

The influence of chloride deicers on mineral nutrition and the health status of roadside trees in the city of Kielce, Poland

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Abstract Application of chemical road deicers has a negative impact on roadside vegetation. Every year, the trees in cities suffer from direct and indirect effects of salt application for winter road maintenance. To elucidate this problem in the city of Kielce, the chemistry of snow, soil, tree bark, and leaf samples has been investigated together with an assessment of the health status of the trees. Twelve investigation sites were selected along the roads with different traffic intensity. Snow samples were collected twice during the winter and analyzed for pH, EC, Na^+ , Ca^{2+} , Mg^{2+} , and Cl^- . In soil (collected from two depth intervals), tree bark, and leaf samples, the concentrations of B, Ca, Fe, K, Mg, Mn, N, Na, P, S, and Zn were determined. The contents of total organic carbon (TOC) in soils, as well as the pH of soil and tree bark samples were also measured. Nega-

tive symptoms revealed by roadside trees included the loss of assimilative apparatus and decreased vitality. The results of chemical analyses indicated that the snowmelt might be a substantial source of chloride ions and alkalizing substances that influenced higher pH of soils. The soil samples displayed elevated concentrations of S and Zn and lower than typical for soil contents of B, Mg, and TOC. The pH of alkaline soils favored greater bioavailability of B and reduced bioavailability of Na and Zn by the trees examined.

Keywords Sodium chloride · Calcium chloride · Magnesium chloride · Roadside trees · Snow · Soils

Introduction

Winter road maintenance requires using chemical substances for ice and snow removal. Sodium chloride was used as a deicer in the USA as early as in 1950s (Godwin et al. 2003), and the first comprehensive environmental impact assessment of highway deicing was published by the US Environmental Protection Agency in 1971 (US EPA 1971). Of the chemical deicers, the most popular is sodium chloride (NaCl), but calcium and magnesium chlorides (CaCl_2 , MgCl_2) are also widely used (Ramakrishna and Viraraghavan 2005). In

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the recent years, alternative road deicers have appeared commonly substituting for traditionally used chlorides in order to minimize their negative influence on the environment, building materials and metal corrosion (Robidoux and Delisle 2001; Hellstén et al. 2005). A variety of organic compounds were also tested as environmentally friendly deicers, including: calcium magnesium acetate, potassium acetate, sodium formate, calcium formate, ethylene glycol, and urea. Recently, new deicing agents, namely salts of levulinic acid produced from grain sorghum, have been proposed as an alternative to chlorides (Ganjyal et al. 2007).

Deicing substances are directly used on the road in the form of dry salts, moisturized/dissolved salts, or in a mixture with abrasive materials. They have widely been applied mostly to iced road surface, but some of the deicers have also been used to prevent ice-cover formation. Various transport mechanisms contribute to spreading of deicers to the roadside environment. By snowmelt runoff and infiltration, airborne spreading and plowing salts are distributed in the vicinity of roads (Blomqvist and Johansson 1999). The salt transport is the most intensive at a distance of about 10–20 m from the road (McBean and Al-Nassri 1987; Blomqvist and Johansson 1999). In other studies, on the influence of chemical deicers on urban soils and groundwaters, a downward migration of salts through the soil profile to bedrock has also been confirmed (e.g., Thunqvist 2004; Cunningham et al. 2008).

A long-term use of sodium chloride for winter road maintenance leads to accumulation of sodium and chloride especially in the urban environment (Godwin et al. 2003; Ramakrishna and Viraraghavan 2005; Green et al. 2008; Cunningham et al. 2008). It should be stressed that all the chemical deicers, regardless of their composition, influence the chemistry of waters, soils, and vegetation. The problem of groundwater and surface water contamination with chloride ions as an effect of road deicer application has been observed in many regions (Rosenberry et al. 1999; Thunqvist 2004; Novotny et al. 2008; Kincaid and Findlay 2009). Some of the authors claim (e.g., Howard and Maier 2007) that the raised concentrations of chlorides in groundwaters which are a source of drinking water for the

growing population of urbanized areas may be a serious problem for the future development of conurbations. The snowmelt runoff and influx of chloride-rich groundwaters to surface waters adversely influence biota, change stratification, and density gradients in lakes, leading to disturbance of water circulation (Judd 1970; Thunqvist 2004).

In addition, a long-lasting use of chemical deicers has a detrimental impact on soils. Chlorides used for melting ice and snow change the soil properties. The cation exchange sites in soils absorb Na^+ , Mg^{2+} , and Ca^{2+} , whereas the chloride ions are transported through the soil to ground and surface waters (Ramakrishna and Viraraghavan 2005). If these absorbed cations substitute for important plant nutrients, such as Zn, Cu, K, and Mn, this exchange can have a potentially detrimental effect to plant physiology. High concentrations of sodium in soil cause dispersion of soil colloids, leading to reduction in soil permeability and increased erosion rate (Cunningham et al. 2008). Accumulation of deicing salts in urban soils may lead to changes in ecological structure of soil organism communities (Green et al. 2008) with an increasing population density of salt-tolerant (halophilic) species.

