Indicators of soil and snow mass exposed to deicing agents for improving the system of hygienic regulation and pollution control

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Abstract. The potential danger of long-term accumulation of salts and negative impact on the objects of the environment exposed to their influence, dictates the need to prepare the material that will be in demand for the implementation of measures to control the use of deicing materials (DIM). The following parameters were analyzed: electrical conductivity (EC); concentrations of Na⁺, Ca²⁺, Mg²⁺, Cl⁻ ions; heavy metals and semi-metals (HMS) in the soil and snow mass. The total soil contamination indicator (Zc) and the sodium adsorption ratio in the soil solution (SAR) were calculated, the results were statistically processed. Indicators and their critical values for the soil and snow mass are proposed in order to regulate the use of deicing reagents (DIR) based on easily soluble salts.

1. Introduction

Along with the use of classical methods for assessing the risk to human health associated with environmental pollution, in order to avoid possible negative effects, the principles of determining the threshold values of pollutants for conducting preventive measures are considered and remain relevant [1–3]. The impact associated with the active use of various qualitative and quantitative characteristics of deicing materials (DIM) in winter is added to the complex of factors that have a negative impact on a person. When they are used, the load on the soil, water, air and plants increases significantly [4–6]. Taking into account the long-term accumulation of salts, which are the main components of DIM, and the need to improve the principles of assessing their impact on the environment and public health, the purpose of the research was to prepare material based on experimental studies to substantiate a number of critical values in samples of snow mass and urban soil (anthropogenically transformed soil), which can be used to organize measures to control the use of DIM.

Due to its specific properties, the soil largely determines the conditions of human life in the city through the performance of its sanitary and recreational functions. Performing important environmental functions, the soil affects the chemical composition of groundwater and is a universal depositing medium. The sanitary and hygienic functions of the soil are very important, as it is a good antiseptic that destroys pathogenic microorganisms and decomposes organic residues, metabolic products of living organisms. Anthropogenic disturbances of the soil continuum lead to serious disturbances and...
degradation of the entire natural complex, which poses a threat to human health and life in the city. Urban soils that are urbanozems (Ur), whether they are man-made urbanozems (Uteh) or agrogenicurbanozems (Uagro) [7], experience a significant anthropogenic load, whereas the protective properties of these soils are significantly less pronounced (compared to soils of natural genesis) [8].

Long-term monitoring of urban soils provides detection of abnormal salinization associated with anthropogenic activities. One of the observed changes in connection with the use of DIM in winter is salinization of urban soils. Part of the salt mixture (up to 50% of the total amount of scattered chemicals), the main components of which are NaCl and CaCl₂, penetrates from roads with snow to adjacent soil areas. Na⁺, Ca²⁺ ions form the main fund of exchange cations. Moreover, in contrast to Ca²⁺ that is able to stabilize the soil absorption complex (SAC), Na⁺ ions in the SAC cause peptization of the sily part of the soil. Na⁺ is associated with the development of the soil alkalinity process, i.e. together with other ions, it is actively involved in the regulation of soil processes.

In this regard, it is necessary to improve the principles of environmental control, taking into account the environmental and public health hazards of DIR when using it. Urbanozems formed along highways are the depositing medium for many pollutants, including DIR [9–10]. Intensive human activity within large cities leads to significant and often irreversible changes [11–13]. Undoubtedly, any protective measures will prevent soil degradation, therefore, maintain a healthy mode of functioning of the environment and protect human health. The quality indices of soil that functions within the urban ecosystem remain the most common and informative indicators for their quantitative assessment. The total indicator should represent the total effect (physical, chemical and ecotoxicological) and include all indices reflecting the impact of various changes in soil properties [14]. According to the results of the research, a number of these soil indicators can be discussed as criteria for the ecological characteristics of an urban area under the conditions of DIM application. Along with the soil, snow can be a good diagnostic material for assessing the dynamics of salt intake into environmental objects during the winter due to the seasonal use of DIM.

2. Materials and methods
Samples of snow cover, as a transit material with which DIR are received [15], and soil samples – depositing material, where DIR get into in connection with the cleaning of road surfaces and their scattering, were taken from more than 20 check points in Moscow.

