Derivation of Matrix Soil Standards for Salt under the British Columbia Contaminated Sites Regulation

Addendum A:

Technical Options Analysis for a Soil Chloride Standard for Drinking Water Protection (Aesthetic)

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Introduction

When deriving risk-based environmentally protective benchmarks for the concentrations of anthropogenic substances in soils, issues occasionally arise for specific substances and scenarios which require that previously established generic guidance be re-visited. Established policy guidance and protocols for deriving "soil matrix standards" under the British Columbia Contaminated Sites Regulation are documented primarily in a report by the British Columbia Contaminates Sites Soils Taskgroup (CSST, 1996) entitled "Overview of CSST Procedures for the Derivation of Soil Quality Matrix Standards for Contaminated Sites" (January 1996).

Recent efforts to develop provisional soil matrix standards for salt ions under the BC CSR are documented in Bright and Addison , (2000, 2002). As part of this exercise, we calculated the soil concentrations of chloride above which contamination of groundwater, and –hence- drinking water above an aesthetic threshold may occur, according to the prescribed procedures. Such procedures, while technically defensible, would result in a soil remediation standard applicable at sites where the drinking water matrix standard applies which is sufficiently stringent that few other practical options would exist other than to undertake a detailed human health risk assessment. It was felt that the calculated soil threshold for chloride ions would mean that very few of the existing salt-contaminated sites in the province (possibly less than 10% to 20%) could be remediated using the generic matrix standards.

We re-visit herein the derivation of a soil chloride standard based on drinking water protection, in light of the anticipated importance of this issue within the province of British Columbia. Any comment on key policy decisions under the British Columbia Contaminated Sites Regulation and Waste Management Act is beyond the scope of this document. Rather, this is intended as a scientific/technical document that addresses the effect of various aspects of scientific uncertainty on the calculated provisional soil standard for drinking water protection. This addendum is complimentary to and should be interpreted in concert with the larger salt matrix standard derivation document (Bright and Addison, 2002). Specific objectives of the addendum are to -

- Further document the sensitivity of the draft matrix standard for chloride in soils for drinking water protection to assumed site properties, and descriptors of subsurface fate of chloride ions; and,

- Provide the results of various alternative scenarios for major site groups based on climatic and soil texture variations.

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Back-Calculation of Soil Thresholds for the Protection of Drinking Water Quality: The Major Components.

Chloride Concentration Thresholds in Drinking Water

Chloride ions in groundwater aquifers are important from an aesthetic rather than human toxicological perspective. There is no known upper limit of human exposure to chloride ion associated with any adverse health condition (chloride is deemed to be virtually non-toxic to human beings). The presence of chloride ion in drinking water supplies in excess of 250 mg/L, however, represents an "aesthetic" threshold, beyond which the strong odor and taste may render the groundwater unusable as a drinking water supply. The aesthetic-based drinking water quality guideline is uniformly recommended in a variety of international, Canadian (CCME, 1999) and provincial jurisdictions, including British Columbia. It represents, therefore, a widely accepted aesthetic drinking water threshold for managing aquifers that may be important present of future sources of drinking water.

The World Health Organization (WHO) in 1999 re-visited the chloride aesthetic drinking water guideline. According to WHO –

> "The 1958 WHO *International Standards for Drinking-water* suggested that concentrations of chloride greater than 600 mg/litre would markedly impair the potability of the water. The 1963 and 1971 International Standards retained this value as a maximum allowable or permissible concentration. In the first edition of the *Guidelines for Drinking-water Quality*, published in 1984, a guideline value of 250 mg/litre was established for chloride, based on taste considerations. No health-based guideline value for chloride in drinking-water was proposed in the 1993 Guidelines, although it was confirmed that chloride concentrations in excess of about 250 mg/litre can give rise to detectable taste in water. At the meeting to plan the revision of the Guidelines, held in Berlin in June 2000, it was concluded that the guideline document did not need to be revised, as there were no new data of significance."

A groundwater concentration of 250 mg/L chloride ion is accepted herein as a targeted upper concentration limit for drinking water based on leaching from salt contaminated soils.