Many authors have emphasized the negative effect of road deicers on roadside vegetation (e.g., Viskari and Kärenlampi 2000; Kayama et al. 2003; Trahan and Peterson 2007). Plants subjected to pollution caused by road deicers show negative symptoms at biochemical, physiological, and community levels. High concentrations of chlorides and sodium inhibit cell membrane functions, causes osmotic stress, and cell plasmolysis and decrease chlorophyll level leading to disorders in photosynthesis. Most of the studies on the influence of chemical deicers on plants concern the most popular salt—sodium chloride. The influence of NaCl on plants can be both direct (a high uptake and accumulation of sodium and chloride ions) and indirect (an influence of this salt on soil properties and plant nutrient bioavailability). The most often recorded response to high NaCl content in soils is leaf chlorosis and in more severe cases also necrosis. Laboratory studies on the influence of sodium chloride on different tree species seedlings revealed that the intensity of leaf necrosis depended on the salt tolerance of a given

tree species to the water regime and soil type (Fostad and Pedersen 2000). The salt tolerance of tree species may be an important factor for urban greenery designing (Sæbø et al. 2003). The currently published data displayed that magnesium chloride regarded as a more environmental friendly deicer (applied in amounts comparable to NaCl-based sand and salt mixtures) was more damaging to two pine species *Pinus ponderosa* and *Pinus contorta* (Trahan and Peterson 2008). Some information is also available on the negative impact of $MgCl_2$, used as a dust suppressant, on vegetation (e.g., Goodrich et al. 2008, 2009).

A negative influence of chemical deicers on the environment is also caused by substances used in technological processes of improving the quality of these products. These chemicals (named anticaking agents) encompass complex cyanide compounds (sodium ferrocyanide, ferric ferrocyanide), chromates, and phosphates (Ramakrishna and Viraraghavan 2005). Ferrocyanide compounds degrade in the environment to free cyanide ions bringing about a toxicological risk to living organisms. This degradation occurs when salt dissociates in water at $pH < 8$ and E_h greater than -600 mV. Fortunately, natural waters scarcely show both high alkalinity and low redox potential values (Novotny et al. 1998).

The recent studies on the environmental impact of chemical road deicers have stressed a complex influence of these substances on the environment. This complexity is highlighted by indirect (or secondary) effects of pollution on geochemical processes, especially on increasing mobility of heavy metals in soils (Bäckström et al. 2004; Norrström and Jacks 1998; Norrström 2005; Rasa et al. 2006), selective retention of Na, Ca, and Mg combined with high mobility of chloride ions in soils (Cunningham et al. 2008), or disturbance of nitrogen transformations in soils (Green et al. 2008). Although the results of comprehensive studies on the influence of road deicers on the environment have been published (e.g., Cekstere et al. 2008; Czerniewska-Kusza et al. 2004), still little is known on the geochemical behavior of salt constituents in snow–soil–plant systems. To give a better insight into this problem, we carried out a study on mineral nutrition and the health status of roadside trees in the city of Kielce (south-central

Poland). The principal objectives of this study were: (1) to compare the chemistry of snow and soils with concentrations of selected elements in leaves and bark of trees growing close to the roads and (2) to assess the health status of these trees and find the possible cause of negative symptoms.

Materials and methods

Location of investigation sites

The study was carried out in Kielce (south-central Poland). The city covers up an area of 109.45 km² and its geographic coordinates are $20^{\circ}38'$ East longitude and $50^{\circ}53'$ North latitude. The city of Kielce is located within the western part of the Kielce–Łagów Valley and is a capital of the Świętokrzyskie Province. The city is surrounded by hills and mountain ranges. The difference in elevation within the city limits ranges from 260 to 406 m above sea level. Kielce plays an important role in a road transport network of Poland. There are three national roads in the city. The mean traffic intensity in the city center is above 1,000 vehicles per hour.

Six streets were selected as investigation sites. Each of these sites encompassed two sampling points localized on opposite sides of the street (Fig. 1). The sites represented three different winter road maintenance strategies. At four sites (1a, 1b, 2a, 2b) NaCl was applied, whereas at eight sites NaCl and other chlorides were alternatively used: $CaCl_2$ (3a, 3b, 4a, 4b) and $MgCl_2$ (5a, 5b, 6a, 6b).

Of the investigation sites, the highest traffic intensity ($>2,000$ cars per hour) occurs at sites 2a, 2b, 4a, 4b, moderate traffic intensity (1,000–2,000 cars per hour) is recorded at sites 1a, 1b, 3a, 3b, and the lowest traffic intensity ($<1,000$ cars per hour) is noted at sites 5a, 5b, 6a, 6b (unpublished data derived from the Municipality of Kielce).

Sampling

Sampling was done in two phases. Snow samples were collected in the winter (February 1 and 26, 2009), whereas soil and plant samples in the late spring (June 9) of 2009. The reason for collecting



Fig. 1 Study area and localization of investigation sites

snow samples in a short period of time in the late winter was the possibility to compare snow chemistry in similar climatic conditions before spring snowmelt. Trees were sampled in June when the leaves were fully developed. Snow samples were placed in plastic containers. At each investigation site two soil samples were collected in plastic bags from the depth intervals of 0–0.15 m and 0.4–0.6 m. Bark and leaves of four tree species growing at a distance of 0.3 to 2 m from the road edge were collected in paper bags at 12 investigation sites and 4 control sites: ash *Fraxinus excelsior* L. (7 sites), linden *Tilia cordata* Mill. and *Tilia platyphyllos* Scop. (3), pine *Pinus sylvestris* L.