The concentration coefficient (equation 1) and the total soil contamination (equation 2) were calculated in accordance with the methodological recommendations [20]:

$$K_c = \frac{C_i}{C_{con}}$$

(1)

where:

$K_c$ – is the concentration coefficient i of the chemical element;

$C_i$ – is the concentration i of the chemical element in the sample;

$C_{con}$ – is the concentration i of the analysis and statistical processing of EC data [16], concentrations of Na⁺, Ca²⁺, Mg²⁺ [17], Cl⁻ [18] ions, as well as a number of heavy metals and metalloids (HMS) [19] in different extracts (mobile, potentially mobile, and total forms of elements) of the studied samples was performed chemical element in the control sample.

$$Zc = (\sum_{i=1}^{n} Kc) - (n - 1),$$

(2)

where:

$Zc$ – is the total soil contamination;

$n$ – is a number equal to the number of elements.

Equation (2) shows the concentration coefficients under the condition of $K_c > 1.5$.

The soil alkalinity processes are usually characterized by the sodium adsorption ratio (SAR) of the soil solution, which is calculated using data on the content of Na⁺, Ca²⁺, Mg²⁺ ions, taking into account their ratio. Here is the equation (3) for calculating SAR, mEq/L:
SAR = \frac{Na^+}{\sqrt{0.5(Ca^{2+} + Mg^{2+})}} \quad (3)

Physical and chemical methods aimed at determining the chemical composition and physical properties of the object are mandatory for complex multicomponent objects. However, it is not always clear which substances need to be determined. In this regard, there is a need to use integral methods – methods of bioassay. Using representatives of hydrobions in the complex of biotests, it becomes possible to control the pollution of soils and the adjacent water environment (rivers, snow, ground water, etc.). Ecotoxicological assessment of snow and soil samples selected from the same check points in Moscow was carried out by bioassay methods. The battery of test organisms used for analytical samples included 2 types of organisms: daphnia and infusoria (tetrachyrena) [21, 22].

3. Results of the study

3.1. List and threshold concentrations of the main components of DIM in snow mass samples

The obtained data enabled to prepare material for substantiating a set of indicators that are informative and selective for the present type of load. In the course of long-term studies of soil samples and snow mass exposed to the use of DIM in the conditions of a metropolis (Moscow), it was found that the results of the following easily changing indicators can be used as the main information indicators for monitoring: the content of chloride anion (Cl\(^-\)) and sodium cation (Na\(^+\)), electrical conductivity.

3.1.1. Sodium and chlorine ions. It was found that sodium (Na\(^+\)), calcium (Ca\(^{2+}\)) cations and chlorine anion (Cl\(^-\)) were most frequently detected in the snow cover, their content reached high values. For example, the content of sodium chloride and calcium along several highways was 2–3 g/L. It should be noted that the increase of the salt content in the samples was also confirmed by an increase of the EC indicator. The maximum values of the present indicator in the samples of the first selection (from 1070 to 8700 mcm/cm) coincided with the highest values for the concentration of Na\(^+\) (from 140.3 to 1261.8 mg/L), Cl\(^-\) (from 278.7 to 3965.3 mg/L) and Ca\(^{2+}\) (from 65.5 to 204.3 mg/L). The data analysis revealed a correlation between the concentration of Na\(^+\), Cl\(^-\) ions and EC in snow mass samples, as shown in table 1. Thus, the relationship of these parameters with the use of DIR is confirmed.

| Table 1. Correlation coefficients between variables. |
|-----------------|----------|----------|----------|
|                 | EC       | Na\(^+\)  | Cl\(^-\)  | Kc       |
| EC              | 1.00     | 0.84     | 0.98     | 0.81     |
| Na\(^+\)        | 0.84     | 1.00     | 0.75     | 0.67     |
| Cl\(^-\)        | 0.98     | 0.75     | 1.00     | 0.79     |
| Kc              | 0.81     | 0.67     | 0.79     | 1.00     |

The group of values (Group 1) with a high concentration of Na\(^+\), Cl\(^-\) ions and EC, presented in figures 1 and 2, was statistically determined and isolated from the entire array of analyzed snow mass samples.

