water

Article

The Relationship among Precipitation, Application of Salt in Winter Road Maintenance and the Quality of Waterways and Soil around Motorway

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Abstract: Construction of motorways and road traffic bring a new line element into the landscape, which fundamentally influences the local hydrological regime and water quality. Impermeable surfaces are introduced into the landscape, affecting the natural infiltration of water in the environment. The aim of the present research was to assess the long-term historical development of changes in the landscape retention potential due to construction of the D1 motorway in the Jihlava region and also evaluate the impact of winter maintenance on the quality of surface waters and soils. During the reference period, the research area showed an increase in land take and related increases in soil sealing by impermeable surfaces due to the construction of the D1 motorway. This fact has clearly contributed to the deterioration of the retention potential of the landscape in the area. Chloride concentrations of both matrices were evaluated in relation to the amount of de-icing salt applied at the sites and precipitation recorded in this region. Water samples collected at the outflow contained in all cases higher concentrations of chloride than samples of the inflow water. The highest chloride concentration in soil was measured in samples collected at a distance of 2 m from the road.

Keywords: land use; soil; water; chloride; contamination; winter maintenance; precipitation

1. Introduction

Transport infrastructure and traffic associated with it are an important factor that has significant impact on the landscape, its individual components and the environment. The primary effects based on the traffic itself are air pollution and environmental (water, soil) contamination; long-term anthropogenic changes in the use of the landscape caused in particular by the availability and development of settlements can be considered a secondary effect [1,2].

Changes in the use of land also have a significant impact on the retention potential of the landscape. The models currently used often combine land cover mapping with the application of runoff curve numbers [3,4]. However, the evaluation of long-term changes depends on the possibility of detailed mapping of the habitats forming the land cover [5]. Such evaluation has only become possible in the post-WW2 era by applying modern remote sensing methods using aerial images; nowadays, such methods are used frequently [6–9]. For evaluation of historically older periods, the land cover mapping often uses historical topographic maps [10,11], but these maps do not provide sufficient detail to make it possible to evaluate the retention potential of the landscape. In the context of road infrastructure, the retention potential of the landscape is changing mainly due to land take and soil sealing resulting from the road construction and maintenance.

In winter, the areas along roads are loaded with de-icing agents due to winter maintenance of roads performed with the aim to ensuring the traffic safety. Since the late 1940s, sodium chloride (NaCl) has been the most commonly used de-icing agent due to its low cost, ease of use and storage and high efficiency (up to −8 °C). In the Czech Republic, during the 2017/2018 winter season, de-icing salt was applied to 369.4 thousand kilometers of motorways, where 24.6 thousand tonnes of salt and 10.1 million liters of brine were used [12]. The primary contaminant of de-icing salts is the chlorine ion Cl^- (chloride), which passes through the aquatic and soil environments surrounding the roads without significant changes in terms of chemical reactions, physical bonds and biological uses. It is easily soluble in water and highly mobile [13,14]. Chloride ions are transported from roads to adjacent soils and surface and groundwater. Therefore, the increased content of chloride ions has a negative impact on ecosystems near the road network. The level of impacts of chlorides on the environment depends on various factors such as the rate and frequency of the application of de-icing salt, the amount of de-icing salt applied, the frequency of rainfall (precipitation), the distance from the site of application and the type of matrix monitored. The extent of contamination is proportional to the amount of de-icing salt used. The impact of the negative effects of de-icing salt on ecosystems is often specific to a given locality [15,16]. Long-term use of de-icing salt also causes problems related to the corrosion of cars and specific parts of road infrastructure (metal structural road elements, steel reinforcement rods, concrete) and accelerates road condition deterioration due to repetitive cycles of freezing and melting on road surface [17,18].

Approximately 60% of the salt applied to paved surfaces is removed each year from the relevant catchment area by surface runoff, the remainder (approx. 40%) accumulates in shallow groundwater and can contribute to reaching equilibrium concentrations. This leads to a gradual long-term increase in concentrations in shallow aquifers [19]. The contamination is most evident during the spring months when snow begins to melt and the high concentrations of chlorides from de-icing salt are washed away with the melting snow into the surface layers of soils and surface water [20]. Furthermore, chlorides permeating to the soil can reach groundwater in several ways depending, for example, on the frequency of precipitation, melting rate and the type and characteristics of the soil around the road [21]. Hydro-chemical research suggests that the amount of NaCl in groundwater increases every spring. The increase is caused by the melting snow water mixing with chlorides, which are washed from the road and seep into the soil and pass into groundwater [22]. The impact of the use of de-icing salt on surface water is not as pronounced as its impact on groundwater. Due to turbulent and rapid flows of surface waters, chemical materials are mixed and diluted almost immediately after entering the watercourse. Groundwater is more sensitive to chloride pollution because there are no turbulent factors that would contribute to diluting of contaminated water [23]. Moreover, since groundwater is often used as drinking water, its contamination with chlorides is not desired; it is therefore advisable to regularly monitor the quality of groundwater in certain areas, not only in winter during the period of application of de-icing salt.

Another negative impact of chlorides from de-icing agents is the increasing salinity of soils and, consequentially, groundwater [24]. Sodium contained in de-icing salt is an undesirable element for vegetation. It causes the displacement of other cations in the soil, such as calcium, magnesium and potassium and makes these cations unavailable for plant intake [14]. High sodium content in the soil disperses organic and inorganic particles that are present in the pores of the soil. This results in reduced soil permeability, reduced aeration and increased surface flow, surface runoff and soil erosion. Increased erosion then leads to the transport of nutrients and heavy metals from roads to surface waters [25,26]. Although chlorine in low concentrations is an important nutrient in terms of soil fertility, its excess in the environment can disrupt the use of agricultural land by reducing its fertility [27]. The chloride anion together with the potassium cation have an important function in the regulation of osmotic conditions in the cell. Chlorine is also involved in the photolysis of water during photosynthesis. In plants, it accumulates in assimilatory organs, i.e., in leaves and needles [28]. The high proportion of salts in the soil affects its physicochemical properties and ongoing biogeochemical soil cycles. The runoff washed from a road contains trace metals such as Cd and Pb [29] and chlorides contained in the runoff have a strong ability to increase the mobility of these trace metals [30]. The mobility of chlorides in soils is so high that chloride can be used essentially as an indicator to assess soil water movements or aquifer saturation [31]. Chloride concentrations also affect pH level of soil. Land in the areas with higher traffic volumes has been showing higher pH values, particularly during the winter period [32].

The aim of the present research was to assess the long-term historical development of changes in the landscape retention potential due to construction of the D1 motorway in the Jihlava region. The impact of winter maintenance on the quality of surface waters and soils was also evaluated in this area due to the nearby specially protected areas within the Natura 2000 network and more frequent application of de-icing salts because of the higher altitude of the assessed area.

2. Materials and Methods

For the research work, two sites (Site A, Site B) near the D1 motorway, which differed in character of the location, were selected; see Figure 1. Detailed characteristics of the sites are given in the chapter below.

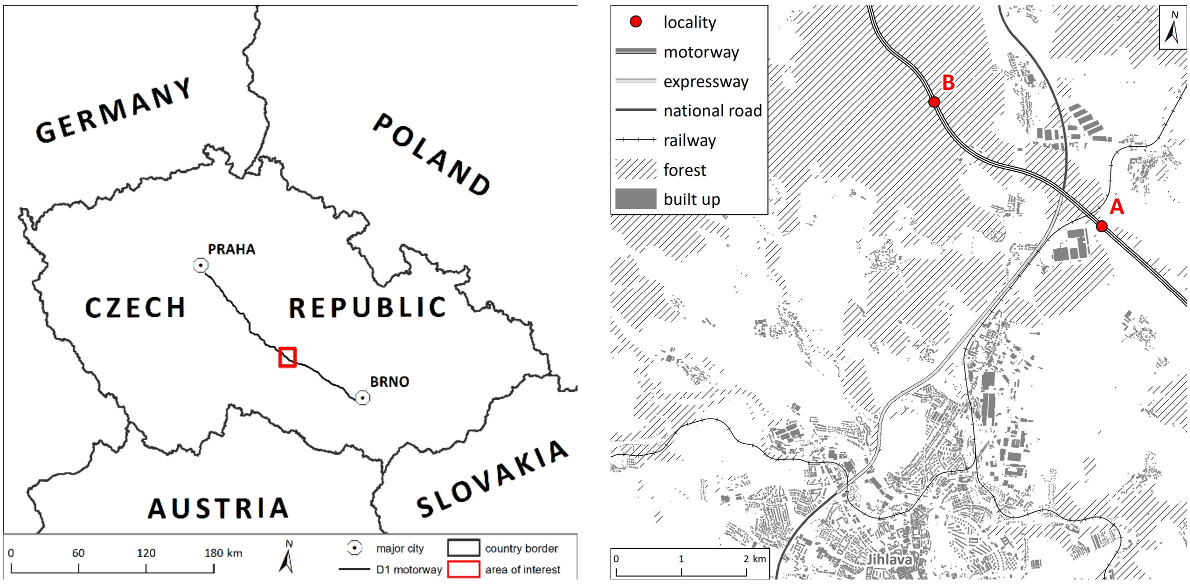

Figure 1. The Jihlava region with marked Sites A and B.