(4) and maple *Acer pseudoplatanus* L. (2). Each composite plant sample consisted of about 10 subsamples. All these samples were transported to the Laboratory of Geochemistry and the Environment Division of Jan Kochanowski University in Kielce. During sampling stainless steel tools were used and precautions were made to prevent the samples from contamination.

Sample preparation and analyses

The snow samples were melted at ambient temperature (about 20°C) and filtered. Subsequently, the pH, and conductivity (EC) were measured with a CP-103 pH-meter and a CC-101 EC-meter (Elmetron, Poland). The concentrations of sodium in the snow samples were determined with a sodium ion selective electrode using a CX-742 multifunctional measuring apparatus (Elmetron, Poland). The chloride ion concentrations were determined with a LF205 spectrophotometer (Slandi, Poland). Calcium and magnesium ion concentrations were analyzed using complexometric titration method.

The soil samples were air-dried and disaggregated with a Fritsch's blender to pass a 0.063 mm sieve. One gram of each soil sample was digested with 50 ml of aqua regia in a closed microwave system (MDS 81, CEM Corp.). The air-dried plant samples were pulverized with an IKA Werke plant material blender and sieved to 0.5 mm fraction. The plant samples were digested in a closed microwave system with HNO₃ and H₂O₂ (0.5 g of sample + 5 ml HNO₃ + 1 ml H₂O₂) (US EPA 1996). In addition, for the purpose of the pH measurements, the disaggregated soil and bark samples were placed in beakers and mixed with deionized water in the proportion: 10 g + 25 ml (soil) and 1.5 g + 20 ml (bark). After 24 h, the pH of filtered solutions was measured using a CP-103 pH meter.

The soil and plant samples were analyzed for B, Ca, Fe, K, Mg, Mn, Na, P, S, and Zn using inductively coupled plasma optical emission spectrometry (ICP-OES; spectrometer Jobin–Yvon model JY 70 PLUS with vertical plasma). For analytical quality assurance, 10% of all samples were randomly selected for routine replicate analyses, and

reference standard materials, i.e., Lake Sediment LKSD-3 (for soils) and Citrus leaves SRM-1572 (for plants), were used. The total organic carbon (TOC) content of soil samples was determined with an automatic Coulometric titration method (Coulomat 702 C/S, Ströhlein, Germany).

The concentrations of total nitrogen were determined using an automated spectrophotometric flow injection analyzer (FIAstar, FOSS) (Lima et al. 2000). Digestion of samples was done in a digestion block (FOSS Tecator DS 12) with H₂SO₄.

The element determinations were performed in the Central Chemical Laboratory of the Polish Geological Institute in Warsaw, whereas the remaining analyses in the Laboratory of Geochemistry and the Environment Division of Jan Kochanowski University in Kielce

Tree health status assessment

The tree health status assessment was done on the day of soil and plant sampling (June 9, 2009). Measurements and observations (tree diameter at 1.3 m above ground level (DBH), degree of defoliation, tree top shape, and vitality) were considered when analyzing the health status of the trees examined. As health status cannot be measured directly, various indicators can instead serve to describe tree vitality (Dobbertin 2005). Field practical methods are of great importance in tree health status assessment. The most importance field methods include crown foliage (degree of defoliation) and crown morphology (tree top shape).

The selected trees were classified according to the following characteristics (Borecki and Keczyński 1992): (1) degree of defoliation (four classes: 0—no defoliation; loss of assimilative apparatus up to 10%, 1—slight defoliation; loss of assimilative apparatus from 11% to 25%, 2—moderate defoliation; loss of assimilative apparatus from 26% to 60%, 3—severe defoliation; loss of assimilative apparatus more than 60%), (2) tree top shape (four classes: 1—conical top, 2—narrowly paraboloidal top, 3—broadly paraboloidal top, 4—flat top); (3) vitality (five classes: 1—trees luxuriantly developed, 2—trees normally developed, 3—trees slightly weakened,

4—trees strongly weakened, 5—dying trees). The degree of tree damage was assessed on the basis of percentage degree of defoliation, according to the European forest monitoring methodology (Lorenz 1995). In determination of vitality all characteristics giving a full picture of the tree were taken into account: degree of defoliation, tree top shape, and some other characteristics as for example length of the crown and degree of crown deformation (Innes 1993). For comparison, the distribution patterns of degree of defoliation, tree top shape, and vitality between sites investigated, the Kruskal–Wallis test was applied. For any characteristic for which a difference was observed with the Kruskal–Wallis test, the significance of differences between groups was examined by the Bonferroni test.

Results

Snow

The results of pH, EC, and ion determinations in the snow samples collected in two series are presented in Table 1. The pH of snow from the first sampling series was in the range of 6.6–8.1 with a mean of 7.3, and from the second series in the range 6.0–7.1 (mean of 6.8). The distinct differences in electrical conductivity were noted between the two sampling series. The much higher EC values (242–5,080 $\mu\text{S}/\text{cm}$; mean of 1,320 $\mu\text{S}/\text{cm}$) were recorded in the snow collected during the first sampling series. The EC of snow from the second series were lower varying from 160 to 492 $\mu\text{S}/\text{cm}$ (mean of 286 $\mu\text{S}/\text{cm}$). The high EC values reflected the distinctly higher ion concentrations. The mean concentrations of chloride ions were four times higher, calcium and magnesium ions almost twice, and sodium ions seven times higher in the snow from the first sampling series.