First of all, the present high concentrations are due to the use of DIM with the main composite substance – sodium chloride (NaCl). In the group of high values, the average concentrations of Na\(^+\) and Cl\(^-\) ions correspond to the values of 1092.1 ± 269.4 mg/L and 2451.5 ± 267.7 mg/L. Given the fact that the analyzed data characterizing biological objects cannot always be described by a normal distribution, it is worth referring to the established median values. In this case, the concentration of sodium cation (Na\(^+\)) ≥ 2000 mg/L and the concentration of chlorine anion (Cl\(^-\)) ≥ 3000 mg/L can be attributed to the indication values. It is these values that were considered to determine the critical indicators for the content of Na\(^+\) and Cl\(^-\), confirming the effect of an external factor (in this case, DIM) on environmental changes.
3.1.2. The total soil contamination indicator. Taking into account the specifics of the use of DIM, their attachment to the highway, it is necessary to put in the indicator of total contamination (Zc). The present integral indicator will reflect the aggregating effect of DIM in conjunction with HM already present in the environment, the source of which is, first of all, motor transport. It is necessary to emphasize the high degree of contamination of roadside areas with HM and semimetals [1], which is confirmed by the studies. Moreover, to determine this indicator, it is necessary to take into account the content of HM not only in the filtrate, but also in the dry residue on the filter. According to this indicator, two samples of snow mass from the entire collection can be placed to the group with a hazardous level of pollution (Zc = 64–256). In samples contaminated with lead, the main amount of this element is found in the dry residue. It can be illustrated with two examples: the level of lead in one of the samples in the filtrate corresponds to 6.1 ± 4.4 mg/kg, and on the filter – 67.5 mg/kg; in the other sample, the level of lead corresponds to 1.5 ± 0.1, and its content on the filter – 33.1 mg/kg. About 15% of all the analyzed samples refer to the category with a hazardous level (Zc = 64–256) of contamination. Extremely hazardous level of contamination (Zc ≥ 256) is absent. The majority of the samples refer to the category of moderately polluted (Zc = 32–64) or uncontaminated.

3.1.3. Electrical conductivity. The integral indicator reflecting the level of salt concentration in the solution is the value of electrical conductivity. The indicator of electrical conductivity in snow mass samples and in soil is not standardized in Russia, but it is present in the documents of the European Union, where its limit value for soil is indicated – 2500 µS/cm. In addition, it is proved that meltwater affects the EC characteristics of the soil [4, 9]. The results of the analysis of the snow samples grouped in the upper cluster shows that their EC value varies from 4840 to 8700 µS/cm. The present data is presented in figure 2. Thus, using the previously described approach (allocation of threshold levels by the median value), it can be assumed that an EC value of 5 mS/cm and higher indicates an extremely high level of salt concentration in the solution.

It is important to compare the established criteria with the indicators of control samples taken from an uncontaminated area where the use of DIR is not provided (55° 19’ 66” N, 37° 87’ 17” E): the concentration of sodium and chlorine ions is 1.1 and 4.6 mg/kg, respectively; EC – 8.2 µS/cm; Zc – 7.0. Thus, the present values of the snow mass characteristic can be considered as critical for assessing the load of DIM in such a transit medium as soil.

3.1.4. Bioassay. The peculiarity of the information obtained with the help of bioassay methods is the integral nature of the reflection of all toxic effects in the studied objects. In our studies, toxicity todaphnia was detected in 50% of snow samples with an increased content of easily soluble salts. Testing using infusoria revealed hypotoxicity in about 33% of snow samples.
3.2. List and threshold concentrations of the main components of DIM in soil samples

In order to determine the irreversible changes occurring in the soil, the focus was placed on a block of chemical characteristics that indicate the ecotoxicity of the soil due to their increasing values. The increase in chemical pollution stipulates a decrease in the self-cleaning ability of the soil, an increase in its toxicity and a negative impact on the environment.

