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Long-term road salting effects on dispersion of organic matter from roadside soils into drainage water.

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Abstract

Sodium chloride has been utilized for decades to maintain road safety in winter and some of its detrimental impacts have been well-documented. However, research on the organic fraction of roadside soils has concentrated upon short-term salt-effects. We hypothesise that decades of past leaching and enhanced mineralization of organic matter have reduced the concentrations of dissolved organic carbon (DOC) flushes currently occurring. We have examined the effects of salt concentration on organic matter mobilization in soils that have already experienced varying degrees of exposure to road salting in the field over decades. Applications of salt at concentrations experienced in the field have been simulated to quantify the extent that DOC and dissolved organic nitrogen (DON) are still being mobilized for three prior salt-impact scenarios. A balance occurs between the effects on organic matter of long-term soil pH increase (due to continued cation exchange during salt exposure) which enhances its solubility and organic matter mineralization, short-term pH suppression (due to the mobile anion effect in soil solution) which reduces its solubility, and short- and long-term sodium-induced dispersion. This now determines the influence of road salt on organic matter leaching from roadside soils and into associated drainage waters.

Keywords: Road salt, soil, DOC, DON, pH, long-term effect.

1. Introduction

Since the 1960s deicing agents have been used heavily on European and North American roads in winter to minimize the risk of accidents due to freezing conditions. Consequently elevated concentrations of Na^+ and Cl^- (preferred deicing agent) have

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been observed from tens to hundreds of metres from the road, and often most of the applied salt is transferred to the roadside environment [1]. Deposition is highest within 10 m of roads [1-2]. Most of the potential detrimental impacts of elevated concentrations of Na^+ and Cl^- on roadside soils, vegetation, and ground- and surface-waters have been summarized recently by Green *et al*. [3].

Changes in soil structure may occur following exposure to salt due to a loss of soil aggregate stability through the accelerated leaching of calcium and magnesium from the soil exchange sites as a result of enhanced competition from sodium [4-5]. Polyvalent cations increase aggregate stability via bridge bonding with the organic matter that helps to bind soil particles together to form aggregates. Hence, once Ca^{2+} and Mg^{2+} are leached, the soil organic matter becomes more mobile.

Increased soil colloid dispersion is a function of the exchangeable sodium percentage and the concentration of salt in the soil solution [6]. High exchangeable sodium percentages and low soil solution salt concentrations can disperse colloids and this may facilitate mobilization of accumulated heavy metals from soil into surface waters and ground waters via colloid-assisted transport [7-9]. Transport of organic carbon and nitrogen to surface waters may also be facilitated. Under storm or snow melt conditions, salt concentrations in soil solutions may, however, be reduced due to a dilution effect in soils directly adjoining roads where de-icing salts are commonly used [9]. Potentially, increases in dissolved organic carbon via colloid mobilisation may restrict light penetration in surface waters, hence reducing productivity of algae and phytoplankton, as well as possibly increasing BOD.

It could be hypothesised that DOC and DON concentrations will be higher in heavily salt-effected soils because of the dispersal effect and the higher soil pH caused by long-term Na^+ retention on exchange sites [3]. However, any such positive pH effect could be countered by a mobile anion effect reducing soil solution pH. In order to maintain soil/soil solution charge balance, cations from the cation exchange sites are released as mobile anions are leached from the soil profile. Hence, excess hydrogen ions are emitted due to the electrostatic attraction to the chloride anion, and thereafter transported to surface waters.

Moreover, leaching over several years and enhanced mineralization at the locally raised soil pH might have removed most of the potentially soluble organic matter, i.e. a when-it's-gone-it's-gone hypothesis, drastically reducing potential for enhanced mobilization of DOC. The objective of this study therefore, was to simulate applications of various salt concentrations experienced in the field to three soils that have had a varying degree of exposure to road salting over several decades, to quantify if, and to what extent, DOC and DON are still mobilized due to flushes of NaCl, and to identify and prioritize the processes that may be generating the responses observed.