Soil

The chemical composition and pH of soil samples are displayed in Table 2. In contrast to the deeper sampling interval (0.4–0.6 m), the surface soil (0–0.15 m) was enriched in most of the elements,

Table 1 Physicochemical characteristics of snow samples

Site no.	EC ($\mu\text{S}/\text{cm}$)		pH		Cl ⁻ (mg/L)		Na ⁺ (mg/L)		Mg ²⁺ (mg/L)		Ca ²⁺ (mg/L)	
	1 Series	2 Series	1 Series	2 Series	1 Series	2 Series	1 Series	2 Series	1 Series	2 Series	1 Series	2 Series
1a	963	477	7.7	7.2	180	105	175	78.1	<1.00	0.97	20.40	16.00
1b	1,800	193	7.8	6.3	290	35	159	20.2	1.22	0.49	26.00	12.80
2a	2,490	445	7.3	7.1	365	79.5	458	58.7	7.29	0.97	56.10	20.80
2b	1,330	278	7.5	6.9	285	45.5	243	28.8	<1.00	<0.10	28.10	18.40
3a	2,980	492	6.6	6.9	445	87	210	23.1	<1.00	0.49	289.00	56.10
3b	3,880	160	7.8	6.0	435	27	450	4.2	<1.00	0.97	377.00	20.80
4a	5,080	219	7.2	7.0	780	40.5	374	8.4	1.22	4.37	351.00	25.60
4b	525	218	6.9	7.0	99	39	27.7	7.0	12.10	1.46	44.10	24.00
5a	830	392	6.9	7.0	175	69.5	148	53.5	2.44	3.40	14.00	14.40
5b	242	176	6.9	6.3	49.5	29.5	37	20.1	2.44	0.49	12.00	6.41
6a	850	302	8.1	7.0	90.5	84.5	141	60.8	12.10	1.46	16.00	14.40
6b	906	331	7.1	6.8	200	69.5	149	44.8	14.60	2.43	16.00	11.20

For sample locations, see Fig. 1

Table 2 The pH and element concentrations in soils

Site no./sampling depth	pH	Ca %	Fe	K	Mg	N	Na	P	S	TOC	B mg/kg	Mn	Zn
1a (0–0.15 m)	7.6	1.27	1.14	0.12	0.20	0.42	0.08	0.10	0.06	5.17	8	447	132
1a (0.4–0.6 m)	8.1	1.38	1.63	0.15	0.17	0.19	0.06	0.05	0.02	1.73	8	478	91
1b (0–0.15 m)	7.8	1.89	1.62	0.22	0.23	0.37	0.05	0.07	0.04	3.32	12	550	145
1b (0.4–0.6 m)	7.7	1.28	2.07	0.19	0.18	0.15	0.03	0.04	0.02	1.23	8	703	84
2a (0–0.15 m)	7.9	1.92	1.73	0.18	0.26	0.40	0.03	0.08	0.06	5.01	14	460	242
2a (0.4–0.6 m)	7.6	2.30	1.74	0.16	0.23	0.27	0.03	0.07	0.04	3.79	14	481	274
2b (0–0.15 m)	7.7	2.24	1.68	0.20	0.28	0.42	0.03	0.08	0.07	3.43	16	567	471
2b (0.4–0.6 m)	8.0	1.92	1.19	0.15	0.14	0.21	0.02	0.06	0.03	1.99	11	554	163
3a (0–0.15 m)	7.4	0.89	1.45	0.16	0.17	0.35	0.02	0.08	0.05	3.74	11	348	184
3a (0.4–0.6 m)	7.6	0.56	1.12	0.10	0.12	0.12	0.02	0.04	0.02	1.60	7	304	127
3b (0–0.15 m)	8.2	1.69	1.60	0.18	0.17	0.42	0.01	0.05	0.04	2.25	9	342	70
3b (0.4–0.6 m)	7.5	1.49	1.65	0.16	0.22	0.37	0.02	0.08	0.05	3.55	13	429	195
4a (0–0.15 m)	8.3	2.86	1.27	0.13	0.29	0.87	0.02	0.08	0.16	8.88	12	343	194
4a (0.4–0.6 m)	7.8	0.76	1.00	0.09	0.12	0.05	0.02	0.05	0.02	3.29	6	136	64
4b (0–0.15 m)	8.0	5.72	1.63	0.17	0.49	0.60	0.04	0.11	0.13	10.4	18	498	391
4b (0.4–0.6 m)	7.7	0.78	1.36	0.14	0.14	0.15	0.02	0.10	0.03	1.91	10	607	112
5a (0–0.15 m)	7.6	2.13	1.13	0.11	0.13	1.62	0.03	0.08	0.17	17.8	17	1570	392
5a (0.4–0.6 m)	6.6	0.16	0.63	0.06	0.03	0.02	0.02	0.02	0.01	1.14	4	848	40
5b (0–0.15 m)	7.8	3.63	1.53	0.14	0.16	0.41	0.01	0.08	0.05	5.62	12	384	148
5b (0.4–0.6 m)	8.0	1.59	1.47	0.12	0.13	0.08	0.01	0.08	0.02	1.69	8	363	102
6a (0–0.15 m)	7.9	3.22	2.37	0.14	0.21	0.43	0.06	0.10	0.10	9.00	23	530	290
6a (0.4–0.6 m)	7.5	0.25	1.05	0.13	0.10	0.01	0.02	0.04	0.01	0.78	7	333	45
6b (0–0.15 m)	8.0	1.05	1.62	0.14	0.14	0.55	0.01	0.05	0.07	9.33	11	448	197
6b (0.4–0.6 m)	7.6	0.70	1.11	0.08	0.08	0.16	0.01	0.05	0.03	3.37	9	299	94