2.1. *Analysis of Long-Term Changes in the Retention Potential of the Landscape in the Selected Locations*

Changes in the landscape retention potential in the selected locations were assessed over the period from 1953 to 2018. Construction of the D1 motorway passing through both these locations took place between 1976 and 1980. When the construction sections 013 Humpolec–Pávov and 014A Pávov–Řehořov were opened, it marked the completion of the motorway connection between the three largest cities in the former Czechoslovakia (Prague, Brno and Bratislava) in its entirety [33].

The Curve Number (CN) method for each category of land cover and the hydrological soil group were used to evaluate the long-term development of land cover and its retention potential within Sites A and B. These are the two most important factors that affect the landscape's ability to retain water in the long term. The hydrologic soil groups (HSG) include soils with similar physical parameters, surface runoff parameters and the rate of infiltration. The groups are divided based on several parameters, including the depth of the impermeable layer, the depth of the ground water level and the saturated hydraulic conductivity (Ksat) of the soil without the land cover. Map data [34] were used to divide the land of the monitored locations into individual hydrologic soil groups, see Table 1.

Table 1. Description of hydrologic soil groups [34].

Hydrological Soil Group	Soil Properties
A	Soils with high infiltration rates (>0.12 mm/min) even at complete saturation, including predominantly deep, well-drained to over-drained sands or gravels
B	Soils with moderate infiltration rates (0.06–0.12 mm/min) even at complete saturation, including mainly moderately deep to deep, moderately well drained to well drained soils, e.g., soils with loamy sand to clay loam texture.
C	Soils with low infiltration rates (0.02–0.06 mm/min) even at complete saturation, including mainly soils with a layer impeding permeation contained in the soil profile, e.g., soils with clay loam to clay texture.
D	Soils with very low infiltration rates (<0.02 mm/min) even at full saturation, including mainly clays with high swelling, soils with persistently high groundwater levels, soils with a layer of clay on or just below the surface and shallow soils above almost impermeable subsoil.

The proposed categorization of the land cover and the determination of CN values of individual elements in the landscape referred to in Table 2 is taken from publications [35,36] and adapted for the local conditions of both monitored sites.

Table 2. Description of hydrologic soil groups (HSG) [34].

LC Category	CN [%]			
	A	B	C	D
Arable land	72	81	88	91
Fruit orchard with grassing	43	65	76	82
Permanent grassland	49	69	79	84
Clearing, windfall	68	79	86	89
Shrub, bush	35	56	70	77
Forest	36	60	73	79
Unpaved road	76	85	89	91
Semi-paved road	83	89	92	93
Railway incl. railbed	59	74	82	86
Other	59	74	82	86
Impermeable surface	98	98	98	98
Water surface	0	0	0	0

The mapping of the historical development of land cover according to the categories listed in the above table was carried out in the ESRI ArcGIS™ software (ESRI, Redlands, CA, USA) for five time horizons based on orthophotomaps and aerial images (Table 3) in the buffer area of 500 m from the center of the motorway at the sampling points (size of the mapped area was 78.5 ha for each monitored location and time period). Individual years were selected from the available data (Table 3) in order to represent the original state of the landscape (1953), the state from just before the start of construction of the motorway with the influence of preparatory works such as creating forest clearings for the planned road (1975), the state after completion of the construction and commissioning of the motorway (1983), the state before the process of complex land modifications and the completion of the first stage of the development of the Bosch industrial site (2001) and the current state of the landscape (2018).

Table 3. Map documents used for land cover mapping.

Map Type	Year	Provider
Historic black-and-white orthophotomap	1953	Czech Environmental Information Agency (CENIA)
Aerial black-and-white image	1975	Military Geographic and Hydrometeorologic Office (VGHMÚř) Dobruška
Aerial black-and-white image	1983	Military Geographic and Hydrometeorologic Office (VGHMÚř) Dobruška
Archive black-and-white orthophotomap	2001	Czech Office for Surveying, Mapping and the Land Registry
Color orthophotomap	2018	Czech Office for Surveying, Mapping and the Land Registry

As the resulting indicator of the hydrological state of the landscape, the spatial analysis in the GIS was used to determine the average CN for each of the monitored sites and the time frame analyzed.

2.2. *Description of Sampling Sites A and B*

Throughout the entire period of operation of the D1 motorway, its continuous traffic in the winter period was ensured by the application of de-icing salt. Therefore, the research was focused on determining the annual increase (November 2017–October 2018) in chloride load in water and soil around the road. The measurements were carried out at the two selected locations (Sites A and B) on the D1 motorway, in the direction from Brno to Prague, which were approximately three kilometers apart. The two sites were selected mainly based on the criteria of the different nature of the area in the context of changes in the landscape retention potential, more frequent application of de-icing salts due to the higher altitude of the area, the traffic intensity, the adjacent watercourses and the nearby specially protected areas within the Natura 2000 network.

2.2.1. Site A—At 113 km of D1 Motorway in the Direction of Prague

Apart from the above reasons, the site was selected also due to the completion of the modernization of the relevant motorway section between intersections Velký Beranov and Jihlava. The modernization had started in August 2016 and was completed in September 2017, i.e., before the soil sampling was commenced. During the modernization, the original soil was excavated from the base of the road to a distance of approximately 10 m from the road, and it was replaced by new soil. It is a straight section of the motorway without a forest cover before the exit to Jihlava (112 km) with the traffic intensity of 39,200 vehicles per 24 h. In the selected site, the road is raised against the surrounding terrain which slowly declines and changes into a grassy field. This transect was used for taking soil samples at five different distances from the road. Approximately 200 m from the soil collection point, Zlatý potok stream flows under the body of the motorway; water samples used for the study were obtained from this stream. The source of the stream is in a forest area near the highway approximately 2.5 km by air from the sampling point towards the south-west. It flows through several ponds, around an industrial site, under the motorway exit, through the body of the D1 motorway, and then, it flows into the Šlapanky river, which belongs to the Vltava river basin. A part of the Zlatý potok stream, starting from the body of the D1 motorway further downstream belongs to the system of protected areas within the Nautra 2000 network–Sites of Community Importance: "Šlapanka a Zlatý potok" (CZ0613332). It is an important and high-value site of permanent occurrence of European otter in the Vysočina region. In view of this fact, it is necessary to preserve this site in its natural state, including the riparian vegetation. The negative impact of the D1 motorway on this site is considered not very serious, as the boundary of the relevant protected area only intersects the D1 motorway in its southern part in a section of about 100 m (Figure 2).

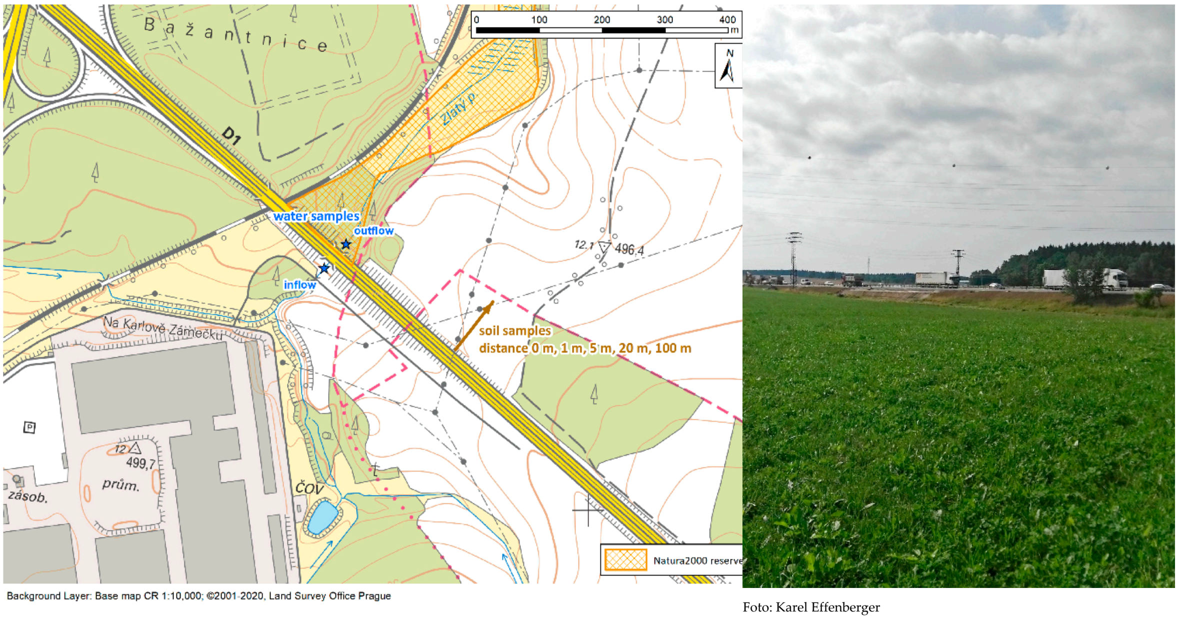

Figure 2. Situation map and photos of Site A.

