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Research for Investigating and Managing Soil Contamination Caused by Winter Maintenance in Cold Regions

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1. Introduction

In the north temperate and arctic zones, large amounts of de-icing chemicals are used during the frozen season for winter maintenance of highways, roads, airports and other surface areas. The transport sector, and civil aviation in particular, has become a major industry and is one of the fastest growing sectors of the world economy (Janic, 1999). Following an increased concern for the environment in general (Lutz, E. & Munasinghe, M. 1994; Willems, 1994; Jackson, 2010; EEA, 2009), pollution from airports and roads (EPA, 1995) has received more attention. It is also considered one of the contamination threats to soil according to the proposed EU Soil Framework Directive introduced by the European Commission in 2006 (COM (2006) 232; Tóth et al., 2008). In this chapter we will discuss various processes that need to be considered from the source to the recipient, which could be the groundwater or surface waters, and how these are affected by cold climate (winter frost). The source will be related to road network or runways, but can potentially be both a line source if no collection or surface drainage is in place, or can constitute a point source. In cases where road or runway runoff is collected in retention systems with subsequent infiltration into local soils or release to surface waters such as creeks or rivers it could potentially be a significant point source. Often there are no legal limitations on total amounts of salt that can be applied on roads and highways, hence there is no control of their release to the environment. Climate change may cause increased fluctuations about the freezing point which is a condition which increases the use of salts (French et al., 2010). Hence these chemicals may constitute a long term threat to soils in areas with frozen conditions in winter. The second process is infiltration into frozen, partially frozen or unfrozen soils, depending on state of soil and snow fall in late autumn/early winter. Frozen soils may create impermeable surfaces and highly affect hydrological conditions and in particular boundary conditions for unsaturated flow. Further, the flow and transport in the unsaturated zone is affected by soil physical and bio-geo-chemical heterogeneities and in cold climate, high temporal variability in degradation potential due to low temperatures during winter and snowmelt. If or when chemicals arrive at the groundwater level heterogeneous conditions continue to influence the fate of de-icing chemicals but the general

mechanisms that apply have been widely documented elsewhere. In the next sections these processes and how they can be measured and modeled will be presented and we suggest how this knowledge can be used in planning of management strategies. The focus of this chapter is on processes near the surface and in the unsaturated zone. In the conclusions we discuss challenges still unresolved.

2. De-icing chemicals

There are two types of de-icing chemicals; non-degradable inorganic salts and degradable organic compounds. Table 1 shows de-icing chemicals and their associated chemical oxygen demand in the case of complete degradation. For the inorganic salts, the only means of reduction is dilution.

Group	Name	Chemical formula	Chemical oxygen demand (COD)[1]
Inorganic salts	Sodium Chloride	$NaCl$	0
	Magnesium Chloride	$MgCl_2$	0
	Potassium Chloride	KCl	0
Organic salts	Potassium Formate	$KCOOH$	0.35
	Sodium Formate	$NaCOOH$	0.35
	Calcium Magnesium Acetate	$CaMg(CH_3COO)_4$	4.28
	Sodium Magnesium Acetate	$NaMg(CH_3COO)_3$	3.21
	Potassium Acetate	KCH_3COO	1.07
	Sodium Acetate	$NaCH_3COO$	1.07
Others	Propylene Glycol	CH_3CHOCH_2OH	1.69
	Ethylene Glycol	$HOCH_2CH_2OH$	1.29
	Diethylene Glycol	$(HOCH_2CH_2)_2O$	
	Fructose/glucose/Sodium Chloride	$C_6H_{12}O_6/ C_6H_{12}O_6 /NaCl$	
	Urea	$(H_2N)_2CO$	2.13[2]

Table 1. Most commonly used de-icing chemicals. [1] Chemical oxygen demand (COD) mg/l based on standard stoichiometrical calculations assuming complete degradation [2] assuming reduction to NH_4.

2.1 Inorganic salts

Some examples of total use of salts on Norwegian national roads per winter season are given in Table 2. The average salt consumption in 2009/2010 was 14 tons per km high priority roads.

Negative consequences of increased salt concentration along roads have been documented (e.g. Nystén and Suokko, 1998; Oberts et al., 2000). Howard & Maier (2007) simulated increased NaCl concentrations in groundwater due to urbanisation near Lake Ontario. According to their Visual Modflow simulations, maximum concentrations of 5000 mg

NaCl/l were predicted near roads with an annual use of 20-250 tons NaCl per km road (dual to multiple lane motorways), and that stabilisation occurred after 700 years. The consequences for the environment can be groundwater of insufficient quality for drinking water. Direct or indirect release to lakes can give hypolimnetic conditions and prevention of biannual full circulation with reduced oxygen levels (Bækken et al., 2006). Release into creeks can give negative effects to biologic systems (Meland, 2010).

Winter season	NaCl (tons)	Sand(tons)
2005/2006	166,000	435,000
2006/2007	137,000	386,000
2007/2008	160,000	484,000
2008/2009	200,000	443,000
2009/2010	201,000	417,000

Table 2. Total consumption of road salts and sand on Norwegian national roads (the Norwegian public roads administration, www.vegvesenet.no).

2.2 Organic de-icing chemicals

Organic salts such as Potassium Formate, Potassium Acetate and Calcium Magnesium Acetate are used on runways and some roads (US EPA, Amundsen et al., 2008). These chemicals are more costly than the inorganic salts, but are used because they are degradable and less corrosive. Normally these chemicals are also released directly into the ground next to the road or runways.

Propylene glycol, Ethylene glycol and Diethylene glycol are the main constituents of de-icing chemicals used to keep planes and other vehicle surfaces free of ice. The de-icing at airport takes place on special de-icing platforms which normally collect the surplus chemicals with subsequent re-cycling or treatment in treatment plants (Øvstedal & Wejden, 2007). Estimations from Oslo airport, Gardermoen, suggest that 80% is collected at the de-icing platforms, about 10% is released to the local environment at take-off, and 10% leaves with the planes (Øvstedal & Wejden, 2007). Still many airports in the world do not collect the de-icing chemicals and they are released into the local environment (US EPA).