For sample locations, see Fig. 1

Table 3 The descriptive statistics of selected tree parameters

Site	Tree species	Mean diameter at breast height (cm)	Defoliation degree (%)			Vitality		
			Min.	Median	Max.	Min.	Median	Max.
1a	<i>Fraxinus excelsior</i> L.	19.0	20.0	27.5	30.0	2.0	2.5	3.0
1b	<i>Fraxinus excelsior</i> L.	19.6	10.0	20.0	50.0	1.0	2.0	4.0
2a	<i>Fraxinus excelsior</i> L.	41.3	20.0	60.0	60.0	3.0	4.0	4.0
2b	<i>Fraxinus excelsior</i> L.	44.5	30.0	60.0	90.0	3.0	4.0	5.0
3a	<i>Tilia cordata</i> Mill.	29.0	10.0	17.5	60.0	2.0	2.5	4.0
3b	<i>Tilia platyphyllos</i> Scop.	23.4	10.0	20.0	60.0	2.0	2.0	4.0
4a	<i>Fraxinus excelsior</i> L.	19.6	20.0	60.0	70.0	2.0	4.0	5.0
4b	<i>Fraxinus excelsior</i> L.	19.7	50.0	60.0	85.0	3.0	4.0	5.0
5a	<i>Pinus sylvestris</i> L.	30.5	25.0	35.0	75.0	2.0	3.0	5.0
5b	<i>Pinus sylvestris</i> L.	47.7	30.0	32.5	35.0	2.0	2.5	3.0
6a	<i>Acer pseudoplatanus</i> L.	20.0	10.0	10.0	10.0	2.0	2.0	2.0
6b	<i>Pinus sylvestris</i> L.	25.5	70.0	70.0	70.0	4.0	4.0	4.0
CS1	<i>Pinus sylvestris</i> L.	28.3	10.0	17.5	45.0	1.0	2.5	3.0
CS2	<i>Acer pseudoplatanus</i> L.	45.0	20.0	20.0	20.0	2.0	2.0	2.0
CS3	<i>Tilia cordata</i> Mill.	36.5	25.0	25.0	25.0	2.0	2.0	2.0
CS4	<i>Fraxinus excelsior</i> L.	28.0	15.0	15.0	15.0	2.0	2.0	2.0

Table 4 The pH and element concentrations in tree bark and leaves

Species	Medium	pH	Ca %	Fe	K	Mg	N	Na	P	S	B mg/kg	Mn	Zn
<i>Fraxinus excelsior</i> L.	1a1 (bark)	5.9	1.83	0.15	0.45	0.18	1.08	0.051	0.090	0.113	26	40	84
	1a2 (leaves)	ND	0.97	<0.01	1.41	0.20	2.21	0.005	0.264	0.148	18	22	22
	1b1 (bark)	5.3	1.48	0.13	0.31	0.13	1.15	0.024	0.086	0.122	23	33	74
	1b2 (leaves)	ND	0.77	<0.01	1.19	0.23	2.15	0.004	0.286	0.172	19	26	26
<i>Tilia cordata</i> Mill.	2a1 (bark)	6.4	3.04	0.22	0.15	0.13	1.18	0.083	0.058	0.193	52	61	150
	2a2 (leaves)	ND	1.31	0.02	1.44	0.29	2.40	0.010	0.279	0.234	35	36	45
	2b1 (bark)	6.0	2.65	0.32	0.19	0.11	0.96	0.086	0.065	0.173	35	75	182
	2b2 (leaves)	ND	1.30	0.02	1.42	0.35	2.18	0.037	0.359	0.199	23	22	31
<i>Tilia platyphyllos</i> Scop.	3a1 (bark)	5.7	3.23	0.18	0.13	0.07	1.22	0.031	0.069	0.167	25	76	86
	3a2 (leaves)	ND	2.42	0.03	1.03	0.26	3.89	0.009	0.383	0.247	69	143	40
	3b1 (bark)	5.3	2.00	0.21	0.14	0.06	1.63	0.015	0.097	0.180	20	53	90
	3b2 (leaves)	ND	1.46	0.02	1.73	0.18	2.99	0.006	0.281	0.214	57	67	20
<i>Fraxinus excelsior</i> L.	4a1 (bark)	5.8	1.73	0.09	0.62	0.16	0.85	0.124	0.095	0.107	34	27	56
	4a2 (leaves)	ND	1.14	0.01	1.43	0.17	2.04	0.005	0.315	0.215	22	13	21
	4b1 (bark)	5.9	2.50	0.14	0.48	0.16	1.07	0.036	0.072	0.112	31	38	91
	4b2 (leaves)	ND	1.06	<0.01	1.15	0.15	2.03	0.007	0.241	0.225	20	9	18
<i>Pinus sylvestris</i> L.	5a1 (bark)	3.6	0.83	0.05	0.04	0.01	0.28	0.014	0.013	0.102	6	76	34
	5a2 (leaves)	ND	0.34	0.02	0.44	0.05	1.50	0.053	0.105	0.099	23	216	43
	5b1 (bark)	4.2	0.66	0.03	0.04	0.01	0.23	0.009	0.013	0.052	6	9	17
	5b2 (leaves)	ND	0.50	0.01	0.55	0.08	1.89	0.015	0.124	0.114	40	26	25
<i>Acer pseudoplatanus</i> L.	6a1 (bark)	5.2	2.88	0.04	0.22	0.09	1.28	0.013	0.071	0.122	20	80	34
	6a2 (leaves)	ND	1.63	<0.01	1.47	0.24	2.44	0.007	0.197	0.190	31	74	36
	6b1 (bark)	3.6	0.84	0.06	0.03	0.01	0.26	0.013	0.012	0.094	6	40	28
	6b2 (leaves)	ND	0.56	0.02	0.40	0.08	1.49	0.065	0.110	0.123	15	65	29
<i>Pinus sylvestris</i> L.	CS1 (bark)	3.8	0.73	0.05	0.04	0.01	0.33	0.009	0.016	0.057	5	16	24
	CS1 (leaves)	ND	0.64	0.02	0.47	0.08	1.70	0.012	0.122	0.104	41	92	41
	CS2 (bark)	5.4	3.36	0.02	0.13	0.06	0.79	0.008	0.042	0.078	15	162	27
	CS2 (leaves)	ND	0.83	<0.01	1.41	0.13	2.04	0.006	0.182	0.177	22	142	27
<i>Tilia cordata</i> Mill.	CS3 (bark)	5.1	1.82	0.06	0.08	0.03	1.03	0.004	0.045	0.104	14	53	24
	CS3 (leaves)	ND	1.07	<0.01	2.11	0.18	3.15	0.003	0.290	0.257	45	253	19
	CS4 (bark)	5.7	1.73	0.03	0.67	0.21	0.86	0.014	0.057	0.073	30	16	51
	CS4 (leaves)	ND	0.97	<0.01	0.83	0.22	2.23	0.006	0.180	0.241	25	22	28