Predicted Soil Chloride Concentrations Based on the Drinking Water Guideline

Appropriateness of the Model

The BC WLAP groundwater model (adapted from Domenico and Robbins, 1994) was previously reviewed in terms of the modeled fate processes used to describe salt ion movement in a homogenous subsurface environment. The generic assumptions adopted through CSST (1996) policy - including an unlimited (non-depleting) contaminated soil

source (30 m wide by 5 m long by 3 m deep), leaching directly into groundwater at the interface between the saturated and unsaturated zone (assumed to occur at 3 m depth) in very permeable sandy, deltaic soils are undoubtedly conservative. They are nonetheless deemed to be appropriate given that generic standards should provide for adequate groundwater protection at any site in B.C. in which they are applied. Such assumptions can be revisited to calculate "Site Specific Standards". Furthermore, the main components of the model, including leachate generation driven by infiltration, introduction at the groundwater surface, lateral (advective) transport, and changes in the mixing layer (via advection and dispersion) at the bottom of the contaminated groundwater plume, were deemed to adequately describe groundwater flow, and – by extension – the subsurface fate of substances such as chloride ion that move conservatively with the groundwater.

It was previously determined that modeling calculations for chloride, using the BC WLAP model and default assumptions, were very sensitive to the assumed magnitude of K_d – the "adsorption coefficient" as an empirical estimate of the relative tendency of the modeled substance to partition into the migrating groundwater as opposed to the surface of soil particles in the saturated zone. Predictions were also somewhat sensitive to the assumed amount of annual infiltration (precipitation minus runoff and evaporation).

The modeled soil concentration for chloride ion was relatively insensitive to assumptions regarding the distance between the contaminated soil mass of point of exposure (for the purpose of drinking water extraction, or within a surface water body).

We were particularly interested in whether the further categorization of site types based on soil texture [coarse versus fine grained, or low versus high Water Holding Capacity (WHC)] or broad regional differences in meteorological conditions (especially number of frost-free days) would be appropriate. The sensitivity of model predictions to soil texture vis-à-vis soil lateral hydraulic conductivity (as Darcy's velocity) and site meteorological conditions is discussed below.

Chloride Adsorption Co-efficient (K_d)

Adsorption co-efficients are not part of the intrinsic properties of a specific substance. Rather, they are site-specific values that may vary across soil types and – to a lesser extent – other site conditions. As such, they are often estimated through empirical studies. Initial attempts to find published, peer-reviewed K_d values for chloride ion were not successful.

According to Krupka *et al.* (1999) –

> "Strenge and Peterson (1989) used 9 categories defined by soil pH and texture in the Multimedia Environmental Pollutant Assessment System (MEPAS) code. The 3 soil texture classes were <10 percent, 10 to 30 percent, >30 percent clay/organic matter/oxide content. The 3 pH classes were >9, 5 to 9, and <5. The 9 cells

defined by the pH and soil texture classes contained literature-derived K_d values and where data was not available, estimated values were included in the table. The inorganic contaminants in the K_d look-up table were actinium, aluminum, americium, antimony, arsenic, asbestos, barium, beryllium, borate, cadmium, calcium hypochlorite, calcium oxide, carbon, cerium, chlorate, chromium (III), chromium (VI), cobalt, copper, curium, europium, fluoride, hydrogen fluoride, iodine, iron, krypton, lead, lead oxide, lithium hydroxide, lithium ion, magnesium, manganese, mercury, molybdenum, neptunium, nickel, niobium, nitrate, nitric acid, nitrogen dioxide, palladium, phosphate ion, phosphorus,, plutonium, polonium, potassium hydroxide, potassium ion, protactinium, radium, ruthenium, samarium, selenium, silicate ion, silver, sodium ion, strontium, sulfate, sulphur, thallium, thorium, tin, tritium, uranium, vanadium, yttrium, zinc compounds, zinc, and zirconium.

For any literature-derived K_d value, it is essential to clearly understand the selection criteria and the logic used to estimate K_d values not found in the literature. For instance, Strenge and Peterson (1989) reported a wide range of literature K_d values for several cells, typically greater than 10-fold and sometimes greater than a 100-fold difference between minimum and maximum values.