An abnormal increase in salt reserves causes an increase in the salt process. Salinization of most soils according to the predominant ionic composition belongs to the chloride-sodium type. Soil alkalization was recorded in comparison with the background soils. When assessing soil salinity, as a rule, anions (CO$_3^{2-}$, HCO$_3^-$, Cl, SO$_4^{2-}$) and cations (Ca$^{2+}$, Mg$^{2+}$, Na$^+$, K$^+$) of easily soluble salts are determined. The toxic effect of easily soluble salts is manifested in an increase in the osmotic pressure of soil moisture, a decrease in its availability to plants, a violation of the normal ratio of mineral nutrition elements, and a negative impact on soil properties [23]. Easily soluble salts in saline soils are found in the composition of the soil solution and solid phases of the soil (in the form of minerals and ions in the composition of the SAC).

NaCl is the main salt that causes salinization in our studies. The accompanying cations Ca$^{2+}$, Mg$^{2+}$, which can also lead to salinization, were not considered indicative due to their relatively low concentration (in relation to Na$^+$). NaCl is one of the most common and toxic substances in saline soils due to its physiological activity and high solubility (264 g/L). Sodium carbonates are the most toxic to plants, followed by chlorides. A mixture of different salts is always more harmless than the predominance of a single salt.

The generalization of analytical data made it possible to determine the indicative values: the content of separate ions (mg/L), the content of exchangeable sodium mol(+)/100 g of soil, the electrical conductivity of filtrates (1:5) (EC) (mS/cm), the sodium adsorption ratio of the soil solution (SAR units), the total soil contamination indicator (Zc).

3.2.1. Exchangeable sodium (mol(+)/100 g of soil). The category of salt-affected soils includes soils containing easily soluble salts or their ions in at least one horizon of the soil profile in quantities exceeding the toxicity threshold – the maximum permissible amount of salts that does not cause plant depression [24].

With the use of DIM, as shown by chemical and analytical studies, together with the snow mass, a large amount of salts enters the soil surface. The content of sodium chloride in the upper soil horizon from which the soil samples were taken correlates (R = 0.80) with the values of their content in the snow mass samples.

Processing of the results on the content of sodium ions in soil samples made it possible to identify an analytical group characterized by high values, obviously due to the use of DIR. Figure 3 represents the sodium content for 4 groups identified by the results of cluster analysis. The average sodium ion content equals 1084 ± 346 mg/L (control sample – 101.5 ± 50.3 mg/kg), which is represented in table 2. In order to determine the indicator value, the median of 1000 mg/L is used. The threshold indicator level can be slightly adjusted upwards (using the upper quartile value of 1835 mg/L), taking into account that the sampling was carried out from the upper horizon in early spring (leaching to the lower horizons is possible due to the washing regime of the present climatic zone).

Thus, the value of 8.0 mmol (+)/100 g of soil can be considered the threshold criterion for the content of exchangeable sodium in terms of mmol (+)/100 g.

3.2.2. Electrical conductivity of the filtrates. Electrical conductivity (EC) or its inversely proportional value – electrical resistance (ER) – are constant soil characteristics: EC = 1/ ER. In Russia and abroad, potentiometric methods for determining EC are widely used – they are used as the main methods for determining the territories of salt-affected soils [25–27].

When comparing the results by electrical conductivity (EC) of soil samples with the EC of snow mass samples previously taken from the same sites (in winter), which is represented in the table 3, a correlation (78%) was established.
Table 2. Results of Na\(^+\) ion content in the soil and snow mass samples by clusters.

| Ranking by groups | Average | St. dev. | Coef. of var. | 25% | Median | 75% |
|-------------------|---------|----------|--------------|-----|--------|-----|
| 1                 | 3232.8  | 940.9    | 29.1         | 2615.5 | 2920.0 | 3850.0 |
| 2                 | 669.5   | 302.5    | 45.2         | 408.5 | 659.5  | 930.5 |
| 3                 | 183.4   | 81.2     | 44.3         | 105.0 | 216.0  | 255.0 |
| 4                 | 28.3    | 2.1      | 15.0         | 6.0  | 19.0   | 29.0 |

Figure 3. Span diagram for Na\(^+\) ions in soil samples.

Figure 4. Span diagram for specific electrical conductivity values in soil samples.

Figure 4 shows the main clusters identified by the analysis using statistical methods, which confirm, as well as by other indicators, the presence of a group of samples with high EC characteristics.