For sample locations, see Fig. 1
 ND not determined; CS control site

except for potassium which was distributed evenly in the soil profile. The total organic carbon and pH also showed increased values in the upper part of the soil profile.

Plant material

The descriptive statistics of selected tree features is given in Table 3 and the chemical composition of bark and leaf samples is presented in Table 4. There are interspecies differences in accumulation of elements in the bark and leaves. The pH of ash bark was similar to that of linden and maple bark (about 5.5–6.00), whereas the pine bark was more acidic (3.6–4.2). Some of the elements were accumulated in the bark (Ca, Fe, Mn, Na, Zn) and the others in the leaves (B, K, Mg, N, P, S).

The health status assessment showed that there were negative symptoms within the trees examined. Statistically significant differences were found in the following parameters: (1) the loss of assimilative apparatus between sites 2a, 2b and 1a, 1b, control site no. 3; sites 4a, 4b and 1a, 1b, control site no. 3 (Kruskal–Wallis test, $p < 0.0001$); (2) the tree-top shape between sites 2a, 2b and control site no. 3 (Kruskal–Wallis test, $p = 0.0023$); (3) vitality between sites 2a, 2b and 1a, 1b, control site no. 3; sites 4a, 4b and 1a, 1b, control site no. 3 (Kruskal–Wallis test, $p < 0.0001$).

Discussion

Chemistry of snow, soil, and tree bark and leaves

Snowpack is the most important pollution source when interpreting the influence of chemical deicers on the environment. Most of the analyzed snow samples exhibited the raised concentrations of ions originating from the chlorides used. This is particularly evidenced by the concentrations of calcium ion at sites 3 and 4. Major variations in the chemistry were noted between snow samples collected on February 1 and 26. This may be explained by the differences in weather conditions (e.g., wind strength and direction, temperature) during sampling, as well as by the duration of snowpack. During the first sampling series, the

snow cover was fresh (samples were collected on the consecutive day after the snowfall) averaging about 2 cm in thickness. In contrast, during the second sampling series, the snowpack was about 30 cm thick following heavy snowfall.

There are strong, statistically significant positive correlations ($p < 0.05$) between EC and chloride concentrations in the snow examined ($R^2 = 0.93$ in the first series and $R^2 = 0.82$ in the second series). Distinctly higher correlation coefficients are also noted between EC and Ca^{2+} ($R^2 = 0.82$) as well as EC and Na^+ ($R^2 = 0.65$) in the first snow sampling series. This evidence suggests that the chloride ion concentrations are the major factor that influences the EC values in the snow samples.

The pH of soil samples was slightly alkaline varying from 6.6 to 8.3 with a mean of 7.7. The similar pH values (6.66–8.11) were found in soils by Green and co-authors at Selside (UK), where roads were maintained with sodium chloride (Green et al. 2008). Roadside soils collected near Stockholm (Sweden) revealed a pH close to neutral in the 6.6–7.0 range (Norrström and Jacks 1998). It should be emphasized that soils of the Kielce area are developed on Upper Devonian marlaceous clayey shales, marls, and limestones, and that is why their natural pH exceeds 6.5. Salts used as deicers may cause an increase of pH values in soils, which is evidenced by a higher pH in the surface layer of eight sampling points.