The treatment of water contaminated with organic de-icing chemicals is usually based on in-situ or off-situ aerobic and/or anaerobic degradation. The tested techniques vary from constructed wetlands, reed beds, constructed soil filters (Roseth and Bjørnstad, 1998; Roseth et al., 1998) natural soil profiles (French et al., 2001, Jaesche et al., 2006) to more conventional wastewater treatment plants (Rusten et al., 1999). The chemicals are supplied during the frozen season which may vary geographically but falls roughly between October to April. The objective of using de-icing chemicals is to reduce the freezing point of water, hence water will remain in a fluid state below 0°C and can infiltrate into the ground as long as there are open pores. Water containing de-icing chemicals may therefore infiltrate the ground prior to the main snow melting period. The preferential melt-out of de-icing chemicals was shown by French & van der Zee (1999) as also shown for other chemicals stored in snow (Johannesen and Henriksen, 1978). Although the organic de-icing chemicals are degradable and the top soil hosts large amounts of bacteria and fungi, the temperatures are negative or close to zero, and there is little degradation during this period. Half lives of Propylene Glycol and Acetate under field conditions throughout melting period and into the summer ranged between 15-46 days (French et al., 2001). Degradation rates of other

airport chemicals, as well as the increase of rates caused by optimizing the C:N:P ratio are reported in French et al. (2002). Although degradability of de-icing chemicals in general is positive for the environment, negative consequences can be anaerobic conditions. Field experiments (French et al., 2001) showed an increase in manganese concentration when propylene glycol and Potassium acetate was supplied to the ground surface even in the unsaturated zone, indicating oxygen limitation. On-going research (French et al., 2009) suggests that nitrate could increase remediation and improve redox conditions in local soils.

3. Non-invasive versus destructive methods for soil characterisation

The theoretical considerations concerning spatial variability, described in the next section requires that we have some knowledge about 1) the geostatistics of the hydrogeological properties, and 2) the scale of the contaminant source or plume relative to this variability.

3.1 Non-uniform infiltration

The boundary conditions influencing the flow and transport during snowmelt are characterized by ground frost and the formation of impermeable ice on the ground surface which redistributes melt water during the snowmelt period (Fig. 1). The ice-cover often inhibits infiltration in sediments with otherwise high infiltration capacity (sandy aquifers). As a result, a large amount of melt water collects in depressions or becomes surface runoff. According to Baker & Spaans (1997); Derby & Knighton (1997); Johnsson & Lundin (1991), infiltration during snowmelt often occurs as focused recharge in local depressions on the surface.

Monitoring water accumulation in snow and also the infiltration below the snow cover is a challenge. The traditional but laboursome way of collecting information about snow cover is to measure snow depths along a transect and estimate the snow water equivalent (SWE) by weighing the snow column. The advantage is that one gets a spatial coverage, but only for single times, also the method is destructive. Snow pillow measurements involves placing a logged scale beneath the snow cover, hence direct measurement of SWE above the snow pillow, this method prevents infiltration and is only representative of one location, also it may give wrong values when ice crusts are formed in the snow which reduces the weight load on the scale. Remote sensing using natural emissions of gamma rays and micro waves has become a common method for mapping snow storage over larger areas (Glynn et al., 1988; Durand et al., 2008; Schaffhauser et al., 2008), but also local measurements can be conducted using this principle (Campbell scientific; Bland et al., 1997).

A snow lysimeter is a method where meltwater is collected via drain pipes from trays below the snow cover, and volumes and quality measured. Hence destructive in the way that water is prevented from infiltrating the ground. Variations in release of water from a melting snowpack was documented by French & van der Zee (1999) by collecting meltwater from snow lysimeters placed beneath a melting snow cover. Despite a fairly uniform snow depth over the monitored area (a few square meters), the total melt volumes varied from nearly 0 to 200% of expected values based on the snow water equivalents measured prior to snowmelt. To further characterise the infiltration pattern in the soil, French & Binley (2004) installed electrodes near the surface and monitored changes in electrical resistivity of the soil volume, which indirectly reflected changes in water contents during snowmelt. Figure 2, shows the spatio-temporal variability of electrical resistivity observed in a horizontal plane during this experiment. This may cause higher velocities through the unsaturated

zone than during evenly distributed infiltration on the surface, hence causing less than optimal conditions for degradation of pollutants. Preferential meltout of chemicals and subsequent redistribution of meltwater may also cause concentrations in soils next to the impermeable covers which are higher than expected from a pure mass-balance point of view. In addition to the temporary and variable surface conditions created by frost and snow, impermeable surfaces, membranes and other installations near the soil surface may highly affect the infiltration and flow pattern in the unsaturated zone next to roads and runways (e.g. Apul et al., 2007).

Fig. 1. Surface conditions at Oslo airport, Gardermoen, near the end of snowmelt, showing patchiness of snow and ponding of melt water due to soil frost.
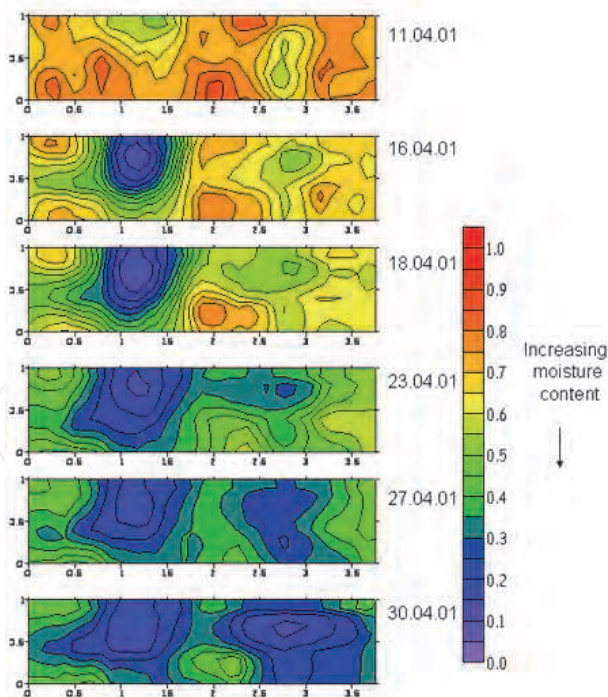

Fig. 2. Variable infiltration, increased moisture content as blue areas, observed as reduced electrical resistivities near the surface of a soil below a melting snow cover (modified from French & Binley, 2004).