The EC value in the analytic samples ranges from ~ 0.1 to 0.5 mS/cm. The EC value of the control sample corresponds to 0.083 mS/cm (~ 0.1). Guided by the existing normative geoeconomic indicators of soils, the electrical conductivity indicator < 2 dSm/m characterizes unsalted soils. The electrical conductivity indicator in the range of 2–4 dSm/m represents very slightly saline ones. In this case, the growth of a number of plant species that are sensitive to salinity (bulbous, umbrella, fruit trees, etc.) is inhibited [28]. Ion concentrations, accompanied by an increase in electrical conductivity from 4–8 dSm/m, characterize highly saline soils and stipulate the death of salinity-sensitive species, inhibition of growth and a decrease in the productivity of most plants up to 50%, as well as adverse changes in the physical and chemical properties of the soil. However, this gradation is designed for analytical studies of soil pastes, which is not comparable to soil water extracts in the ratio of 1:5. Taking into account that EC that corresponds to values from 2 to 8 mS/cm was established earlier in the snow mass samples taken from sites with a high DIM load, and the EC in soil samples taken from the same territories is in the range of 0.420–0.605 mS/cm, it can be assumed that the critical EC level, reflecting the use of DIM in urban soils, is within these limits. In order to clarify the starting point or initial EC values indicating a high concentration of NaCl in the medium (in the soil, previously established according to ecotoxicological parameters), an experiment was modeled to determine the dependence "NaCl concentration – EC value", which is presented in figure 5.
Table 3. Values of electrical conductivity in the soil and snow mass samples by clusters.

| Ranking by groups | Average | St. dev. | Coef. ofvar. | 25% | Median | 75% |
|-------------------|---------|----------|-------------|-----|--------|-----|
| Snow mass samples (n = 20) |         |          |             |     |        |     |
| 1                 | 5967.5  | 1834.0   | 65.0        | 2902.0 | 4984.0 | 8350.0 |
| 2                 | 1463.3  | 519.1    | 28.1        | 553.0 | 604.0 | 1641.0 |
| 3                 | 551.0   | 92.3     | 53.0        | 144.0 | 382.0 | 658.0 |
| 4                 | 108.3   | 37.5     | 18.6        | 28.0 | 34.5 | 161.0 |
| Soil samples (n = 20) |         |          |             |     |        |     |
| 1                 | 504.4   | 73.5     | 14.8        | 446.0 | 460.0 | 596.0 |
| 2                 | 415.3   | 31.0     | 7.8         | 371.0 | 384.0 | 480.0 |
| 3                 | 294.4   | 48.4     | 23.5        | 213.0 | 287.0 | 398.0 |
| 4                 | 145.3   | 40.2     | 27.7        | 103.0 | 150.0 | 283.0 |

Given that the median concentration of sodium ions is 1000 mg/L or 1 g/L, the EC indicator (the formula is shown in the graph: \( y = 1827x + 82 \)) is 1909 µS/cm ~ 2000 µS/cm or 2 mS/cm. It is this level of EC – 2 mS/cm – that can be considered critical for urbanized soils due to the use of DIR. According to the results of the analyses, such values are not established. However, the value that was set for the upper quartile – 0.56 mS/cm – acts as a predictor of anthropogenic load to urban soils.

Figure 5. Relationship between the EC value and the NaCl concentration.

3.2.3. The sodium adsorption ratio (SAR) in the soil solution. The correlation between EC and Na\(^+\) in snow mass samples, shown in table 1, is revealed during data analysis and confirmed statistically, whereas in soil samples it is not so obvious and depends on a number of factors. The soil factors affecting the EC characteristics are formed taking into account the soil functions, which reflect, first of all, the complexity of the anthropogenic impact in these samples. The load from the use of DIM is not prevailing, it is cumulative and reflects the total load formed by the presence of HM and the concentration of DIM ions. Thus, it seems very difficult to distinguish the action of a single factor (for example, DIM) in soil samples, so in order to determine the cumulative action, it is necessary to refer to
the characteristic that reflects the process of soil alkalinity – to the value of SAR (sodium adsorption ratio).