The use of calcium chloride as a deicer at sites 3a, 3b, 4a, and 4b might have been contributed to calcium accumulation in soils. Compared to sodium, calcium is easier retained in soils. The long term application of calcium chloride may trigger the geochemical interactions of Ca with other elements in soils and affect their bioavailability for plants.

Concentrations of some elements in the soils examined do not substantially differ from those reported as typical levels in soils, for example, Fe and partly Mn (Kabata-Pendias and Pendias 2001), or found in natural forest ecosystems of the Holy Cross Mountains, for example, K and P (Gałuszka 2005).

The mean concentration of Na in soils from unpolluted sites is 0.63% (Tan 2005) and in the soils examined the content of this element varied from 0.01% to 0.08%. Nearly the same

concentrations of Na (0.013–0.057%) were found by Cunningham and co-authors (2008) in roadside soils of New York State. According to Kayama and co-authors (2003), roadside soils in Northern Japan contained similar concentrations of sodium (0.009–0.047%). The same levels of this element (0.003–0.033%) were also found in roadside soils in Opole (Czerniewska-Kusza et al. 2004).

The roadside soils of the Kielce area showed lowered contents of magnesium (0.03–0.49%) and TOC (0.78–17.80%). Similarly, low concentrations of this element were also recorded in roadside soils by other authors, for example, 0.03% (Cunningham et al. 2008), 0.007–0.035% (Kayama et al. 2003), <0.001% (Czerniewska-Kusza et al. 2004). Although low concentrations of TOC are typical for urban soils, the decreased contents of organic compounds in the soils studied are a concern. The presence of increased amounts of organic carbon facilitates proper soil structure and cation exchange and, moreover, is a source of nutrients for soil microorganisms. There is a positive correlation between TOC and N concentrations ($R^2 = 0.84$), as well as between TOC and S concentrations ($R^2 = 0.83$) in the soils examined, which provides evidence for the occurrence of S and N in organic form.

The concentrations of sulfur (0.01–0.17%) and zinc (40–471 mg/kg) in the soils of the Kielce area exceeded those (0.05% S and 50 mg/kg Zn) recorded in soils by Foth and Ellis (1996). In a majority of investigation sites these elements were accumulated in the surface soil layer (0–0.15 m).

The results of plant material analysis showed that some elements are accumulated in the tree bark (Ca, Fe) and others in the leaves (K, Mg, N, P, S, Zn). Low concentrations of iron in these leaves were similar to those found in edible plants (Kabata-Pendias and Pendias 2001). The concentrations of potassium in the tree bark did not differ from those recorded previously in the Holy Cross Mountains (Gałuszka 2005), but the levels of K in the leaves were higher than those reported from the Holy Cross Mountains (Migaszewski et al. 2004). No substantial differences between K concentrations in the samples from investigation and control sites were found.

The concentrations of magnesium showed interspecies differences. The lowest Mg concentra-

tions were found in the bark of linden (0.03–0.07%). For comparison, the typical contents of Mg in plants are in the 0.1–0.3% range (Kabata-Pendias and Pendias 2001). The bark exhibited a positive correlation between the Mg content and pH ($R^2 = 0.72$).

Of the elements determined in plant material samples, only the concentrations of sulfur and sodium were higher than those characteristic for plants. Sodium accumulated in most of the bark samples. The concentrations of sulfur in the tree bark (0.052–0.193%) and leaves (0.099–0.257%) were twice as high as typical contents of this element in trees growing in forest ecosystems of the Holy Cross Mountains (0.023–0.110%) (Migaszewski et al. 2004). Sulfur is an important constituent of polipeptides responsible for detoxification of phytotoxic metals in plants. Accumulation of sulfur in roadside trees may thus be linked to the enrichment of urban soils in metals. However, this hypothesis needs further studies. The maximum concentrations of sodium in the bark and leaves were 0.124% and 0.065%, respectively. These concentrations differ from those reported for tree leaves by Czerniewska-Kusza et al. (2004) (0.18–1.01%) or by Cekstere et al. (2008) who recorded maximum values of 1.93–2.1%. However, it should be stressed that Na concentrations in trees change seasonally (Cekstere et al. 2008) and the time of sampling may play an important role when comparing the results derived from different studies.

Elements transfer from soil to plants

Soil pH is a very important factor that influences bioavailability of elements for plants. The mean pH of the soil examined was 7.7, whereas the observed range was 6.6–8.3. The optimal soil pH value for plant nutrition is 6.5 and if it exceeds 7.5, the uptake of important elements, such as Zn, P, Mn, Cu, and Fe, is hampered (Adams and Early 2004).

The use of chloride deicers at the sampling sites resulted in increased soil alkalinity, which, in turn, influenced higher bioavailability of boron for plants. The contents of this trace element in the soils were low (4–23 mg/kg). In contrast, the

tree samples showed concentrations of B about three times as high as those in soils. In twelve samples of plant material concentrations of B were higher than the optimal level (30 mg/kg) for plants (Kabata-Pendias and Pendias 2001). There was a statistically significant positive correlation between the B concentrations and pH in the upper soil and tree bark ($R^2 = 0.81$).