3.2 Characterisation of soil heterogeneity

Conventional techniques, such as soil sampling and core drillings provide more or less disturbed samples on which one can measure porosity, unsaturated hydraulic conductivity and Pf curve (if undisturbed) or grain size distribution and water content on disturbed samples. The Pf curve describes the relationship between soil suction and soil water content, this curve determines the unsaturated hydraulic conductivity (ref). Well established pedo-transfer functions such as Hazen's equation (ref.) can be used to calculate hydrogeological parameters such as the hydraulic conductivity. The samples can also be used for bio-geo-chemical characterisation interactions between contaminants and soil. A number of spatially collected samples can then be used to establish geostatistical properties of the different hydrogeological parameters (discussed in the next section). More recently advanced direct push technology, which provides opportunities for in-situ measurements e.g. of hydraulic conductivity by the use of specialised probes at the end of the direct push probe. The disadvantage of these methods alone is that they are destructive, time consuming, expensive; and do not give a continuous image of the subsurface. Deep geophysical exploration has been around since the beginning of the last century, and is a common method for geological characterization in oil exploration, mapping of lithostratigraphy, fracture patterns in bedrock and is described in several text books (e.g. Kearey & Brooks, 2002). In the last couple of decades geophysical techniques such as those described in Table 3 have become more common for hydrogeological applications, for further reading see e.g. Regli, et al, 2002; Hubbard & Rubin, 2000, Kowalsky et al., 2001; Rea and Knight, 1998; Rubin & Hubbard, 2005; Veerecken et al., 2006.

As an example we discuss briefly the principle of the electrical resistivity method. The electrical properties of soils are a function of the soil type, water content, soil temperature and ion content of the soil water. Measurements of soil bulk electrical resistivity are most commonly conducted by placing a set of electrodes in the ground along a line on the surface or in vertical boreholes. By inserting a known current and measuring the resultant voltage consecutively over the set of electrodes, one can after an inversion of the collected data obtain an image of the distribution of electrical resistivities in the soil volume next to the electrodes (see eg. Reynolds, 1997). A single measurement may reveal geological features of the subsurface, while the comparison of images taken at different times (time-lapse measurements) can help quantify spatial and temporal variability caused by changes in water (Daily et al., 1992) and ionic contents.

The advantage of geophysical techniques over the more conventional and invasive techniques is that they are non-destructive and provide continuous images of the subsurface. The challenge of geophysical methods however is the ambiguity of their interpretation. The non-invasive geophysical methods map zones or layers of different physical characteristics (Table 3). The interpretation of such data requires that the data is run through an inversion code which basically "suggests" a likely distribution of the specific geophysical responses in a 1, 2 or 3D space. The results are optimised with respect to measurements conducted on the surface or in boreholes. Forward and inverse modelling of the system that is being studied is required for optimising the configuration of measurements, this technique can also be used in a stochastic framework in order to include uncertainty and coupling to soft and hard data for hydrogeological characterisation (Rubin and Hubbard, 2005). Another recent development to reduce the non-uniqueness of the interpretation is to combine different geophysical data sets collected at the same location and time through joint inversion (e.g. Gallado & Meju, 2004; Linde et al., 2006). However

combining measurements from geophysical techniques and ground truthing (bio-geo-chemical data) is still required for more complete and accurate characterisation of spatial variability of hydrogeological parameters such as porosity, hydraulic conductivity and contaminant concentration.

Method	Sensitive to	Procedure	Can represent	Example references
Seismic refraction/reflection	P-wave velocity and reflectivity	Sound wave, Sound source and set of geophones	Top of bedrock, lithology, water table, faults	Steeples, 2005;
Ground penetrating radar	Dielectric constants	Electromagnetic waves, Transmitter and receiver antennas,	Lithology, water table, water contents	Annan, 2005; Farmani et al., 2007
Electrical resistvity	Electrical resistivity	Electrical current injected and voltage measurements, usually electrodes in the ground	Lithology and zonation, water table, contaminant plumes, water contents	Binley & Kemna, 2005; Daniels et al., 2005;Kemna et al., 2006; Godio & Naldi, 2003
Electro-magnetic	Electrical resistivity	alternating or pulsed current through long wires or coils	Zonation, water table, contaminant plumes, water contents	Everett & Meju, 2005;
Induced polarisation	Chargeability	Same as for electrical resistivity, but non-polarisable electrodes	pore-fluid conductivity and grain-surface polarisation – can be used with ER data to separate lithological units	Binley and Kemna, 2005; Kemna et al., 2006; Vanhala et al., 1992
Self potential	Streaming or Electrochemical potential	Passive voltage measurements, non-polarisable electrodes	Zones with concentrated flow, zones with high degradation	Naudet et al, 2003; Revil et al., 2006; Arora et al., 2007

Table 3. Geophysical methods for soil physical and hydrogeological characterization.

3.3 Characterisation of contaminant plumes

Solute transport, which is basic to natural attenuation processes, requires new measurement techniques to provide spatial distributions and internal spatial characteristics as current modelling of these phenomena is conditioned by availability of experimental data.

Traditional monitoring techniques may neither capture the contaminant distribution nor their removal rates sufficiently (Aagaard et al., 2004). Subsurface characterization of contaminant distribution over large scales is challenging, since the contaminants may have moved erratically through the profile as illustrated in Figure 3, and point samples as provided with conventional sampling techniques (as discussed above) may not provide a representative measurement of the situation. The most common measurement technique for monitoring contaminant transport in the unsaturated zone is sampling of soil water through suction cups. These consist of a porous material such as ceramics or an inert material e.g. Teflon and quartz with a pore size small enough to ensure contact between the filter and the soil. An example of such a system is the experimental lysimeter trench at Moreppen near Oslo airport, Gardermoen, Norway (French et al., 1994). This experimental site has more than 100 Prenart suctions cups and various other soil physical measurements. It was constructed to conduct controlled experiments of transport of Propylene glycol and Potassium Acetate during snowmelt. Several studies were carried out at the same site in order to examine the hydrogeological properties in the unsaturated and saturated zones and the transport processes.

Contaminants may, depending on their chemical properties, affect the geophysical signature of the soil. Salts will increase the electrical conductivity (EC) of the pore fluid, while hydrocarbons will have the opposite effect. The organic and inorganic de-icing salts will reduce the electrical resistivity of the soils, while Propylene glycol will not affect the electrical conductivity of the pore fluid. Electrical and electromagnetic methods are widely applied for soil mapping and detecting of contaminated plume. Over the last decade new geophysical methods such as induced polarisation (e.g. Godio and Naldi, 2003; Slater & Mansoor, 2006), electromagnetics, GPR, micro-sesimics and self potential (Naudet et al., 2003; Arora et al, 2007) have been explored as methods for exploring contaminated sites. Low frequency electromagnetic (EM) methods are usually adopted for fast mapping and preliminary assessment of the aerial extent of the potentially contaminated land. A qualitative image of the soil mineralization, due to degradation of hydrocarbons, could be inferred by integration of resistivity and induced polarisation data (e.g. Godio and Naldi, 2003, Slater et al. 2006). Electrical Resistivity Tomography (ERT) is a powerful tool for investigating pore fluid properties (Olsen at al., 1999; Kemna et al., 2000; Depountis et al., 2001; Damanesco and Fratta, 2006;) as shown in laboratory experiments (Comina et al., 2005) and for solute transport in undisturbed soil columns (Binley et al., 1996) and field sites (Slater et al., 2000; French et al., 2002, Binley et al. 2005). How to estimate hydrogeophysical parameter is still one of the major challenges, state-of the art knowledge is described by Linde et al., (2006). Another challenge for combined interpretation of geophysical and point measurements is that the support scale of different methods varies; hence a statistical framework is required for joint interpretation.