2.2.2. Site B—At 110 km of D1 Motorway in the Direction of Prague

The sampling point was located in a slight climb of the motorway in a wooded area two km behind the exit for Jihlava (at 112 km), with a traffic intensity of 39,439 vehicles per 24 h. At this place again, the road is raised due to the surrounding terrain with the embankment of the D1 motorway steeply descending into a wooded area. In this transect, soil samples at five different distances from the road were taken. Water samples for this site were taken from a nameless stream located approximately 100 m from the soil collection point towards the exit for Jihlava. The stream springs in the forests near the D1 motorway and flows from the source only through the forest. The whole area is part of the Natura 2000 network–Sites of Community Importance: "Vysoký kámen u Smrčné" (CZ0610003). It is one of the few larger and relatively well-preserved flowering beechwood areas and Tilio-Acerion forests in the Bohemian-Moravian Highlands. The territory has a considerable potential for spontaneous restoration of the nature of the nearby deciduous forest. The D1 motorway poses a certain threat to the site (Figure 3).

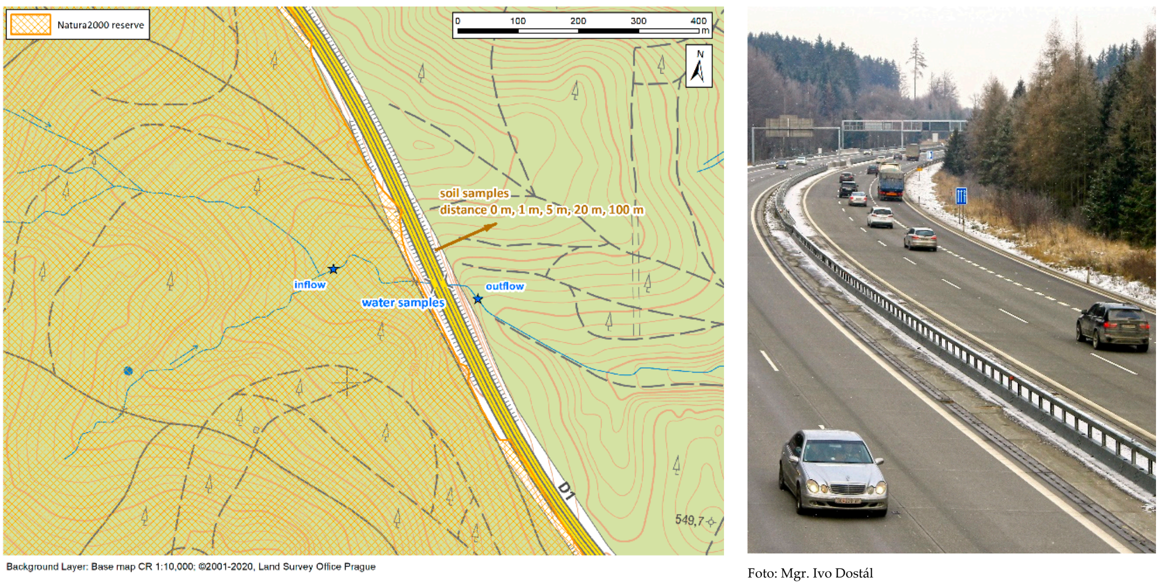

Figure 3. Situation map and photos of Site B.

2.3. Collection and Laboratory Processing of Soil and Surface Water Samples

Soil and water samples from both sites were taken over a period of thirteen months between October 2017 and October 2018, at regular monthly intervals. The soil collection was carried out at a depth of 30 cm below the surface at five distances from the road—i.e., at the distance of 0, 1, 5, 20 and 100 m. During the sampling within the given distance, 6 to 8 soil samples were first taken from which a modified composite soil sample of approximately 1 kg was obtained by quartering method. Surface water samples were taken at each site before and after the stream intersects the motorway (inflow/outflow), always as point samples collected to dark glass sample containers in the volume of 1 liter. Soils and water samples were placed in a cooling box immediately after the collection and transported to the laboratory without further delay.

For the water samples obtained, pH and conductivity values were determined immediately after delivery to the laboratory using laboratory pH/Orion 4 Star conductometer (Thermo Scientific, Waltham, MA, USA) according to ČSN EN ISO 10523:2010 [37]. The procedure is intended for determining the pH value in samples of leachates from roads, aqueous extracts and surface waters in the range of values from pH 2 to pH 12, in the temperature range 0 °C to 50 °C. The chlorine ions were determined by spectrophotometry using Spectroquant Prove 300 (Merck KGaA, Darmstadt, Germany) with selective test Spectroquant® - Test for chloride determination by photometry, measuring range 2,5–250 mg/L chlorine ions (Merck KGaA, Darmstadt, Germany). The quality of the analytical results was checked via frequent analyses of reference material (RM ION-96.4 A natural river water with certificate value of Chlorides).

The soil samples were first dried at room temperature and then sieved through a sieve with mesh size of 2 mm. Subsequently, an aqueous leachate was prepared from the modified samples according to the procedure for extracting soils with highly purified demineralized water in the liquid and solid-phase ratio of 10 l per 1 kg of soil [38]. A plastic container of 500 ml volume was filled with 25.0 g of dry sieved soil sample and 250 ml of ultrapure water. The containers were put into a Reax 20 rotary shaker (Heidolph, Schwabach, Germany) and left shaking for 24 h at laboratory temperature and a rotation speed of 5 rpm. After the extraction, the samples were set aside for 20 min before being further processed. Soil samples extracted with water were centrifuged (Universal 320 R benchtop centrifuge) for 20 min at the rotation speed of 4000 rpm. If needed, the samples were further filtered through 0.45 µm membrane filter. The samples thus prepared were used for the spectrophotometric analysis of chlorides using Spectroquant Prove 300 with selective test Spectroquant®. The measured concentrations in aqueous soil leachates were converted to corresponding concentrations in one kilogram of soil collected.

2.4. Quantity of Applied De-Icing Salt and Precipitation at Sites A and B

For the period October 2017–October 2018, information on precipitation was also obtained from the nearest measuring station of ČHMÚ *(Czech Hydrometeorological Institute)* in Přibyslav, which is located approximately 13 km away from the sampling location. Furthermore, the data on the consumption of de-icing agent for the period from November 2017 to March 2018 obtained from the relevant D1 maintenance center were taken into account. Data on the total amount of de-icing salt applied to the 47.6-km section of the motorway to which the two monitored sites belong have been converted into the amount of de-icing salt used per 1 km of motorway.

Data on the amount of de-icing salt applied and precipitation are identical for both monitored sites. From Figure 4, it is apparent that the highest amount of precipitation in the monitored area was recorded in October 2017 (93 mm/month). Moreover, precipitation fluctuated between 15 and 40 mm/month in the period from November 2017 to April 2018. Since May 2018, the amount of precipitation has increased on average to 50 mm/month, with the driest month being July with 18.5 mm/month.