2. Methods

2.1 Field site

The study site was an upland area along the A6 at Selside, Cumbria, UK (NY 554 046 GB Grid; Lat: 54.434849N Long: 2.689080W), with altitudes up to 458 m above sea level. The road section used runs parallel to the Crookdale Brook over a distance of *ca*. 1.5 km [3]. The brook has an uncontaminated catchment area of acid grassland of ca. 7 km^2 upstream of where the road comes close to it. Soil types consist of podzols, often with poorly developed E and B horizons, with variable depths of organic-rich surface horizons. The bedrock is primarily Upper Ludlow, Ludlow series, Upper Silurian. The vegetation consists mainly of grasses, Juncus and bryophytes and the land is used for light grazing by cattle and sheep. Road drainage is piped directly onto the soil surface at regular intervals. Hence this site presents a valuable opportunity, providing three possible pollution impact scenarios (a) direct (drainage plus spray), (b) indirect (spray) and (c) controls on an adjacent hillside at the same altitude and with the same aspect and land use within the catchment. A wall *ca*. 1.5 m high separates the road from the salt-affected sampling sites, but as this is down slope of the road spray readily passes over it. In the case of the directly affected-soils, the roadside drains are piped directly onto the soil surface through the wall. The sampling scheme was designed to sample each of the three scenarios six times, at a constant distance from the highway for the salt-impacted sites or on soils with identical parent material, age, land use, altitude, slope and aspect in the case of the controls, in order to compare the effects of the three salting scenarios.

2.2 Sample collection

Soils for each salt impact scenario were sampled in sets of six at spacing of at least 10 m. The direct (drain-affected) soils were sampled from sites immediately down slope of drainage pipe outlets (direct); indirectly effected soils (indirect) were sampled approximately midway between drain outlets. The control soils (control) were on the adjacent hill side, well clear of the road. The characteristics of the control soils visibly matched those of soils upslope of the road (above the salt impacted sites). For this reason, and because of the constancy of the soil properties at all six control sites, the authors are confident that the differences between soil and soil extract data for the three scenarios are indeed due to the presence of the road. Soil samples from 0-10 cm were collected with a stainless steel trowel at 3 m from the wall, and thus 4 m from the road itself, on the 19th March 2007. The top 10 cm of the soil were examined as this is the portion of soil receiving the highest concentration of runoff/spray containing road salt, and generally contains the majority of the organic matter which is of prime interest in this study.

The samples were transferred to polyethylene bags after rapid hand-sorting to remove stones and obvious roots, and placed immediately into a cold box. They were stored at 2-4 °C prior to the simulation experiment and chemical analysis. Residual small stones and identifiable vegetation fragments and roots were removed by careful hand sorting immediately prior to use.

2.3 Simulation experiment

For soil from each of the six sites for each of the three salt impact scenarios, sub-samples equivalent to 10 +/- 0.1 g of freshly collected and homogenized field moist soil were inserted into a series of eight 140-ml polyethylene bottles (i.e. 144 bottles in total). To test effects of salt concentrations, four concentrations of NaCl (0, 100, 1000 and 10000 mg l^{-1}) were added to duplicates of each soil. The concentrations of NaCl selected reflect those observed in soil solution at the site across the three salting scenarios [10]. Samples were shaken with 100 ml of appropriate salt solution, and filtered through Whatman No. 42 papers after a 4 hour experimental period. The samples were equilibrated at 2-4 °C for the 4-h period to simulate approximately the winter conditions prevailing when road salt is usually being applied.

2.4 Soil and water analysis

Soils were analyzed for pH, water content, loss-on-ignition (LOI), 1M ammonium acetate extractable base cations (Na^+, K^+, Ca^{2+}, Mg^{2+}), and CEC (80% ethanol pre-wash followed by acidified 1M NaCl). Methods are described in detail elsewhere [3]. Filtrates from the simulation were analyzed for pH, DOC (Elementar Liquitoc) and

Total-N (Bran and Luebbe segmented flow system, AutoAnalyzer III). Dissolved organic-N (DON) was calculated by subtraction of NO_3-N and NH_4-N from Total-N.