4. Modelling implications

As evidence shows, the subsoil is in general heterogeneous (or spatially variable), and often this heterogeneity is partly irregular. This irregular variation has been the motivation to consider soil as an intrinsically random material, i.e., as a material that can only be described statistically. This assumption has resulted in a large body of literature (Bellin et al., 1993; Dagan, 1997; Keijzer et al., 1999; Janssen et al., 2006; Cirpka, O.A., P.K. Kitanidis, 2000; Fiori et al., 2002), that is still actively being developed and is quite mathematically inclined:

stochastic groundwater hydrology and contaminant hydrology. For the present purpose, it is not useful to emphasize the mathematics, although some of it cannot be avoided. Rather, emphasis is given to illustrate the effects of heterogeneity and to give an impression of what that means for decision making.

4.1 Stochastic hydrology

The mentioned stochastic approach to contaminant hydrology (that focuses on the transport behaviour and fate of soil and groundwater contaminants) implies that particular properties are considered to be random space functions (RSF): they vary irregularly in the horizontal plane and with depth. However, they do not do so completely randomly: due to the genesis of layering, and soil horizons, patterns of large and small values can be observed. These patterns are described by autocovariance functions. Examples of strongly variable properties are the hydraulic conductivity, and for the water unsaturated zone also the soil water retention function (Van der Zee & Boesten, 1991). Much research has established that the hydraulic conductivity, and the scaling parameter in this function that also is used to describe water retention, are well described with a lognormal distribution. Also for several important properties that control the adsorption behaviour of contaminants, the lognormal distribution appeared to be reasonable (Van der Zee & Van Riemsdijk, 1987, 1988; Boekhold & Van der Zee, 1991).

Randomness can have a large effect on flow and transport (Rode et al., 2010). Basically, this effect is (i) a more irregular transport behaviour of the contaminants, and (ii) a larger uncertainty about this behaviour. To give an impression of how heterogeneity affects the transport pattern, Figure 3 shows the leaching behaviour of both inert chloride and degrading de-icing chemical at Oslo Airport, Gardermoen (French et al., 2001). We observe a very irregular 'plume' of contaminant. A problem with such complex patterns is that they are difficult to communicate, other than by sending a picture.
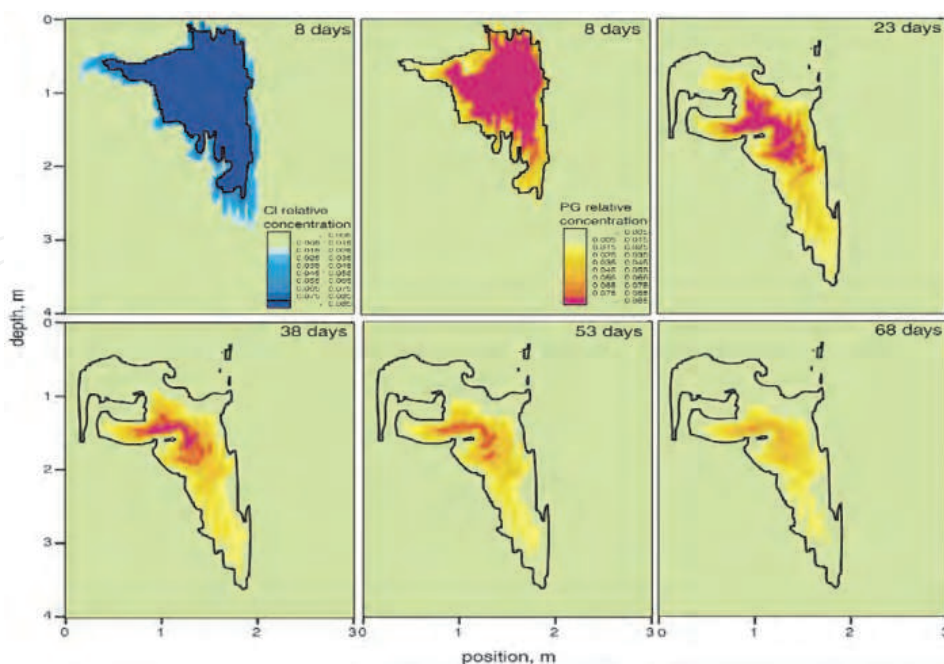

Fig. 3 The erratic pattern of chloride and propylene glycol transport in a heterogeneous soil representative of the Moreppen site at Gardermoen (French et al., 2001).

In addition, we have to deal with uncertainty, because the pattern if the contaminant were released on a spot a few meters away, might look similarly irregular but not exactly the same! It might be slightly deeper or shallower, moved to the left or right, and have another shape. Whereas for contaminated sites, it may be meaningful to know the exact pattern, for making predictions, the precise shape of the plume becomes less important: a calculation for another spot would look always more or less different. For this reason, we need more robust measures to quantify the transport behaviour, than the pictures of Figure 3.

4.2 Spatial moments

To this aim, we explain the moment theory, with which such quantification is feasible at different levels of detail. Using this moment theory, it is feasible to illustrate the profound impact of spatial variability on contaminant transport in terms of pollution front, breakthrough time, and of course remediation efficiency.

If soil and aquifer properties are spatially variable, two limiting situations may arise. These two limiting situations are related with those of point source pollution and of diffuse source pollution, respectively. The point source pollution problem can be defined as the situation where the source is relatively small (or of similar order of magnitude) compared with the scale of (random) heterogeneity. In that case, the source of contaminants might be visualized as being in a spot of large or of small hydraulic conductivity. In the first case, contaminants may readily spread in the environment, whereas in the latter case, this occurs slowly. For the point source pollution event, it is difficult to predict whether the source will be at in contact with the fast or the slow route of transport, which leads to high uncertainty. In the diffuse source pollution case, contaminants enter the soil or aquifer over a large surface, and both fast and slow transport routes are 'sampled'. In that case, uncertainty is much smaller, and heterogeneity leads to large spatial variability of fluxes. This distinction of two limiting situations is illustrative, but in view of the nested scales of heterogeneity of soil and aquifers, many real situations will be somewhere in between.