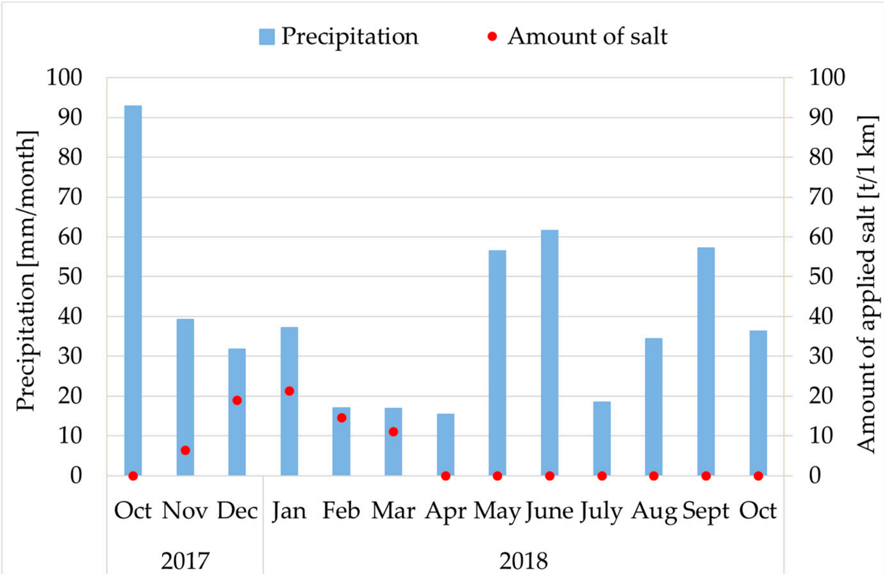

Figure 4. Amount of precipitation and applied de-icing salt during the sampling period for both sites.

Every year, the application of de-icing salt is carried out according to winter road maintenance plans specific for the relevant area of the motorway. Within Sites A and B, winter maintenance of the road begins every November of the relevant year and ends at the beginning of April of the following year. In the reference period (October 2017–October 2018), based on the weather conditions, the highest amount of de-icing salt was applied in the month of January (21.3 t/km of motorway), followed by December and February (19 and 21.3 t/km of motorway, respectively), i.e., during periods of low temperatures with the highest incidence of freezing rainfall and snowfall. Figure 4 shows an overall trend of increasing application of de-icing salt from the beginning of the winter period (November, December and January, 6.4, 19 and 21.3 t/km of motorway, respectively) and a decreasing trend towards the end of winter (February and March, 14.5 and 11.1 t/km of motorway, respectively).

2.5. Statistic Methods

Multidimensional analysis module of the QC.Expert 3.3 statistical software package (TriloByte Statistical Software, Czech Republic) was used for statistical data processing [39]. Pearson correlation [40] was used to compare the interrelationships of the measured parameters (chloride concentrations at different sites, distances and periods; conductivity at different sites, distances and periods). Pairwise comparisons were used to assess the identity of the data sets of chloride concentrations ad different sites. Principal component analysis was used for graphical representation of the relationships among chloride concentrations in soils and waters, the amount of salt used and rainfall.

3. Results and Discussion

3.1. Historical Development of the Retention Potential of Landscape at Sites A and B

The retention potential of the landscape was assessed on the basis of two parameters. First by monitoring the historical development of land cover and, second, using the hydrological characteristics of soil cover at both monitored sites. The results of the analysis of the long-term development of the land cover of both sites are presented in Table 4 and show the completely different characteristics of the landscape at both sites and the different development and changes in the individual monitored periods.

Table 4. Land cover categories and their proportions [%].

	Site A					Site B				
Category	**1953**	**1975**	**1983**	**2001**	**2018**	**1953**	**1975**	**1983**	**2001**	**2018**
Arable land	50.71	55.63	52.67	41.31	36.39	-	-	-	-	-
Fruit orchard with grassing	0.74	0.13	0.13	-	-	-	-	-	-	-
Permanent grassland	20.40	11.31	11.01	8.41	11.05	-	-	3.32	1.05	1.91
Clearing, windfall	-	2.40	0.45	2.44	-	7.36	9.89	5.04	1.33	14.49
Shrub, bush	0.75	3.19	3.42	5.40	2.64	-	-	-	1.76	4.89
Forest	25.73	25.71	25.09	30.07	33.99	90.90	88.33	86.38	90.65	73.81
Unpaved road	0.42	0.60	1.37	1.63	0.65	1.74	1.78	1.69	1.64	1.30
Semi-paved road	0.37	0.12	0.12	0.08	0.18	-	-	-	-	-
Railway incl. railbed	0.83	0.83	0.79	0.79	0.79	-	-	-	-	-
Other	-	0.01	1.03	1.08	0.10	-	-	-	-	-
Impermeable surface	0.05	0.07	3.92	8.58	14.01	-	-	3.57	3.57	3.60
Water surface	-	-	-	0.21	0.21	-	-	0.00	0.00	0.00
Total	100	100	100	100	100	100	100	100	100	100

Figure 5 shows that in 1953, arable land accounted for more than a half of the mapped area at Site A because it is a landscape used intensively for agriculture. Forests (approx. 25%) and permanent grassland (20%) also account for significant shares of the land cover. By 1975, agricultural use intensified, which is manifested in the growth of the proportion of arable land at the expense of permanent grassland in particular; moreover, the share of shrubs and clearings also increased. During the construction of the D1 motorway, the proportion of impermeable surfaces, which was very little up until this period, had increased to approximately 4%. The proportion of arable land began to decrease and the category of "Other" was recorded, which represented the form of previously uncultivated areas after the construction of the motorway. Between 1983 and 2001, there was a significant decrease in the category of arable land, while the share of the permanent grassland, shrubs and forest areas increased. The proportion of impermeable areas increased significantly as a result of the construction of the first part of the industrial site in the southwestern part of the mapped area. In the last period between 2001 and 2018, a further decrease in arable land areas and the continued increase in impermeable surfaces caused by the development of the industrial site should be highlighted. There was also an increase in forest areas at the expense of clearings; similarly, the share of permanent grasslands also increased.

The land cover at Site B (Figure 6) corresponds to the local focus on forestry. In all the examined periods, the dominant categories of land cover are forest or "clearing, windfall", which corresponds to the logging and forest restoration activities of the area. The dramatic increase in clearings and windfall areas in the last analyzed period can be attributed to forced spruce logging in connection with the excessive occurrence of the European spruce bark beetle (*Ips typographus*). The construction of the motorway between the mapping years 1975 and 1983 was manifested by sealing of the soil by impermeable surfaces (3.57% of the area) and the emergence of the categories of permanent grassland and shrubs.

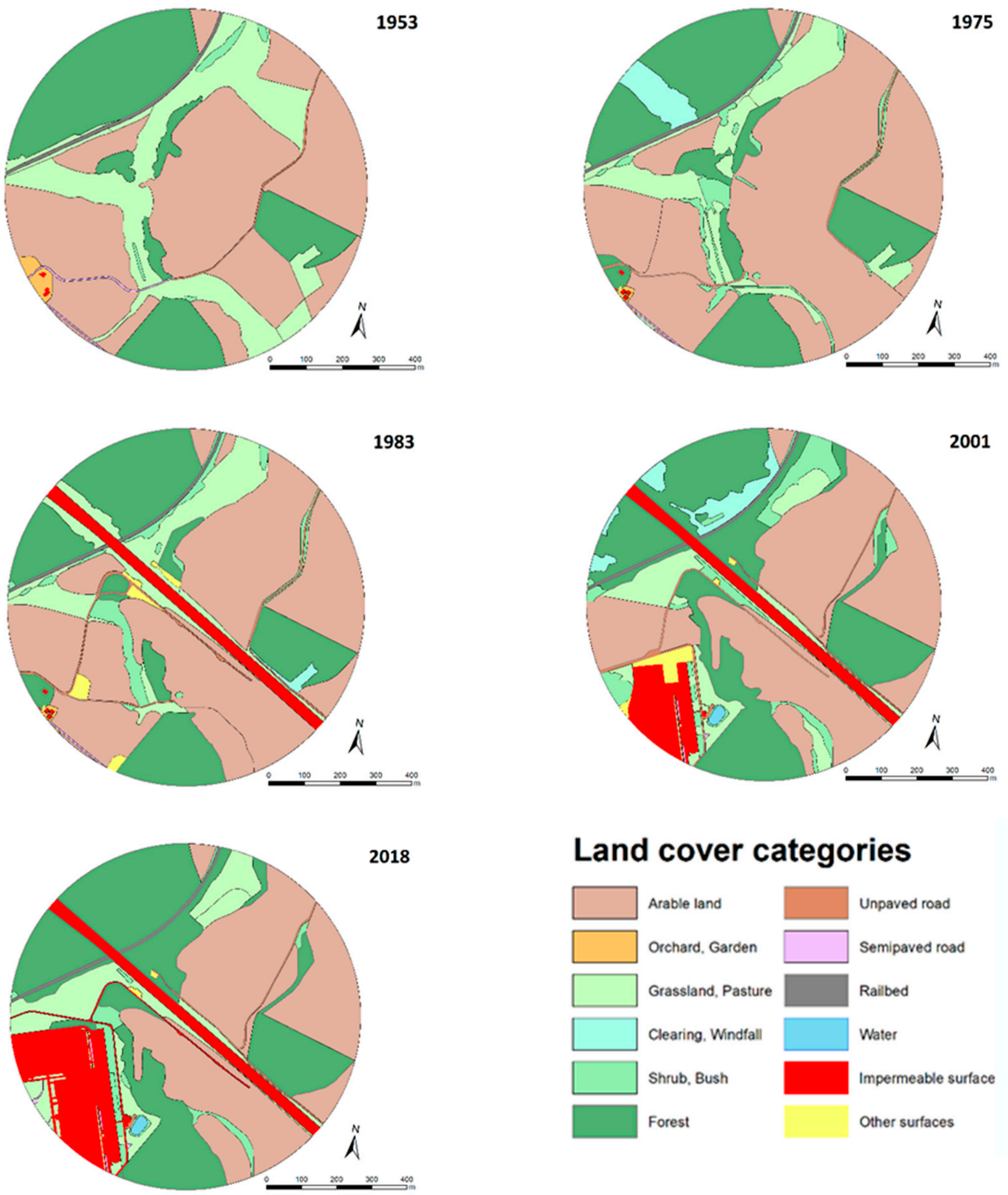

Figure 5. Land cover development at Site A.