2.5 Statistical analysis

Backward multiple linear regressions were completed to determine the degree to which pH and NaCl concentration influenced DOC and DON concentrations. Chemical properties of the different salting scenarios were also statistically compared using One-Way ANOVA, and SNK Post-Hoc. Two way-ANOVA was used to statistically compare DOC, DON and pH with respect to scenarios and salt concentrations. For all statistical tests, significance was accepted at $\alpha = 0.05$. Analyses were performed using SPSS version 11.0.1 (2001).

3. Results

3.1 Soil chemical soil properties

Table 1 summarizes the chemical properties of the soils prior to treatment application. It demonstrates clearly there are significant differences (at the 1% level) in soil pH and base saturation, expressed here as % occupation of cation exchange sites by sodium, magnesium, calcium, and potassium ions, between pollution scenarios. There is sodium dominance for the drain-impacted soils, with declining contribution of sodium to exchangeable cations for spray-affected soils and for the controls, thereafter. This corresponds to a higher dominance of CEC by hydrogen ions for the control soils, and progressively lower H^+ dominance for both spray contaminated transects and drain-impacted soils (significant at 1% - data not shown). The proportions of magnesium and calcium follow slightly different trends to that for sodium ions. LOI increases significantly from the drain-affected soils to the controls.

The soil for drainage-influenced soils had a much higher pH (6.6) than that from the spray-contaminated transects (5.32), which were in turn less acid than the control soils (3.78). This corresponds to the high sodium dominance and lower hydrogen content of the CEC for the salt-impacted soils.

3.2 DOC and DON after equilibration with 0-10000 mg l^{-1} NaCl

Figure 1 shows how DOC (mg l^{-1} and mg kg^{-1} of organic matter (i.e. mg kg^{-1} loss on ignition)), DON (mg l^{-1} and mg kg^{-1} of organic matter), pH and DOC:DON ratio changed with increasing sodium chloride concentration over the range 0 to

10000 mg l^{-1} (filtration after 4 h equilibration). As expected for all the soils with a pH below 7, solution pH declined significantly (1% level) with increasing salt concentration for all salt impact scenarios (Fig. 1e). In addition pH was significantly different at the 1% level between scenarios, with pH declining from drain-affected to spray-affected to control soils (Table 2).

DOC and DON concentrations were significantly different with respect to NaCl concentration applied (1% level), with highest concentrations being released at 0 mg l^{-1}, and the lowest at the intermediate concentration of 1000 mg l^{-1} (Tables 3 and 4). Soil DOC concentrations declined over the salt concentration 0 to 1000 mg l^{-1}, and increased over the range 1000 to 10000 mg l^{-1}.

Soil DOC concentrations released from drain-, spray-affected and control soils were not significantly different from each other. However, DON released was significantly greater from salt-impacted soils than control soils alike pH.

For DOC concentration there is a significant interaction between the degree of prior exposure of soils to road salt and the application of sodium chloride concentration applied to the soils (1% level – Table 2). However, there was no such significant interaction for either DON concentrations or pH (p-value: 0.081 and 0.940, respectively; Tables 3 and 4).

For the most impacted soils, both DOC and DON declined significantly (1% level) with increasing salt concentration over the entire salt concentration range. This was in marked contrast to the control soils. For these, control, soils DOC concentrations declined over the salt concentration range 0 to 1000 mg l^{-1}, but increased over the range 1000 to 10000 mg l^{-1} (Figs. 1a and 1b). The trends for the more moderately impacted soils were intermediate.

3.3 Multiple regressions

The influence of solution pH values and salt concentrations on DOC and DON (mg kg^{-1} per unit organic matter) was assessed by backward multiple regression (Table 5, 6 and 7). Concentrations of DOC for drainage-affected soils are significantly driven down by increasing salt concentration and declining pH (Table 5 - 1% level). However, only 33.6% of the variance in DOC from soil (mg kg^{-1} of organic matter) was associated with pH and salt concentration. DOC concentrations for spray-affected soils are not significantly driven by either NaCl concentration or pH, nor a combination of the two (Table 6). The DOC concentration in the control soil extracts were significantly driven by both pH and NaCl concentration (1% level), explaining 52% of the variance observed (Table 7).