The upper soil pH and concentrations of Ca, Mg, N and P in the tree bark displayed positive correlations between the following pairs: pH–Ca ($R^2 = 0.58$), pH–Mg ($R^2 = 0.63$), pH–N ($R^2 = 0.59$), pH–P ($R^2 = 0.62$). These relationships may be an indication of the influence of soil pH on translocation of elements within the tree organs.

A need for determining K/Na ratio in urban soils and plants has been recently mentioned (Cekstere et al. 2008). The K/Na and Ca/Na ratios are indicative for soil salinity (Porcelli et al. 1995). In the present study, these ratios were the highest at control sites (Fig. 2), which were not influenced directly by chloride road deicers. It is interesting to note that at investigation sites, in which the trees have shown the most intensive negative symptoms, the leaves displayed the lowest K/Na and Ca/Na ratios.

It is a known fact that roadside trees are especially prone to decline. There are many reasons for this vulnerability. Chloride deicers applied on the roads every year cause negative symptoms such as chlorosis and the loss of assim-

ilative apparatus. Other pollutants, for example, engine exhaust components are often phytotoxic. Besides, various building foundations, sidewalks, or roads restrict root growth and contribute to tree damages. The combination of different stress factors influencing roadside trees often leads to tree death.

The loss of assimilative apparatus and decreased vitality were the most intensive at sites 2a, 2b, 4a, and 4b. The trees growing at sites 1a, 1b, 3a, and 3b were the healthiest. A type of salt used for winter road maintenance does not seem to contribute to intensity of negative symptoms in the tree examined. In the winter season of 2008/2009 the roads at sites 1a, 1b, 2a, 2b were treated with sodium chloride, while the roads at sites 3a, 3b, 4a, and 4b with calcium chloride.

The most probable cause of negative symptoms is the road traffic intensity (the highest at sites 2a, 2b and 4a, 4b). Another factor is the distance of the tree from the road (the lowest was <0.5 m at site 4a). The recent study by Zehetner et al. (2009) indicated that the road salt residues (as well as PAHs and total and mobile heavy metals) decreased exponentially with distance from the road. Different studies on the influence of chemical deicers on roadside vegetation also provided evidence for the highest risk of toxicity as a result of direct deposition of salt on plant organs (Trahan and Peterson 2007; Viskari and Kärenlampi 2000).

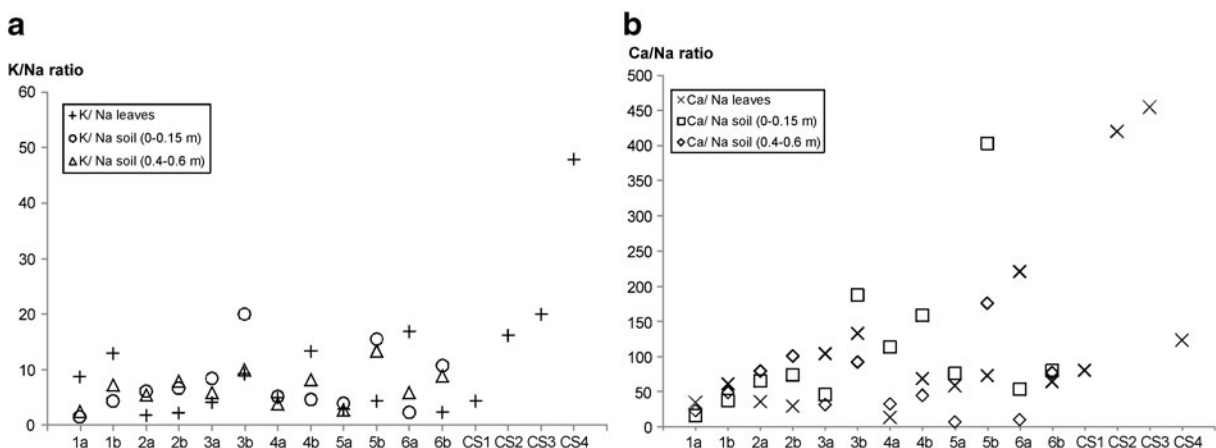


Fig. 2 K/Na and Ca/Na ratios in tree leaves and soils

Conclusions

Chemical analyses of snow collected in the vicinity of roads maintained with three different chloride deicers showed that the snowmelt might be an important source of alkalizing substances and chloride ions in soils. This process may bring about an increase of soil pH which may decrease bioavailability of important plant nutrients. The concentrations of S and Zn in the studied soils were higher and the concentrations of B, Mg, Na, and TOC were lower compared to the chemistry of typical soils. The alkaline soil pH brought about increased bioavailability of B and decreased bioavailability of Na and Zn. Negative symptoms revealed by the roadside trees, i.e., the loss of assimilative apparatus and decreased vitality, were most frequently observed at the sites with the highest traffic intensity and at the sites where trees grew closest to the road. The roadside trees decline is most probably caused by dissolved contaminants released to soil during snowmelt, but it also may be increased by a possible synergistic effect with winter temperature extremes.

The center of a city provides very difficult conditions for plant growth, and the seasonal systematic use of chemical deicers is just one of the possible causes of chemical stress for urban greenery. The influence of road deicers on the city environment is more complex than the direct influx of chemical substances to soils or waters. This influence affects biogeochemical processes in water–soil–plant systems with a serious consequence for plant nutrition.

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