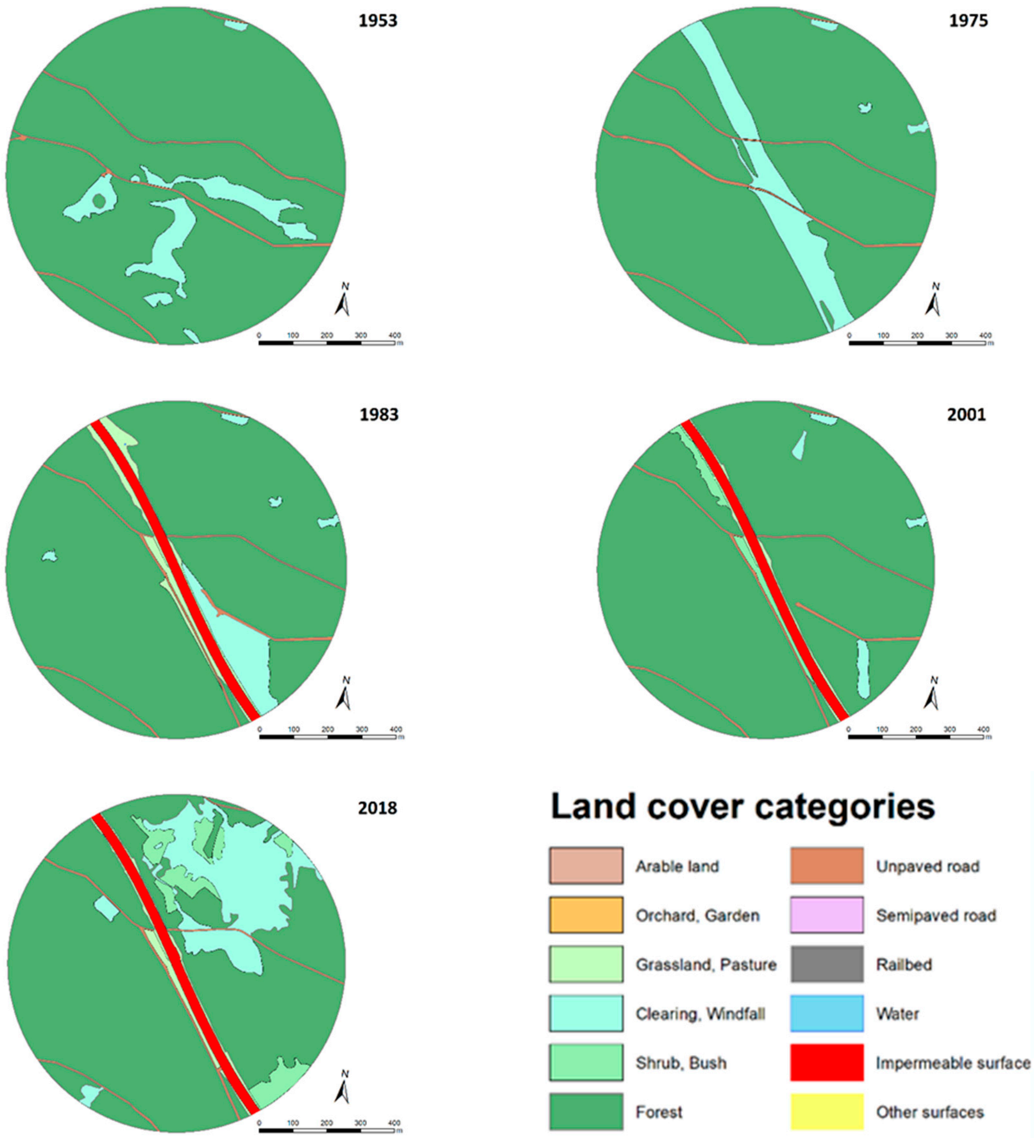

Figure 6. Land cover development at Site B.

The two analyzed sites differ significantly in terms of the classification of the soil in the different hydrological soil groups. While the entire area of Site B belongs to HSG "B", Site A has a significantly more varied composition (Figure 7a,b) with the hydrological soil groups "B" and "C" being the dominant types.

The category of impermeable surface became more prominent in the land cover of the sites only in the context of the period of construction of the motorway between 1975 and 1983. The proportion of impermeable surfaces in Site A continued to grow in subsequent periods due to the construction of the industrial site. The majority of the soil sealing took place on the soils of the hydrological category C; in other categories, the soil sealing increased only slightly. At Site B, there was recorded only a slight increase of 0.02 ha during the last period, which corresponds to the construction of a safe area (hard shoulder) for emergency parking of vehicles during the modernization of the highway; see Table 5.

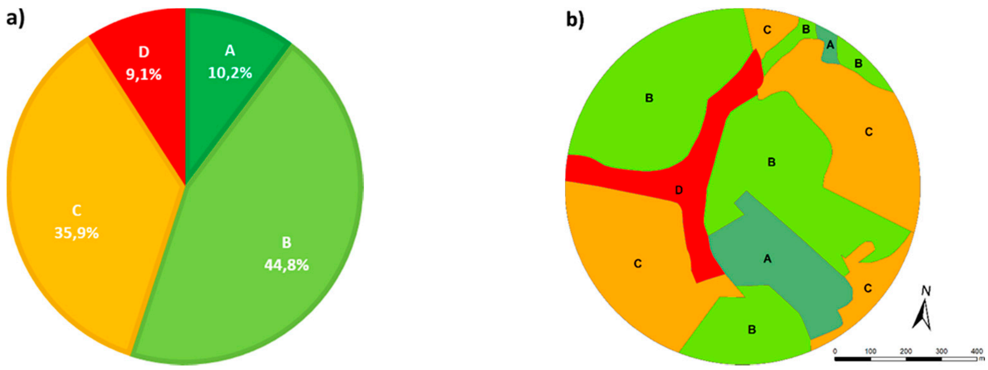

Figure 7. Proportion of individual hydrologic soil groups (HSG) types in the area of Site A (**a**) and their spatial distribution (**b**).

Table 5. Absolute soil sealing by impermeable surface according to individual HSG types [ha].

	Locality A					Locality B				
Category	**1953**	**1975**	**1983**	**2001**	**2018**	**1953**	**1975**	**1983**	**2001**	**2018**
A			0.54	0.54	0.65					
B			1.95	1.95	2.04			2.81	2.81	2.83
C	0.04	0.05	0.36	4.02	7.91					
D			0.22	0.22	0.39					

The development of the average CN value as a basic indicator of long-term changes in landscape retention potential had approximately the same course at both of the monitored sites (Table 6). However, due to their different character and different trajectories of long-term changes in land cover development (see Table 4 and Figures 5 and 6), the reasons for the changes in the average CN value for each site were different. Both sites have a similar decline in retention potential when comparing the period of 1953 to 1975, or 1975 to 1983. In the subsequent period, a positive trend appeared in the form of a decrease in the average CN, where the value increased again in the last monitored period. The same factor influencing the CN value at both locations was, in the period between 1975 and 1983, the construction of the motorway and the subsequent emergence of new impermeable surfaces. The area of these impermeable surfaces was also partially increased in the following periods, especially near Site A, in connection with the construction of the industrial site at the expense of arable land in the hydrological group "C". Despite the increasing area of the impermeable surfaces, the average CN decreased between 1983 and 2001 because of further changes in the land management. These included, in particular, afforestation and growth of areas of shrubs, which have positive effect on increasing water retention in the landscape. Site B was mainly affected by various forms of deforestation, i.e., the creation of areas of the category "Clearing, windfall" as a result of forestry activities.

