the simplest to compute and to interpret. It is a useful conclusion that a simple distance measure helps explain salt distributions, even in this urban environment. Contrary to expectations, percentage impervious within 30 m of a sampling site and downhill distance provided little explanation. This small explanatory effect may have resulted from the short distance used (30 m), which included little variation in percentage impervious cover, and from our relatively small sample size. It seems likely, however, that multiple surrounding paved surfaces contribute to the wide distribution of elevated salt levels, as those levels declined less steeply in our study area than in rural environments. Although downhill

distance included two important factors—slope and distance—this measure was also less useful than simple distance. This result may reflect the coarse elevation data available for the area (10 m cell size), combined with the importance of very subtle topography for movement of overland flow, and the importance of other depositional mechanisms.

Conclusions

Deicing salt applications leave detectable levels of Mg^{2+} and Na^+ in urban soils, even at the end of the summer season in a humid climate. Salt concentrations in this semi-urban study area were more like urban than rural conditions: our samples had less dramatic declines in salt with distance to pavement, compared to rural highway studies, widespread high levels of salt, greater variety of salts used (including $MgCl_2$), and apparently rapid leaching of Na^+, possibly accelerated by Mg^{2+}. Despite leaching, Na^+ levels in our samples were high enough to pose risks to land plants. Multiple vectors, including airborne dust and spray, appear to help distribute salt in this environment, as indicated by elevated levels of both cations, even tens of meters away from salted surfaces. Water is therefore not the only transport mechanism, and drainage systems do not entirely remove salts from the terrestrial system. In this study area, we failed to find sampling sites distant enough from roads to be unaffected by salt inputs. The simultaneous effect of a network of paved surfaces is likely to contribute to widespread elevated salt levels. Wide distribution of salt raises serious questions about impacts of deicers on vernal pools and other habitats.

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