Fig. 4. Photo of a heterogeneous soil profile.

It is clear from Fig. 4, that the thickness and intensity of soil horizons varies in the horizontal plane. As soil genesis processes involve longer time periods, spatial variability must be due

to persistent differences in soil composition and do not necessarily result from differences in 'input' at the soil surface.

For the two limiting cases mentioned above, the 'diffuse' pollution case is called an ergodic situation, because 'all' heterogeneity is sampled simultaneously. This leads to a distributed answer, such as a spatially variable contaminant plume or breakthrough curve. If these answers are measured or calculated at another place, they do not significantly change. Hence uncertainty is limited.

However, the distributed answer is complicated and difficult to communicate to others who cannot see the picture. Moreover, it is commonly too detailed for e.g. management decisions. Therefore, it is appropriate to consider robust measures of the results, which focus on the major issues. Useful examples of such robust measures are found in the theory of moments. We will give the definition of these moments in terms of spatial moments (representing the situation at one instant in time), but temporal moments (at one particular location, plane or volume) are equally feasible. The spatial moments of a property P, where the space coordinate is x, are given by

The zeroth' moment, or the mass of the distribution:

$$M_0 = \int_{-\infty}^{\infty} f_P \, dP \tag{1}$$

The first moment, or the mean of the distribution:

$$M_1 = \frac{1}{M_0} \int_{-\infty}^{\infty} P f_P \, dP \tag{2}$$

The second central moment, or the variance of the distribution:

$$M_2^c = \frac{1}{M_0} \int_{-\infty}^{\infty} \left(P - M_1\right)^2 f_P \, dP \tag{3}$$

For a contaminant plume as shown in Figure 3, the zeroth' moment is equal to the amount of dissolved chemical in the contaminant plume, if P is equal to the position x, and the function fx represents the spatial distribution of the contaminant concentration multiplied with the water-filled porosity. The first moment represents the mean position (in direction x) of the contaminant distribution and the second central moment represents the width of the zone over which the concentration distribution occurs. Whereas we illustrated the moments where P is equal to position, many other properties can be chosen. Which to choose depends on the primary interest, but examples are the concentration or mass of contaminant, the quantity or fraction of contaminant leached beyond a reference plane, such as groundwater level and so on. In principle, the transport problem for de-icing chemicals is a very complicated one, even in one direction such as depth. The reason for that is that a complex of interactions is affecting this transport. Examples are infiltration conditions, ad/desorption, microbial degradation according to different kinetics (1st, Monod, 0th order), and a host of different redox-sensitive components that may affect the degradation rate (Beltman et al., 1996; Keijzer et al., 1998) may occur, depending on ambient conditions as well as the influx of degrading substrate (continuous, instantaneous). To give an impression

of the complexity, we provide graphs in Figure 5, that reveal that the concentration pattern may be completely different from what we are used to, and that the transport process is quite irregular in space.
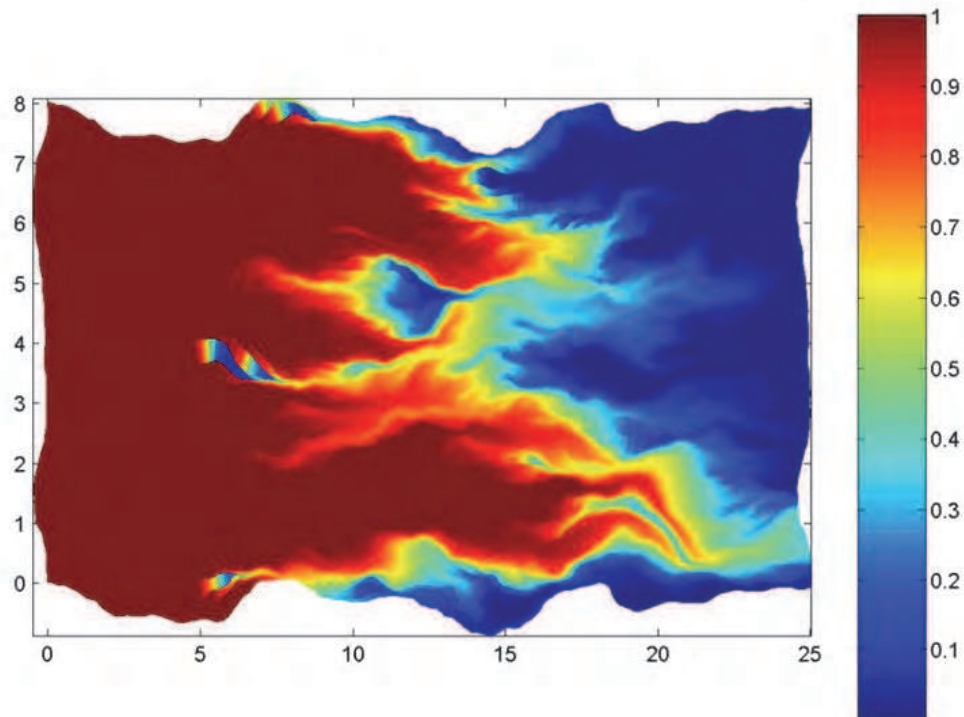

Fig. 5. Computationally determined map of concentrations of an electronacceptor (EA) such as oxygen or nitrate with scaled concentrations ranging from 0 to 1. The EA enters the aquifer from the left and moves towards the right, and is used by micro-organisms to degrade initially resident gasoline that contaminates the aquifer. Computationally, this situation is quite demanding to solve, and a streamline dependent spatial discretization was needed to suppress numerical dispersion (Janssen et al., 2006).

Figure 6 gives a representation of the behaviour of Figure 5, in terms of concentration distributions in the longitudinal direction. This result implies that the three linked concentration curves for initially resident degradable contaminant G (gasoline, or kerosene, for instance), an incoming (from the left) electron acceptor (EA such as oxygen) C, and resident, growing and decaying bacterial mass M, show patterns that for each time are directly related with each other. What is complicated is that the electron acceptor concentration distribution for all three times is different, and that between time 1 and 2 the injected electron acceptor seems to withdraw to the left again. This seemingly illogical result is purely the consequence of the increasing microbial mass when both substrate (G) and e.g. oxygen are present, as then, the consumption of G suddenly increases and its concentration decreases. As Figure 5 reveals, spatial variability of the hydraulic conductivity may strongly affect the transport of electron acceptor and the related degradation of resident contaminant. Erratic patterns may develop, that are difficult to communicate.