Table 6. Long-term development of average CN value for the individual sites [%].

	Locality A					Locality B				
	1953	**1975**	**1983**	**2001**	**2018**	**1953**	**1975**	**1983**	**2001**	**2018**
CN average	75.9	76.6	77.3	76.4	76.7	61.8	62.3	63.0	62.0	64.4

3.2. Contamination of Soil with Chlorides due to the Application of De-Icing Salt

In soil samples, the highest concentrations of chlorides at both sites were always measured at the first distance near the road (0 m); see Figures 8 and 9. More pronounced movements in concentration values were also evident in the sampling distances 1 and 5 m at Site A and at the distance 1 m at Site B. These results are also confirmed in the publication [14] which states that the impacts of chlorides on soil are mainly observable at a distance of approximately 10 m from the body of the road. Another research performed in Lithuania confirmed that the amount of chlorides in ground water within 10 m of a road reached the concentrations of 112–500 mg/L [41].

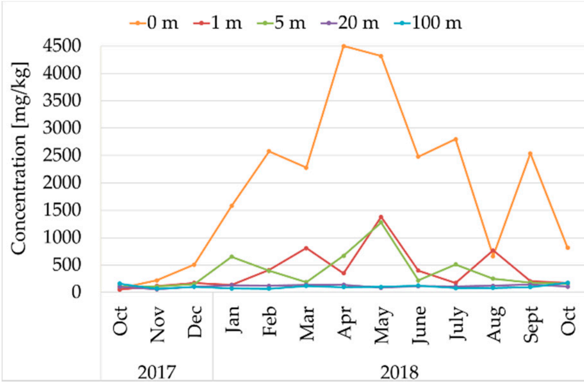

Figure 8. Chloride concentrations in soils at Site A measured at five distances from the motorway [mg/kg].

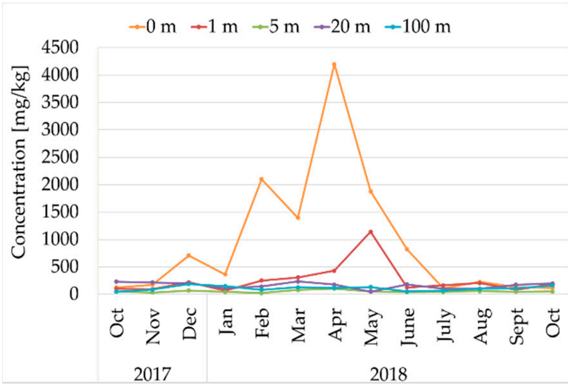

Figure 9. Chloride concentrations in soils at Site B measured at five distances from the motorway [mg/kg].

In the last two distances (20 and 100 m) of Site A, chloride concentrations ranged in approximately the same values over the months of the study as the concentration of chlorides at Site B at the third, fourth and fifth distances (5, 20 and 100 m). The measured values at these distances of the motorway were one order lower than the concentration measured in the distance 0 m at both sites. The results show that, in the reference period from October 2017 to October 2018, soils at distances of 20 and 100 m from the road were basically not affected by the winter maintenance of the motorway.

The trend of the analyzed period showed that the soil samples taken from the distance 0 m at both sites at the beginning of the reference period (October 2017) showed relatively low concentrations of chlorides, specifically, 74 mg/kg (Site A) and 127 mg/kg (Site B). During the following months, the concentrations were gradually increasing until April when they reached the maximum values of 4500 mg/kg at Site A and 4200 mg/kg at Site B; see Figures 8 and 9. After reaching the maximum, there was a decrease in the concentrations, which was different for each site due to different use of the land at the sites. The natural content of chlorides in the soil should not exceed the value of 10 mg/kg; at higher values, the soils are described as "chemically affected". Chloride concentrations at the level of 50–150 mg/kg can be described as slightly saline soils; at concentrations of 150–300 mg/kg, we are talking about moderately saline soils and at concentrations of 300–600 mg/kg about highly saline soils. Chloride concentrations exceeding 600 mg/kg have a negative impact on the conditions for plant growth, in particular in relation to trees and shrubs [42]. Chloride and sodium ions transferred from winter maintenance agents are accumulated in the assimilation organs of conifers growing near roads and have a negative effect on their health [43]. Thus, the assessment of chloride concentrations at Site A shows that the values in distance 0, 1 and 5 m from the road can be classified in the category of highly saline soils. Soil in distance 20 and 100 m can be considered slightly saline soil. Soil at Site B can then be classified in distance 1, 5, 20 and 100 m as ranging between slightly and moderately saline soil and in distance 0 m even as highly saline soil.

At Site A, chloride concentrations were gradually decreasing from the month of May following the amount of rainfall. The site consists of 45% of loamy sand to clay loam soil, 35% of clay loam to clay soil and 10% of almost impermeable soil; see Figure 7a. These soil categories significantly increase the surface runoff ratios and the site can be classified in the category with low to medium water infiltration. The higher intensity of rainfall in May and June therefore caused the gradual washing of chlorides from the earth surface layers; see Figures 4 and 8. The decrease in concentrations caused by washing was always evident at the site with one-month delay compared to the incidence of rainfall.

Site B shows a trend of more rapid decrease in chloride concentrations, which can be attributed to the forests at the location and a different type of soil cover. The decrease in chloride load already reached its low point in July; see Figure 9. In the spring period, with higher incidence of rainfall, most of the groundwater containing chlorides washed from the roads was probably absorbed by the forest cover and vegetation where the chlorides accumulate [43]. The faster decrease of the chloride load on the soil can be attributed to the fact that soils at this location belong to the categories of loamy sand to clay loam soils with medium infiltration rate. Thus, a proportion of the chlorides was probably transferred from the surface water to groundwater due to rainfall. On the basis of research data obtained in Lithuania, it was determined that the washing of chlorides is very significant in sandy soil; on the contrary, washing is low in clay soil [44].

At distances of 1 m (Sites A and B) and 5 m (Site A), the maximum values were recorded one month later than at the distance 0 m where it was recorded in May. For these distances the chloride values ranged around 1200 mg/kg. The increase in the chloride concentration at distances of 1 and 5 m at Site A showed delay, i.e., from January. Over the entire research period, concentration values, except for the maximum, fluctuated in hundreds of mg/kg. Development trend of the chloride content at distance 1 m at Site B copied the trend of distance 0 m; the first increase in concentration occurred in February; see Figures 8 and 9.

Statistical analysis of the data shows that chloride concentrations at both sites are statistically significantly correlated. Correlation coefficient between chloride concentrations at Site A (cCl(s)A) and chloride concentrations at Site B (cCl(s)B) was R = 0.7861 (Sample size $n = 65$). At Site A, chloride concentrations are significantly higher than at Site B, which was confirmed by a pairwise comparison between datasets from both Site A and Site B ($p = 0.00031$). A more detailed analysis found that the concentrations measured at the distances of 0 m from the motorway at both site A (cCl(s)A-0) and site B (cCl(s)B-0) and 1 m from the motorway at both Site A (cCl(s)A-1) and Site B (cCl(s)B-1) correlated significantly (R = 0.7331 and R = 0.8439, respectively, sample size $n = 13$). Correlations between chloride concentrations at Site A and chloride concentrations at Site B at other distances were insignificant at level of significance alpha = 0.05.

Using correlation coefficients, the dependence of chloride concentrations in soil at individual distances from the D1 motorway was further compared with the amount of de-icing salt applied in each month to 1 km of motorway (MNaCl-0). The correlation coefficients characterizing these dependencies proved to be statistically insignificant on level alpha = 0.05. In the following step, correlations of chloride concentrations with the amount of applied de-icing salt and time shift were also tested. The results of this correlation analysis are shown in Table 7 for Site A and Table 8 for Site B. Both tables show correlation coefficients between the chloride concentration in soil cCl(s)A-X or cCl(s)B-X, respectively, at distance X where X equals 0, 1, 5, 20, 100 m, respectively, and the amount of de-icing salt (MNaCl-i) applied, where i equals Time Shift 0 to 5 months (according to the number of months that have elapsed between the application of de-icing salt and the measurement of the chloride concentration). Statistically significant values of correlation coefficients are indicated in bold.

Table 7. Pearson correlation coefficients between chloride concentrations in soils and the amount of applied de-icing salt—Site A. Sample size *n* = 13.