Concentrations of DON for heavily salt-affected soils were driven down by increasing salt concentration (5% level), explaining 7% of the variance observed (Table 5). As for DOC concentration, for the spray-affected soils DON concentration was not associated with pH or salt concentration, nor with both variables combined (Table 6). DON release in the control soils was driven by both pH and NaCl concentration (1% level), explaining 54% of the variance observed (Table 7).

3.4 DOC:DON ratio

The results reported so far suggest that the DOC:DON ratio of the DOM changes markedly with salinity in all soils (Fig. 1f). The controls show an increasing DOC:DON ratio as the concentration of salt increases until 1000 mg l^{-1}, but then a decline. The drain- and spray-affected soils appear to continue to show an increase in the ratio with increasing salt concentration, but the effect is not significant.

4. Discussion

4.1 Soil chemical properties

The low standard errors shown in Table 1 for the soil analyses of the control soils show that the soil is quite consistent. This strongly supports the idea that the differences between the control soils and the impacted soils must be due to road runoff and/or residual road construction effects. The differences between directly and indirectly impacted soils are also consistent, supporting the concept of a large effect of salting rather than one of road construction. It is important to establish that the differences in soil chemical composition for the three contamination scenarios are primarily due to impacts of road drainage water and not a product of natural soil spatial variations. Our preliminary field survey showed that the near-surface soils subjected to the diverse pollution scenarios at the sampling distance from the road were all highly organic, and when not adjacent to the road consistently very acidic (pH 3.78). However, the control soils were apparently more organic rich, and this was confirmed by subsequent LOI % measurement (Table 1). Furthermore, there was no evidence, comparing soils immediately adjacent (at 4 m distance) to the road with the soils further down-slope, of any foreign mineral matter from road construction. However, a road construction effect cannot be unequivocally ruled out, as it is difficult to isolate effects on the roadside environment. It is highly probable that differences between soils were attributable not to natural variation, but to combined effects of road salt in runoff, insoluble particulates from the salt, the additional water flux and associated soil wetness and erosion effects, and possibly to soil particulates redistributed by vehicle flow in both directions along the A6 being flushed from the road surface down drains. The salt used in the area contains 7.5 % insoluble solids, which include small amounts of gypsum and carbonate (Kay Monaghan, Salt Union Pers. Comm.). Thus, the elevated pH observed down-slope of the road is likely to be a product of sodium displacing H^+ from cation exchange sites, and similar effects from magnesium and calcium present as impurities in commercial road salt. As % Ca > % Na on CEC for spray-affected soils (Table 1) it is clear that particulates and/or soluble calcium in splash from the road must make an important contribution to soil partial neutralization.

Both short-term (mobile anion) and longer-term (field) effects of salting on solution are apparent in this experiment (Fig. 1). As the concentration of NaCl solution applied increases there is a switch between impact of mobile anion effect depression of pH and dispersal, depending on the prior degree of road salt exposure in the field. The long-term effects of salt exposure on soil and soil experimental solution are substantial.

Over the range 0 – 1000 mg salt l^{-1} the concentrations of DOC and DON mobilized decline with declining pH (Fig. 1), which reflects the mobile anion effect on pH for all three soil scenarios. This is a consequence of reduced solubility of organic matter at a more acidic pH leading to lower concentrations of DOC and DON in the filtrate. Protonation of functional groups can reduce the solubility of DOM by altering the steric conformation when intra-molecular bonds are cleaved, with van der Waals forces and proton bridging becoming more effective [11].

It is also clear that at 1000 and 10000 mg salt l^{-1}, lower organic carbon concentration is mobilized from salt impacted soils than from the control soils. This supports the hypothesis that so much carbon has already been removed from the impacted soils over many years that salt-induced dispersion has become much less significant at the present time in heavily impacted soils.