Whereas the complexity of the transport problem is profound, the strength of identifying the right robust features may inspire to equally profound simplifications. Considering the downward transport of a degrading contaminant towards the groundwater level, the mass balance equation for linearly adsorption and 1st order degradation reads:

$$\rho \frac{\partial q}{\partial t} + \theta \frac{\partial c}{\partial t} = \theta D \frac{\partial^2 c}{\partial x^2} - \theta v \frac{\partial c}{\partial x} - k\left(\theta c + \rho q\right) \tag{4}$$

Where q is adsorbed amount, c is concentration, k is degradation rate, v is pore velocity, D is dispersion coefficient, θ is volumetric water fraction, ρ is density.
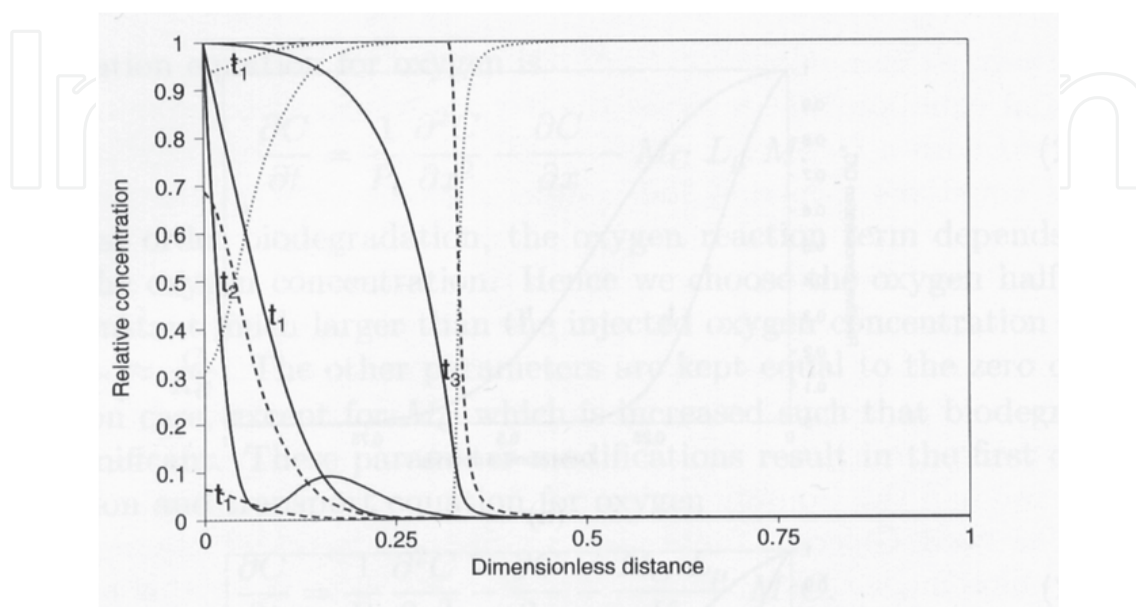

Fig. 6. Concentration distributions for electron acceptor (solid), contaminant (dotted), and microbial biomass, for times increasing in the direction of t_1 to t_3. Concentrations scaled with regard to their maximum concentration.

In essence, according to this transport equation, adsorption/desorption is instantaneous, and the degradation occurs (according to the last term) both in the liquid and in the solid phases. In terms of system properties, physical properties such as the hydraulic conductivity and water retention, control the development in time of the volumetric water fraction (θ) and flow velocity (v). Biogeochemical properties of importance are those related with the adsorption equation, s(c), e.g. affinity coefficient and a sorption maximum, and those related with the degradation rate, e.g. Monod kinetics parameters that describe the degradation rate as a function of the concentrations of contaminant, of electron acceptors or reductors, and microbial activity. Commonly, degradable contaminants are assumed to degrade according to first order kinetics. For the given transport equation, the transport and leaching process can be simulated with a computer model such as PEARL (Leistra et al., 2000). The PEARL model considers one-dimensional transient flow of water in the soil (vadose zone) profile, which is linked to the convection dispersion equation (CDE) for transport of chemicals. Besides that the model has modules for crop growth and energy and water interactions with the atmosphere, it is also possible to account for (monocomponent) adsorption/desorption and first order degradation. The PEARL model(see Beltman et al., 2008) is used in pesticide screening in the EU, for which the FOCUS scenarios have been developed ((FOCUS, 2000).). These scenarios harmonize the soil type, climate and weather variations within the EU for which the assessment of pesticide leaching to groundwater is evaluated, with several aims, among which the need to consider a limited number of modeling permutations, while still enabling comparison of results EU-wide.

5. Implications for management strategies

5.1 Simplified modeling approach

For decision making, we propose to first make an approximate assessment based on several simplifications. Thus, diffusion-dispersion is considered to be of secondary importance and flow is assumed to be steady state (as a first step). For groundwater quality protection, particularly the leached fraction of applied or spilled contaminant is of interest, because that quantity will control the concentration in the annual recharge of the topmost aquifer. For a particular year, both the leached fraction and the recharge (precipitation minus evapotranspiration) may be difficult to predict, which is an example of lack of ergodicity in time. However, if the interest is primarily for long term predictions as is the case with de-icing chemicals applied every winter, rather than for incidental spill events, then the uncertainty becomes considerably smaller. Regular leaching leads in that case to repetitive concentration jumps in the upper groundwater aquifer, and whether these jumps lead to a gradual building up of concentrations in groundwater, can be analysed according to the method of Beltman et al. (1996) and Van der Zee et al. (2010).

In those papers, we showed how the regular leaching of contaminant can result in a building up of concentrations in groundwater, depending on the capacity of the soil to degrade the contaminant: Figure 7. Whereas Beltman et al. (1996) focused on degradation and transport in an aquifer, Van der Zee et al. (2010) considered leaching from a mixed reservoir analogous of the unsaturated soil. In essence, the mathematical formulation does not depend on whether the first order kinetics of the contaminant transport and removal are due to transport processes or to degradation. Hence, in analogy to the approach of Van der Zee et al. (2010), we can consider an unsaturated soil volume, that looses contaminant to the groundwater. The quantity that is lost, we consider below, but if the lost quantity is equal to M and the volume of water in the unsaturated soil zone equals V, then

$$C = \frac{M}{V} \frac{\left[1 - \left(exp\left(-\frac{j\Delta t}{V}\right)\right)^n\right]}{1 - \left(exp\left(-\frac{j\Delta t}{V}\right)\right)} \tag{5}$$

For the maximum concentrations of the resulting saw tooth pattern of concentrations, where j is the water flux leaching from the unsaturated soil, n is the number of applications of the contaminant, and the time step is for one year. Observe that the minimum concentrations of this pattern are given by C-M/V.
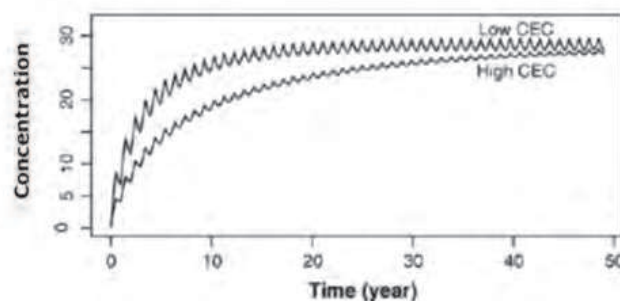

Fig. 7. Saw tooth patterns of concentration for an application of contaminant every year for two different values of CEC (cation exchange capacity), which controls concentration buffering. From: Van der Zee et al. (2010).