	Time Shift [Month]	cCl(s)A-0	cCl(s)A-1	cCl(s)A-5	cCl(s)A-20	cCl(s)A-100
MNaCl-0	0	−0.2365	−0.1808	−0.0567	0.1041	−0.3737
MNaCl-1	1	0.2125	−0.0143	0.1476	0.4909	−0.3705
MNaCl-2	2	**0.5620**	0.4790	0.3832	0.4009	−0.2247
MNaCl-3	3	**0.7377**	**0.5757**	0.4369	0.3096	0.0050
MNaCl-4	4	**0.8313**	0.5496	**0.7029**	0.0166	−0.0486
MNaCl-5	5	0.4971	0.5250	0.5013	−0.1412	−0.0970

Table 8. Pearson correlation coefficients between chloride concentrations in soils and the amount of applied de-icing salt—Site B. Sample size *n* = 13.

	Time Shift [Month]	cCl(s)B-0	cCl(s)B-1	cCl(s)B-5	cCl(s)B-20	cCl(s)B-100
MNaCl-0	0	−0.0032	−0.1990	−0.1077	0.0696	0.5044
MNaCl-1	1	0.4668	−0.0536	0.0818	0.0030	0.2699
MNaCl-2	2	**0.7304**	0.4320	0.3069	−0.0396	0.1361
MNaCl-3	3	**0.8337**	**0.5735**	**0.5978**	0.0254	0.0161
MNaCl-4	4	**0.6280**	**0.7003**	0.4179	−0.3696	−0.1305
MNaCl-5	5	0.1324	0.4713	−0.0123	**−0.5807**	−0.3487

The results in Tables 7 and 8 indicate that the effect of the application of de-icing salt resulted in an increase in chloride concentrations in soils with a delay of 3 to 5 months. At the distances of 20 and 100 meters from the road, the effect of the application of de-icing salt on soil did not prove to be statistically significant at level of significance alpha = 0.05.

3.3. Contamination of Surface Water with Chlorides due to the Application of De-Icing Salt

Sodium chloride (NaCl) also has a significant influence on water geochemistry as it increases the mobility of ions, especially metal ions, through the ion exchange and contributes to forming chloride complexes [45–47]. De-icing salt may alter the stratification of bodies of water and thereby indirectly aggravate their eutrophication processes in connection with the reduction of water oxygenation and the release of phosphates from organic sediments [48]. Extremely high concentrations of chloride ions in water have a toxic effect on certain organisms [49,50]. Salinity creates a toxic environment for benthic and other freshwater organisms and affects the reproduction capabilities of invertebrates [51]. It may also change the species composition of plant and animal communities in terrestrial and aquatic ecosystems [52,53]. In high concentrations, it reduces the development of biomass and increases the mortality of plant and animal species [54–56].

The research shows that chloride concentrations in outflowing water were at both sites in all cases higher than concentrations in samples of inflowing water; see Figures 10 and 11. Therefore, the results confirm an obvious effect of de-icing salt on the water quality. However, the range of concentrations was different from one site to the other.

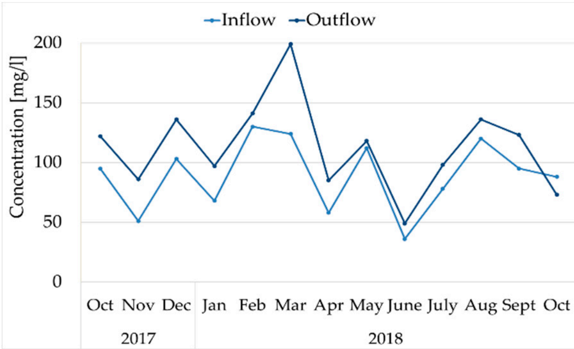

Figure 10. Chloride concentration in inflow and outflow water at Site A.

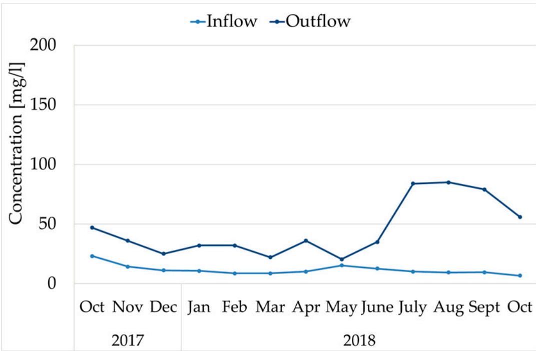

Figure 11. Chloride concentration in inflow and outflow water at Site B.

The amount of chlorides contained in inflow water at Site A fluctuated during the reference period in the range of 36–130 mg/L without any significant maxima and minima; see Figure 10. The concentrations in the outflow water essentially copied the chloride concentrations in inflow water, but the values were higher by approximately 30 mg/L up until April. The maximum was reached in March, when the difference in inflow and outflow concentrations was 75 mg/L and the chloride concentration in the outflow water reached 199 mg/L. In the following period, the differences between chloride concentrations in inflow and outflow water were no longer as pronounced, reaching an average of 20 mg/L.

The pH values measured over the reference period ranged from 7 to 9. The lowest values were measured in both inflow and outflow in May and the highest in October 2018; see Table 9.

Table 9. Monitored parameters of inflow and outflow water at Site A.

Year	Month	Inflow				Outflow			
		Conductivity [μS/cm]	SD [μS/cm]	pH	SD	Conductivity [μS/cm]	SD [μS/cm]	pH	SD
2017	October	261	1.2	7.53	0.15	376	1.2	7.66	0.01
	November	555	0.6	7.39	0.08	664	0.0	7.50	0.12
	December	645	1.2	7.86	0.03	831	0.0	7.77	0.02
2018	January	492	1.2	7.18	0.06	617	1.7	7.42	0.12
	February	626	1.0	7.68	0.03	817	1.0	7.63	0.13
	March	647	0.0	7.15	0.07	787	1.2	7.48	0.05
	April	538	0.6	7.17	0.07	661	0.6	7.64	0.03
	May	617	0.6	6.94	0.02	716	1.0	7.09	0.02
	June	443	1.0	7.28	0.04	450	1.0	7.66	0.03
	July	784	1.2	7.15	0.02	845	1.2	7.32	0.02
	August	970	1.5	7.15	0.02	1023	2.0	7.16	0.02
	September	639	0.0	8.24	0.01	656	0.6	9.06	0.02
	October	650	0.0	8.74	0.02	723	0.6	9.14	0.03

Chloride concentrations at Site B in inflow water ranged in low values throughout the entire reference period, on average around 12 mg/L; see Figure 11. This was due to the location of the inflow sampling site, where the watercourse flows from the source to the sampling point only through a wooded area unaffected by human activity. Chloride values in outflow water were higher by approximately 20 mg/L until April. After a decline recorded in May, they increased during June and July, when the difference reached 76 mg/L. The maximum chloride concentration in outflow water was reached in July increasing to 85 mg/L. Subsequently, the concentrations were slightly decreasing until the end of the reference period (October 2018) to 56 mg/L, achieving a slightly higher concentration than at the beginning of the reference period (October 2017)—47 mg/L.

This site shows the effect of seasonal precipitation. The dry spring months were followed by a period of relatively higher amounts of rainfall in May and June which caused the washing of concentrated chlorides from adjacent soils into the watercourse. The phenomenon of washing of chlorides from soils into surface waters is discussed in detail in other research works [14,57]. The impact of seasonal precipitation on the quality of runoff water flowing into a retention tank used to collect such water is also discussed in [58]. The highest chloride concentration and conductivity was achieved in the summer months, with low precipitation and higher temperatures. On the contrary, in the autumn months, chloride concentration decreased due to increased precipitation and lower temperatures and essentially returned to concentrations recorded in spring [24].

The measured pH values at the inflow and outflow of this site fluctuated between 6.4–9. The minimum and the maximum were reached in the same months as at Site A, i.e., in May and October 2018, respectively; see Table 10.

Table 10. Monitored parameters of inflow and outflow water at Site B.

Year	Month	Inflow				Outflow			
		Conductivity [μS/cm]	SD [μS/cm]	pH	SD	Conductivity [μS/cm]	SD [μS/cm]	pH	SD
2017	October	164	1.5	7.69	0.10	272	0.6	7.57	0.01
	November	164	0.4	7.77	0.04	259	1.1	7.37	0.02
	December	162	0.2	7.60	0.01	243	0.6	7.38	0.01
2018	January	142	0.4	7.68	0.03	226	0.2	7.15	0.05
	February	135	0.2	7.32	0.07	235	0.8	6.90	0.07
	March	118	0.0	6.99	0.10	167	0.2	6.71	0.03
	April	135	0.1	7.20	0.32	248	0.3	7.19	0.03
	May	140	0.2	6.44	0.07	155	0.3	6.41	0.01
	June	151	0.1	7.41	0.02	237	0.5	7.37	0.06
	July	165	0.4	7.35	0.02	531	0.6	7.13	0.04
	August	185	2.4	7.48	0.02	429	0.6	7.16	0.01
	September	177	0.1	7.29	0.02	433	1.2	7.65	0.02
	October	162	0.7	8.14	0.04	309	0.0	9.03	0.02

To determine the relation in the data, a multidimensional analysis was carried out, which included chloride concentrations in watercourses at both of the monitored sites at the inflow (cCl(l) i) and at the outflow (cCl(l) o), the amount of applied de-icing salt per 1 km of motorway (MNaCl) and monthly rainfall (Precipitation). The summary evaluation is shown in Figure 12 (BiPlot Graph), which shows the relationship between the first (Comp1) and second principal components (Comp2).