Above 1000 mg salt l^{-1}, the less salt-effected soils differ markedly from the heavily salted soils in their response to the increasing concentration of salt. There is a clear shift towards a growing dispersal effect and the concentrations of DON and DOC increase, even although the pH of the soil solutions is still declining. It seems that the threshold levels for the control and spray affected soils are exceeded, leading to the mobilisation of organic matter due to dispersal induced by sodium ion dominance. Dispersal is more significant by far for the control soils.

If sodium chloride additions and/or pH had no effect of organic matter solubilization, and a fixed proportion of the organic matter was soluble at a particular moment in time, then the three scenarios would exhibit identical DOC and DON per unit mass of organic matter at each concentration of NaCl. The soils would behave, relatively, exactly the same irrespective of their overall organic matter content. Figures 1b and 1d clearly show a lower soluble organic carbon and nitrogen per unit mass of organic matter with declining exposure to road salt at low sodium chloride addition. Thus, it may be proposed that long-term exposure to road salt has increased the degree of solubilization per unit mass of organic matter and hence; increased the relative concentration of DOC detected at lower concentrations of sodium chloride additions. As the pH becomes suppressed due to the mobile anion effect the amount of DOC released into solution is lower as solubilization is suppressed.

The hypothesis outlined prior to this study stated that once the organic matter had been dispersed over many years, it is gone from the roadside system; this seems to be the case with drain affected-soils, as the organic matter experiences minimal dispersal as the concentration of NaCl increases with a continually declining pH and therefore dispersal resistant organic matter is retained in the soil profile. The drain-affected soils have been subjected to decades of road salting runoff and spray, which is likely to have solublised and leached the majority of labile organic matter that was present in the past; this would be aided by increased mineralization over the years at the higher soil pH; hence, the low content leached in this experiment for drain affected soils. The spray-affected soils provide an intermediate response between the heavily-salt affected soils and the control soils.

4.2 *Competing effects*

The results highlight the importance of three competing effects: (a) pH suppression (mobile anion effect)/depression of solubilisation with increasing acidity; (b) dispersion of soil organic matter by sodium, and (c) the long-term effect of road salting on increasing the soil pH. The latter would facilitate the loss of organic matter over decades from the most heavily salt-impacted soils and also increase mineralization of organic matter. Both undoubtedly contribute to the large reductions in LOI compared with the values for the control soils. Hence, for heavily salt-impacted soils no dispersal impact is observable now, supporting the "when it's gone it's gone" hypothesis. Therefore, the LOI data provide supportive evidence that the drain-affected soils have experienced OM loss due to a long-term salting effect. Reduced plant growth in heavily salt-effected soils could also be a contributing factor by lowering annual litter input. For the controls at low salt concentrations, falling pH was an important factor, but increased dispersal was marked above 10000 mg salt l^{-1} (1% level). These conclusions are further supported for the control soils by the results from the backward multiple regressions. This is may be a product of more organic matter being available for solubilization indicated by a significantly higher percentage LOI, 72.22 compared to 17.09% for drain affected soils (1% level).

The loss of organic matter may be considered rather high; however, several studies have highlighted major losses of organic matter due to pH increases through the application of lime. Nilsson *et al.* [12] recorded a pH as high as 6.5 in the O horizon after an application of dolomite lime in coniferous forests on podolised soils in Sweden. Over a ten-year period (1984-1994) a *ca.* 12% decline in carbon storage between the control and lime treatment sample areas was observed. A significant increase in DOC and DON leaching (*ca.* 33 and 52%, respectively) was also documented above that of the control 8 years post-application. Similarly consistent declines in carbon storage have been reported in the forest floor as documented by Persson *et al.* [13] for four other sites in Sweden. Hence, the differences in LOI% between salt-affected soils and those of the controls are fairly typically of experiments with liming products to increase soil pH.