For a homogeneous soil, the leached fraction, *F*, according to the Convection Dispersion Equation is given by

$$F = \exp\left[\frac{-\mu RL}{v}\right]; \qquad R = 1 + \frac{\rho}{\theta}K_s \qquad (6)$$

if dispersion is ignored, where R is the retardation factor and K_s is the sorption coefficient for linear sorption. In this solution, we recognize the soil physical and atmospheric forcing controlled properties (θ, v), and the contaminant specific properties (K_s, and degradation rate parameter μ). Hence, a leaching vulnerability assessment system for the applicability of de-icing chemicals could involve a water flow and tracer leaching investigation, followed by a contaminant specific leaching investigation. In real soil systems, the various parameters of equation (6) vary in space and time, and this may have a large effect on the leached fraction. Therefore, Van der Zee & Boesten (1991) simulated leaching for the heterogeneous case. It appears that if there is a weak spot in the soil where leaching is relatively large and fast, then this one spot can contaminate a large volume of water up to the level of the water quality standard even if elsewhere no leaching of contaminant occurs. For moderate variability of soil properties, the leaching of contaminant in such a heterogeneous environment still behaves as 'first order degradation and transport', as described by equation (5), but the constants in this equation should be appropriately averaged. How this averaging should be done, focused on the leached fraction, is explained by Van der Zee & Boesten (1991).

Aimed at the problem of contamination with de-icing chemicals, it appears feasible to approach surface runoff also from a partly stochastic approach. Therefore, some first indication of ways to deal with irregular soil surfaces in the case of overland flow are presented using the framework of Appels et al. (2011). Using these approaches, it appears to be possible to give a stochastic analysis of the in situ aquifer bioremediation of degradable contaminants such as Propylene Glycol. Besides illustrating this aspect, many problems and unresolved questions are outstanding. For instance, mostly soil and groundwater formations are assumed to conform to Gaussian spatial structures, but in reality, more complicated structures that are distinctly non-Gaussian, may be more realistic.

5.2 Remediation in heterogeneous soils

Because transport of nonlinear reacting contaminants in spatially variable soils is a very active field of research, many problems are unresolved, yet managers need to make decisions, for instance concerning choice of remediation technique. For this reason, it is appropriate to indicate in what respect management decisions need to take into account this complexity. For instance, the applicability of the still recommended (e.g. by USA, EPA) air sparging as a methodology to purify groundwater that is contaminated with organic biodegradable or volatile contaminants can be judged well on the basis of the physics of this technique.

In air sparging, air is injected below the phreatic groundwater level, with the purpose to strip the aquifer from volatile contaminants, but also to increase the oxygen content in those strata as certain organic contaminants require it to become degraded. Much effort has been devoted to experimentally investigate how air will flow through the aquifer (upwards, as air is much less dense than water): in a narrow, vertical funnel or in a gradually upward broadening cone. The latter occurs if the aquifer material is sufficiently fine textured sand,

but finer, loamy or clayey textures cannot well be sparged in view of their air entry values. Engineers were particularly interested in the so called radius of influence (ROI), which is the radius of the supposedly circular cross section of the cone through which air flows at the phreatic groundwater level. In addition, air saturations in this cone are of interest.

A mathematical analysis has been worked out by Van Dijke et al. (1995) and was later refined by Philip (1998). In both cases, relatively simple solutions were obtained, but in both cases for a single cone and for a homogeneous aquifer. In a refinement, Van Dijke & Van der Zee (1998) considered a two layered medium, i.e., air was injected below the groundwater level, in a coarse layer that is overlain by a slightly finer textured layer. The contrast in physical properties of the two layers was very small (hydraulic conductivity differed only by a factor √2, i.e., hardly measurable). Even with such a small contrast in properties, the effect on the sparged area was major, as can be seen from Figure 8. It appears that at the interface between the two layers, the cone rapidly expands to larger radial distances away in a horizontal direction from the injection point, and air saturations rapidly decrease. Important is to note, that these calculations were done for an axially symmetric situation. The practical impact of these results is the following: if the air flow pattern is so sensitive to slight spatial (vertical) variability of the subsoil, for real subsoils with intricate spatial patterns in 3D, this must also be expected and (in view of variability) in an enhanced way. The air flow will be along a path of minimal resistance, and along this path, almost all flow will concentrate. Therefore, although injected air will oxygenate the subsoil, this will be a very inefficient way to do so, as flow is excessively preferential. Moreover, it cannot be well steered, to get the air to flow to a particular part of the subsoil, e.g. the contaminated part. In reality, the flow may follow many possible paths around a contaminated volume.
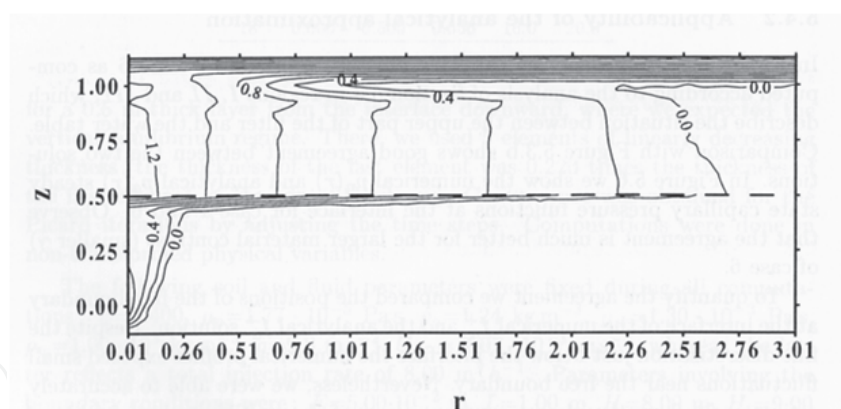

Fig. 8. Iso-concentration lines of air saturation for a two-layered medium with layer interface at dimensionless depth Z. Dimensionless radial distance is r (Van Dijke and Van der Zee, 1998).