In most cases, the SAR value in the studied soil samples does not exceed 1, this indicates the absence of soil alkalinity processes. The present value was equal to 1.5 in several territories (№ 1, 9, 11, 19) and exceeded this indicator in only three cases and was equal to 3.9 and 4.3. The given values of SAR indicate a low risk of irreversible processes of soil alkalinity. This suggests that the primary process in the studied areas is soil salinization, and the processes of soil alkalinity are less pronounced. When determining the functional geochemical and sanitary changes in urban soils caused by the combined action of DIM, it is advisable to use an informative indicator of the sodium adsorption ratio (SAR) – the coefficient of potential sodium absorption. The hazard of salinization of soils in the natural environment is determined not only by mineralization, but also depends on a number of other factors – the properties of soils and subsoils, climatic indicators. Given the high mineralisation of precipitations in winter, combined with the leaching regime of soils in the spring and autumn season, it is recommended to take into account 8–10 SAR limit values.

3.2.4. Total soil contamination indicator (Zc). The total value of HM accumulation in the soil was determined according to the results of chemical analysis, followed by the calculation of the ratio of the concentration of chemical elements (Ki) and the pollution index (Zc) obtained when comparing the concentration of HMS in the soil with their background content that indirectly takes into account polyelemental pollution. As a background, the indicators of regional soils of this type, located outside the area of anthropogenic pollution were used. Thus, the basic requirements for the comparison sample were implemented: the same set of defined elements and the same methods for their determination. In order to analyze the results of the content of HM, the sanitary and hygienic criteria, i.e. MPC and APC for soils, taking into account the forms available in the standards (total, mobile) were also used. When comparing the obtained results of the analysis on the content of HM in the studied samples with the MPC (total forms), there are excess standards for some elements in different areas. The excess of Zn by almost 100% (Kzn = 0.88) and Cd by almost 200% (Kcd = 1.80), the excess of Zn by almost 200% (Kzn = 1.95) and the excess of Pb by more than 100% (Kpb = 1.23) were found in individual samples. In other samples, the excess of As was found by 80–140% (Kas= 0.81; 0.88; 1.4). The total content of HM and the content of potentially mobile forms are within the normal range for the remaining elements and samples. Based on the analysis of the composition of the elements of pollutants, Zinc is the leading one, then the effect of lead and copper is manifested; the excess of cadmium is not so significant; As and Sr polluted several territories selectively. The hazard criteria for the Zc index are as follows: low (non-hazardous) < 16; medium (moderate-hazardous) 16–32; high (hazardous) 32–64; very high 64–128; extremely hazardous > 128. The results of the analysis of the pollution index (Zc), which takes into account only those elements whose concentration coefficient is >1.5, indicate a predominantly low level of hazard. Only in individual samples a very high level of pollution and a high level of pollution are noted. It should be emphasized that the very high and high levels of pollution are obviously not connected with the application of DIM, but are due to the proximity to the highway.

3.2.5. Bioassay. The conducted soil bioassay showed that hypotoxicity on the part of the generative function of tetrachymenes was detected only for one soil sample. The remaining samples did not have a negative effect on the test organisms and even had some stimulating effect, probably due to the presence of organic substances in the soil. Toxicity of soil samples using daphnia as a test organism was not detected.

4. Conclusion

Based on the results of long-term studies of the impact of deicing materials on the environmental objects, indicators and their critical values were established and presented in Table 4. They will help determining the optimal conditions for the use of reagents, as well as contribute to limiting their irrational use in urbanized areas.
Table 4. Summary data of indicative parameters for assessing the negative impact of DIM on environmental objects.

| Parameter                                           | Research object | Indicative value | Critical value |
|-----------------------------------------------------|-----------------|------------------|----------------|
| electrical conductivity (EC), mS/cm                 | Snow mass       | 5                | 2              |
| Concentration of Na\(^+\) ions, mg/L                | snow mass       | 2000             |                |
| Content of exchangeable Na\(^+\), mmol(+)/100 g     | soil            | 8.0              |                |
| Sodium adsorption ratio (SAR)                        | snow mass, soil | low (non-hazardous) < 16; medium (moderate-hazardous) 16–32; high (hazardous) 32–64; very high 64–128; extremely hazardous > 128 |
| Total soil contamination indicator (Zc)              | snow mass, soil | 8–10             |                |

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