Even with the lower CEC, there is considerably more calcium in the salt-affected soils than in the control soils (Table 1). The higher proportional occupation of the exchange sites with calcium ions could be contributing to greater organic matter stability (i.e. suppressed DOC release) in salt-affected soils compared to controls soils at the highest salt additions (Fig. 1a). It was initially thought that organic matter loss may be controlled by the cation exchange reactions and the overall displacement of calcium and magnesium by sodium ions, hence, instability of the organic fraction and thereafter, dispersal. However, the calcium additions via the road salt itself, and/or washed of particulates may increase the organic stability. However, over time the accumulation off sodium and calcium ions on the CEC displacing hydrogen ions has resulted in an increasing pH over the last 40 years. The pH and dispersal effects appear to out weigh that of the enhanced stability of increased occupation of calcium on the CEC.

The long-term and short-term pH effects of differing salt concentrations for the three soil pollution scenarios were evident after 4 hours of exposure (Fig. 1e). Continuous addition of high levels of NaCl to roads over long periods of time leads to raised pH values in the soils and soil solution directly alongside the road (Table 1). The soil pH data demonstrate that the drainage-influenced soils have approximately, on average, one and a half units higher pH than the spray-affected soils, and two and a half units higher than the control soils (1% level), which corresponds to the pH trends in Fig. 1e at 0 mg salt l^{-1} addition. As a result of a higher pH there is a greater concentration of leached DON and DOC, as more alkaline pH promotes solubilisation and mineralization of organic matter. Therefore, a lower concentration of NaCl still may relate to higher solubility of organic matter.

DON leaching is also of particular interest as the ratio to DOC reflects changes in the quality of organic matter. The results suggest that the DOC:DON ratio of the DOM significantly changes with salinity in control soils. Possibly salt induces changes in the structure of the soil organic matter, and the ratio of fulvic and humic acids. Humic acid absorption is higher than fulvic acid absorption at a given pH and salt concentration [14]. So in response to changes in pH and salt concentration, there may be an alteration in the ratio of humic to fulvic acids retained to the mineral surface. Such a change in ratio may influence the changes in DOC:DON due to the differing molecular components of each type of acid. However not all apparent changes were significant in the salt-affected soils; thus further research is necessary to provide a full explanation.

5. Conclusions

The effect of pH suppression, and hence the inhibition of organic matter solubilisation with falling pH, is overcome by the tendency for soil colloids to disperse above 1000 mg salt l^{-1} only for the control soils in this study. There is clear competition between the two factors, which, together with the longer-term effects of increases in soil pH in roadside soils, eventually determine the current influence of road salting on organic matter in these soils and its solubility and transport to associated surface waters. The degree of soil exposure to conditions experienced in the field alongside roads is clearly important. The sodium dispersal effect is not present for heavily salt effected soils as the majority of the potentially mobile organic matter has already been leached and/or mineralized at the enhanced soil pH. Hence, potential for DOC and DON loading of waterways due to leaching is limited. The release of DON and DOC is unlikely in salt-affected soils as, over time, labile organic matter has been lost, and will only become a potential concern if low salt exposed soils become heavily exposed in the future. Thus, a mass flush of organic matter/heavy metals and/or an annual cycle of salt effect impacts for existing roads is unlikely to be observed in adjacent surface waters unless new road construction occurs.

In conclusion, once the organic matter has been solublised and/or mineralized under the influence of road salt and other roadside influences, and thereafter leached, it is gone from the system, and “once it has gone, it’s gone”. Conclusions drawn in earlier studies in which the effect of long-term exposure in the field has not been included are thus almost certainly inappropriate.

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Table 1: Chemical properties of soils for drainage-affected, spray-affected and controls at 3 m from the wall. All results are means with standard errors within the parentheses (n= 6). Cations are expressed as % of CEC. Soils were sampled and field moist soils analyzed, in March 2007.

Pollution scenario	**pH (H_2O)**	**Water Content (%)** †	**LOI (%)**	**CEC ($mmol_c$ kg^{-1})**	**Na^+ (% CEC)**	**Ca^{2+} (% CEC)**	**Mg^{2+} (% CEC)**	**K^+ (% CEC)**
Direct	6.60 (0.18)	37.5 (1.6)	17.09 (1.69)	188 (22)	11.74 (3.39)	9.26 (0.96)	1.35 (0.21)	0.27 (0.04))
Indirect	5.32 (0.71)	42.3 (2.8)	20.87 (3.43)	210 (34)	1.89 (0.82)	11.2 (3.02)	2.82 (0.59)	0.85 (0.17)
Control	3.78 (0.08)	77.8 (1.2)	72.22 (2.90)	847 (80)	0.44 (0.03)	0.93 (0.08)	1.37 (0.08)	0.21 (0.03)

† Soil water content is expressed on a wet weight basis here.