In more general terms, spatiotemporal variability leads to uncertainty and therefore some suggestions can be given regarding soil/water contamination decision making under uncertainty, focused at the de-icing chemical contamination context.

6. Guidelines for decision making

Many contaminated sites and polluting surface activities rely on natural or manipulated remediation to remove pollutants in the soil. In order for this approach to be acceptable it is

important to have control of: (1) the pollution source, (2) the behaviour of pollutants in soils as influenced by spatial variability of the subsurface, (3) their removal rates (adsorption, degradation, precipitation) and how these can be optimised, and (4) long term effects on the soil system. In the above sections some of these aspects related to contaminants from winter maintenance in cold climate have been discussed, as well as different methods for subsurface characterisation and contaminant transport monitoring. The importance of dealing with heterogeneities in modelling strategy has also been dealt with.

6.1 Legal and political incentives

How contaminants are dealt with at contaminated sites in general, is not only determined by whether the scientific community can overcome present challenges as those discussed in the previous sections, but also within which political and legal framework contaminated site managers work under. Whether the optimal strategies for monitoring and cleanup are chosen depends on the following:

- A correct conceptual understanding of the processes which determine flow and transport of the contaminants, which is the prerequisite for correct choice of modelling tool to assist management at the site.
- The scientific knowledge, do we have the right methods and understanding of the processes to be able to deal with the contamination in a good way? which implies
  - Good methods exists for mapping and monitoring surface and subsurface heterogeneities, at present a combination of destructive and non-invasive techniques are recommended in combination with transport modelling.
  - Robust relationships exist between geophysical properties and hydrogeological properties and sufficient resolution of survey at the site.
  - Robust relationships exist between geophysical properties and contaminant situation in the soil, and sufficient resolution.
  - A sound method for coupling of different methods destructive and non-invasive methods, for quantitative description of contamination levels and remediation efficiency.
  - Appropriate remediation techniques for heterogeneous systems exist.
- Managers' knowledge about the system they are in charge of, the quality depends on:
  - Size of company, in view of available resources made available for such purposes.
  - Type of contaminant – available information about reactive properties
  - Company regulations and policies
- The authorities knowledge about contaminants, survey possibilities, modelling and risk factors, as well as their capacity to follow up contaminated sites and validate that accepted procedures are being followed.
- Legal international and national framework regulating expectations to contaminated sites.

6.2 Cold climate implications

Scientifically many processes are still not well described for low and freezing temperatures, such as physical conditions in freezing soil and how to model these, especially the combination of de-icing chemicals which lower the freezing point in a soil system near 0ºC is complicated. Degradation and biological dynamics as a function of low temperature is generally not well described. Instrumentation may work poorly under frozen conditions,

good methods for measuring the total water content in a frozen soil are for instance still lacking. What scale of heterogeneity is relevant during snow melting conditions compared to the thawed situation? Cold climate conditions add extra complications to the contaminant site management, for one; the need for use of de-icing chemicals. Since the use is generally not regulated, there is a great potential for reduced environmental impacts by implementing such limitations. At Oslo airport, Gardermoen, only a certain amount of de-icing chemicals is allowed per winter season. De-icing chemicals that are highly concentrated such as at the de-icing platforms are collected, hence reducing the amount released to the green areas. Since soil temperature has a large effect on surface infiltration patterns, this should be monitored as part of the regular monitoring programme along roads and at airports. Strategies to deal with both frozen and non-frozen conditions during snowmelt must exist, e.g. pump and store contaminated melt water in local depressions for later infiltration when conditions are more optimal, e.g. in summer.

7. Conclusion

This chapter gives an overview of properties of de-icing chemicals and the processes that affect their fate in the unsaturated zone. Inorganic salt concentrations can only be reduced by dilution and is only discussed briefly here, while degrading organic de-icing chemicals are the main focus. Conventional methods of soil and soil water samples are still required for interpretation of emerging technologies such as different geophysical techniques. The importance of heterogeneity of the infiltration process and unsaturated flow is emphasised. The method of spatial moments is an efficient way of describing average movement of contaminants in the unsaturated zone. An ergodic situation, i.e. when the size of the contaminant source is much larger than the scale of soil heterogeneity, makes average assumptions more certain while small plumes in a system of large variability causes great uncertainty. Geophysical methods can be used in time lapse mode to obtain a more continuous impression of the flow and transport processes and is recommended in combination with point measurement for quantification of absolute levels of contaminants. Flow and transport modelling within a stochastic framework is another important complementary technique for optimised management strategies at contaminated sites.

8. Acknowledgement

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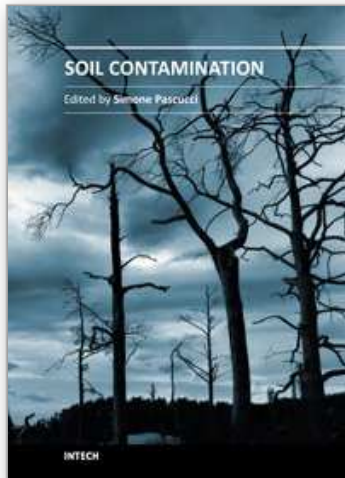
Soil Contamination

Edited by MSc Simone Pascucci

Soil contamination has severely increased over the last decades, mainly due to petroleum hydrocarbons, solvents, pesticides, lead and other heavy metals from industrial wastes and human activities. The critical point regarding contaminated soil monitoring is the intrinsic difficulty in defining fixed monitoring variables and indicators as the establishment of any a priori criterion and threshold for soil quality can be still considered subjective. This book is organized into eight chapters and presents the state-of-the art and new research highlights in the context of contaminated soil monitoring and remediation strategies, including examples from South America, Europe and Asia. The chapters deal with the following topics: - monitoring of dioxin, furan, hydrocarbons and heavy metals level in soils - bioindicators and biomarkers for the assessment of soil toxicity - use of reflectance spectroscopy for soil contaminants and waste material detection - remediation technologies and strategies.

How to reference

In order to correctly reference this scholarly work, feel free to copy and paste the following:

Helen K. French and Sjoerd E.A.T.M. van der Zee (2011). Research for Investigating and Managing Soil Contamination Caused by Winter Maintenance in Cold Regions, Soil Contamination, MSc Simone Pascucci (Ed.), ISBN: 978-953-307-647-8, InTech, Available from: http://www.intechopen.com/books/soil-contamination/research-for-investigating-and-managing-soil-contamination-caused-by-winter-maintenance-in-cold-regi

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