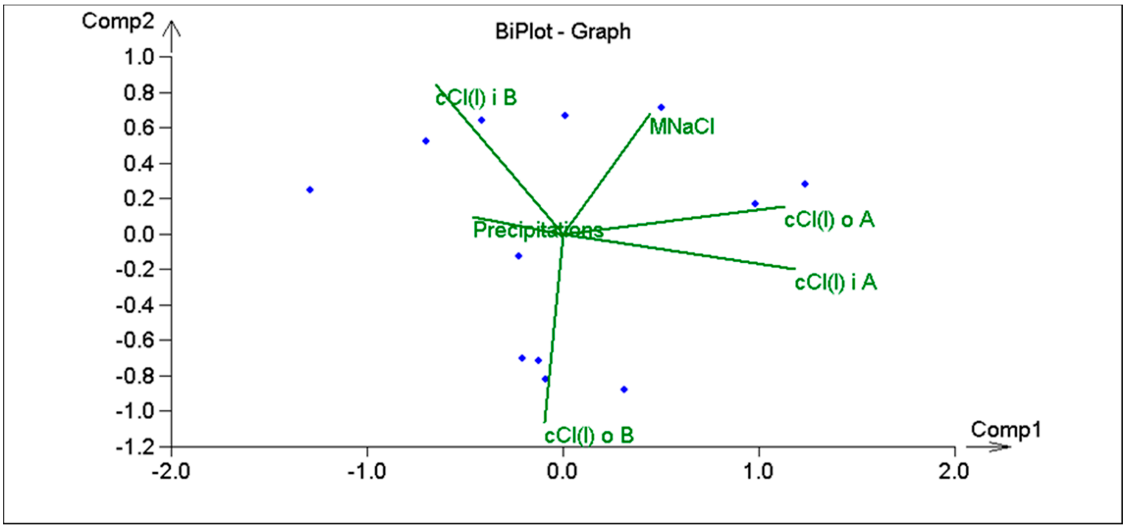

Figure 12. Relationships between variables shown in the BiPlot chart for both sites.

The principal component analysis (PCA) shows that the data includes 3 main components. The first principal component contains the chloride concentration in the inflow (cCl(l) i A) and outflow (cCl(l) o A) at Site A and the amount of de-icing salt applied (MNaCl); the second component contains the rainfall (Precipitation) and the chloride concentration in the inflow at Site B (cCl(l) i B); and the third component consists of the chloride concentration in the outflow at Site B (cCl(l) o B). The correlation between concentrations measured at Site A ((cCl(l) i A and cCl(l) o A) was significant (R = 0.8405). The highest positive correlation coefficients for the amount of de-icing salt applied (MNaCl) were correlation coefficients between the chloride concentrations measured at Site A ((cCl(l) i A, cCl(l) o A) and MNaCl (R = 0.2102 and R = 0.3836, respectively). The correlation coefficient between the Precipitation and the chloride concentration in the inflow of Site B (cCl(l) i B) was R = 0.5142. The correlation matrix is presented in Table 11.

Table 11. Correlation matrix for the evaluation of chloride concentrations in watercourses at Sites A and B (Pearson correlation). Sample size $n = 13$.

Variable	cCl(I) i A	cCl(I) o A	cCl(I) i B	cCl(I) o B	MNaCl	Precipitations
cCl(l) i A	1.0000	**0.8405**	−0.3726	0.0307	0.2102	−0.2915
cCl(l) o A	**0.8405**	1.0000	−0.2700	−0.1264	0.3836	−0.4079
cCl(l) i B	−0.3726	−0.2700	1.0000	−0.3918	−0.0660	0.5142
cCl(l) o B	0.0307	−0.1264	−0.3918	1.0000	−0.5286	0.0306
MNaCl	0.2102	0.3836	−0.0660	−0.5286	1.0000	−0.3065
Precipitations	−0.2915	−0.4079	0.5142	0.0306	−0.3065	1.0000

The differences in these correlations between the chloride concentrations in the inflows and outflows at Sites A and B (Table 11) are likely to be caused by the different flow of watercourses through the landscape within the respective sites. The watercourse at Site A is significantly affected by human activity because it flows through a system of ponds before reaching the inflow sampling point and crosses a D1 motorway junction just before it reaches the D1 motorway. The contribution of chloride load from the application of de-icing salt is evident from the statistically significant correlation between the concentrations measured in the inflow and the outflow at this site. Chloride concentrations in water between inflow and outflow at Site B are not statistically significantly correlated. This is probably due to the fact that the stream at this site springs in a wooded area before reaching the inflow collection point. Thus, the stream is not significantly affected by human activity, and its water is relatively highly pure. Chloride concentrations in this watercourse are therefore most affected by rainfall and the application of de-icing salt on the D1 motorway. The difference in chloride concentrations between the inflow and outflow at this site is more noticeable due to the purity of the inflow water; see Figure 11.

The correlation between the conductivity values is shown in Table 12. Statistically significant were the correlation between the conductivity of the inflow and outflow at Site A and the correlation between the conductivity of the inflow and outflow at Site B. However, the conductivity values between the sites were not correlated. The correlation between water conductivity and chloride concentration was only significant on the outflow at Site B. Based on these correlations, it is evident that conductivity measured at the monitored sites was most likely not caused only by chlorides but also by other ions. Another reason for statistical insignificance of the correlation may be the fact that the overall conductivity values in the monitored water samples were low, on average approximately 705 µS/cm. The literature review suggests that the relationship between chloride concentrations and conductivity values has been demonstrated for conductivity values exceeding 1400 µS/cm, where these values were not affected by other types of ions contained in the water sample [50]. The correlation between conductivity and the content of chlorides in water has also been demonstrated in [59,60].

Table 12. Correlation matrix for water conductivity. Sample size n = 13.

Variable	Conductivity i A	Conductivity o A	Conductivity i B	Conductivity o B
Conductivity i A	1.0000	**0.9122**	0.5286	0.6307
Conductivity o A	**0.9122**	1.0000	0.2847	0.3854
Conductivity i B	0.5286	0.2847	1.0000	**0.7584**
Conductivity o B	0.6307	0.3854	**0.7584**	1.0000

4. Conclusions

During the reference period of 1953 to 2018, both sites showed an increase in land take and related increases in soil sealing by impermeable surfaces due to the construction of the D1 motorway. This fact has clearly contributed to the deterioration of the retention potential of the landscape in the area. Due to the maintenance of the motorway in the winter period, the surrounding soils and watercourses are contaminated by chlorides. Monitoring of the soil chloride load at distances of 0, 1, 5, 20 and 100 m from the road clearly demonstrated that the highest concentrations of chlorides were recorded at the distances of up to 5 m from the road. The level of contamination was related to the amount of de-icing salt applied, precipitation and the character of the site, where the chloride concentrations at Site A were significantly higher than at Site B. The concentration maxima were observed at both sites in April. However, their decline varied from one site to the other in relation to different hydrological soil groups and precipitation. Soils at the distance of up to 5 m from the road can essentially be considered saline soils. Soils at the distance of 20 and 100 m from the road were marked as slightly to moderately saline. The chloride load at these distances is probably caused by the gradual accumulation of chlorides from the winter maintenance of the D1 motorway from the beginning of its operation. During the period under review, there was no significant increase in chloride concentrations at distances of 20 and 100 m.

The impact of the motorway maintenance on surface water quality was clearly demonstrated by increased chloride concentrations in outflow waters at both sites as compared to concentrations of inflow water. Overall, the values of chlorides in the watercourse at Site A were higher compared to the values measured at Site B. This is probably caused by the fact that the watercourses flow through different landscapes before they reach the inflow sampling point. The inflow of Site A is already partially affected by human activity, while the inflow of Site B is unaffected. This is reflected in the significantly lower level of chloride contamination in inflow at Site B compared to Site A and more noticeable differences between the inflow and outflow at Site B in the summer and autumn months. It has been demonstrated that during the reference period from October 2017 to October 2018, the chloride concentrations at both sites were clearly affected by the amount of de-icing salt applied and precipitation.

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