Table 2: Two-way ANOVA testing the hypothesis that pH is not significantly different from each other with respect to salt-impact (scenario) and NaCl concentration applied.

Factor	SS	d.f	MS	F-statistic	P-value
[NaCl]	16.03	3	5.34	15.10	0.000
Scenario	200.08	2	100.04	282.77	0.000
Interaction	0.620	6	0.103	0.292	0.940
Error	46.70	132	0.354		
Total	3971.6	144			

Table 3: Two-way ANOVA testing the hypothesis that mean DOC concentrations are not significantly different with respect to salt-impact (scenario) and NaCl concentration applied.

Factor	SS	d.f	MS	F-statistic	P-value
[NaCl]	2470297	3	823432	37.893	0.000
Scenario	26659	2	13330	0.613	0.543
Interaction	96332	6	160605	7.391	0.000
Error	2868415	132	21730		
Total	3.348E7	144			

Table 4: Two-way ANOVA testing the hypotheses that mean DON concentrations are not significantly with respect to salt-impact (scenario) and NaCl concentration applied.

Factor	SS	d.f	MS	F-statistic	P-value
[NaCl]	280876	3	93625	9.254	0.000
Scenario	497785	2	248892	24.602	0.000
Interaction	116937	6	19489	1.926	0.081
Error	1335428	132	10117		
Total	4008241	144			

Table 5: Multiple backward regression output for DOC and DON (mg kg^{-1} per unit OM) for drain-affected soils.

Variable		SS	Df	MS	F-statistic	P-value	R-square
DOC (mg kg^{-1} per unit OM)[a]	Regression	1217139	2	608570	11.368	0.000	0.336
	Residual	2408946	45	53532			
	Total	3626085	47				
DON (mg kg^{-1} per unit OM)[b]	Regression	111495	1	111495	8.124	0.007	0.150
	Residual	631289	46	13724			
	Total	742784	47				

a Predictors in the model: (constant), pH, [NaCl]

b Predictors in model: (constant), pH

Table 6: Multiple backward regression output for DOC and DON (mg kg^{-1} per unit OM) for spray-affected soils.

Variable		SS	Df	MS	F-statistic	P-value	R-square
DOC (mg kg^{-1} per unit OM)[a]	Regression	34988	2	17494	0.477	0.624	0.021
	Residual	1650596	45	36680			
	Total	1685584	47				
DON (mg kg^{-1} per unit OM)[b]	Regression	28167	2	14083	0.664	0.520	0.029
	Residual	955043	45	21223			
	Total	983210	47				

a Predictors in the model: (constant), pH, [NaCl]

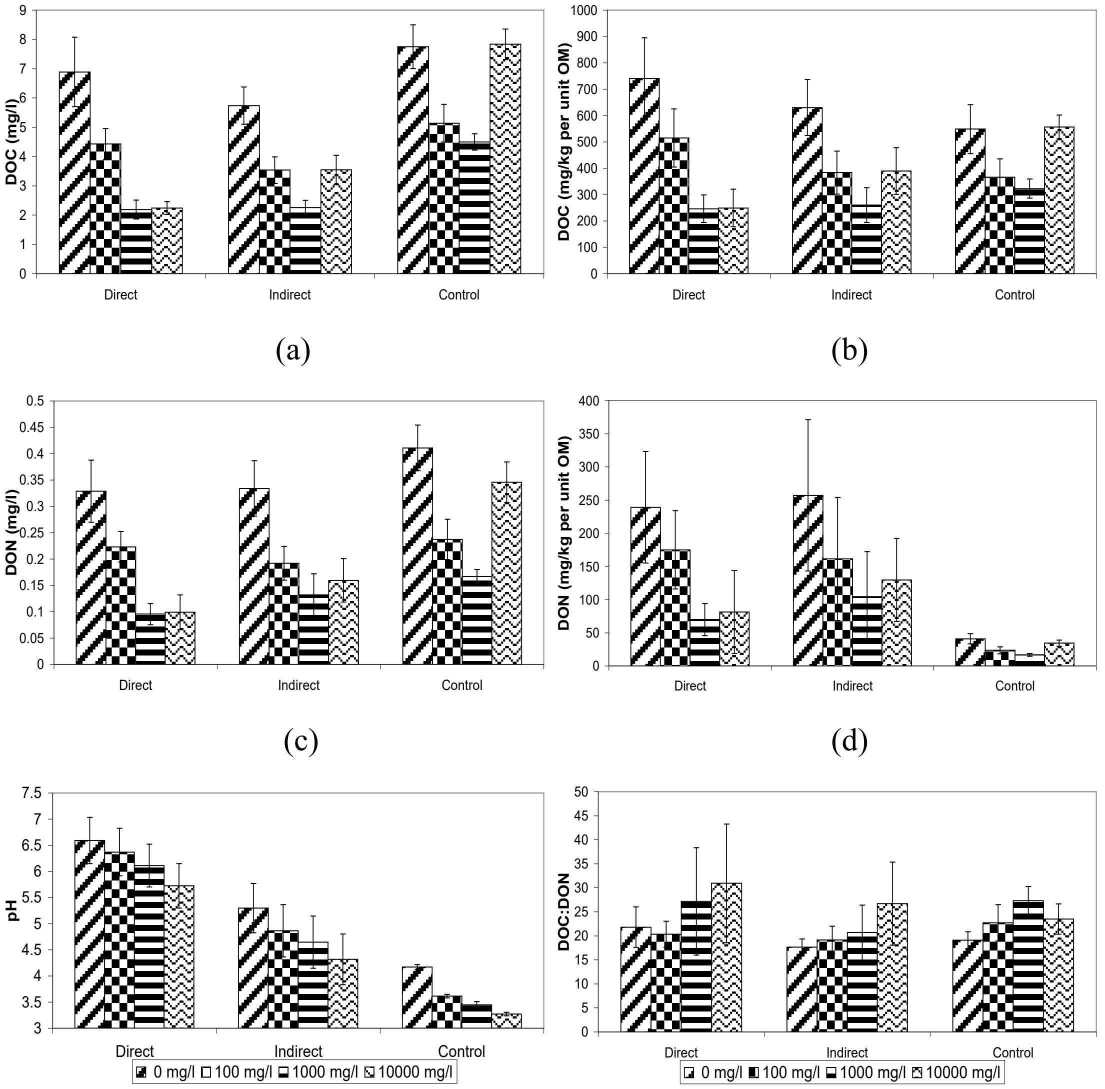
b Predictors in model: (constant), pH, [NaCl]

Table 7: Multiple backward regression output for DOC and DON (mg kg^{-1} per unit OM) for control soils.

Variable		SS	Df	MS	F-statistic	P-value	R-square
DOC (mg kg^{-1} per unit OM)[a]	Regression	514058	2	257029	24.268	0.000	0.519
	Residual	476616	45	10592			
	Total	990674	47				
DON (mg kg^{-1} per unit OM)[b]	Regression	3911	2	1956	26.379	0.000	0.540
	Residual	3336	45	74.13			
	Total	7247	47				

a Predictors in the model: (constant), pH, [NaCl]

b Predictors in model: (constant), pH, [NaCl]



(a) (b)

(c) (d)

(e) (f)

Figure 1: The effect of increasing salt concentrations over the range 0 – 10000 mg l^{-1} on DOC and DON concentrations in filtrates in mg l^{-1}, and in mg kg^{-1} per unit OM, and on filtrate pH and DOC:DON ratio (filtration after 4 h) for drain-affected soils, spray-affected soils and control soils. Error bars reflect 95% confidence intervals. Note changes in scale.