The excellent review provided by Krupka et al (1999) serves to illustrate two points. First, there is a general lack of K_d estimates for chloride ions in various soil types. Second, such estimates, if available, are likely to be highly variable, and very much dependent on the experimental conditions and calculation method.

It is generally considered that chloride ions are conservative tracers of groundwater movement, and in most soil types it is generally accepted that chloride ions exhibit very little if any potential to interact ionically or otherwise with the surfaces of soil particles in the saturated zone. The K_d for chloride, therefore, is often assumed to be zero ml/g.

We noted earlier (Bright and Addison, 2002) that some United States regulatory agencies recommended use of a chloride ion K_d of 0.1 ml/g, toward the modeling of subsurface fate of radionuclides, using the RESRAD model.

The interaction of chloride with soil particles, leading to limited retardation relative to groundwater flow velocities is theoretically possible, as would be a site-specific K_d value that departs from zero. In the absence of a reasonable scientific knowledge base, it is assumed that coarse, low organics soils (sands and coarse glaciofluvial materials, for example) would exhibit very limited ability to retain chloride ions. Soils that include less than 10% by weight of soil fractions with a least some potential to transiently retain chloride ions (clays, organic matter, some complex oxides) are likely to exhibit chloride K_d values of zero. On the other hand, it is conceivable in the absence of better scientific information that medium-grained (10 to 30% clay/organic matter/oxide content) to fine (> 30% clay/organic matter/oxide content) to fine-grained soils would exhibit K_ds that could approach 0.1 ml/g or higher. Given the uncertainty in these estimates, experimental

analysis of chloride ion K_ds would be invaluable in allowing more accurate model predictions, using methods provided in Krupka *et al.*, 1999.

The model predictions *vis-à-vis* soil concentrations that are protective of groundwater and surface water uses and ecological functioning are sensitive to assumptions about the chloride dissociation constant. Using the CSST default assumptions (Bright and Addison, 2002), previous estimates of a provisional chloride concentration in soil for the protection of drinking water (≤ 250 mg/kg Cl$^-$) were as follows:

Table 1: Relationship between assumed chloride adsorption coefficient and predicted soil quality threshold for protection of drinking water.

K_d	soil quality threshold (mg/kg Cl$^-$)
0	47
0.05	89
0.1	130
0.2	210

(from Bright and Addison, 2000; Table 6.23).

Assumed Site Properties

CSST default assumptions (Annex A) specify a near worst-case situation for the mobility of various organic and inorganic contaminants in groundwater. The generic case used in modeling exercises is based on a highly permeable, homogenous sandy soil with low organic carbon content.

We are not concerned herein with those aspects of the model that predict fate in the unsaturated zone (Henry's law constant). There is no potential for salt biodegradation, so the environmental half-life in both saturated and unsaturated zone is zero. The organic carbon content is not relevant to subsurface fate of ionic as opposed to non-ionic hydrophobic contaminants. For ionic substances such as chloride, the soil and groundwater pH is generally a very important factor that influences adsorption/desorption, ion exchange, and - hence – K_d estimates. Since no information was available however on the pH dependence of chloride soil adsorption, it is assumed to be uniform across the range of pHs encountered.

Important site aspects that both have the potential to vary substantially across sites in British Columbia and might theoretically influence chloride fate in the saturated zone include –

- Saturated zone groundwater velocity as estimated from the Darcy Velocity.
- Infiltration rate.
- Number of frost free days.

Other site aspects such as the source dimension, depth of the unsaturated zone, and depth of the unconfined aquifer might be important determinants of chloride ion travel time and predicted concentration at any given point in space and time. These properties are highly variable across all the candidate application sites in B.C., however, and the rationale for simplifying assumptions for the generic case have been thoroughly discussed previously.

When considering soil texture and allied properties, it is important to appreciate that some of the model inputs are linked from a theoretical perspective.

Table 2: Linked variables in the BC WLAP groundwater model

Suite 1 source soil volume		Suite 2 climatic conditions		Suite 3 subsurface environ.	
X	source dimension length	**P**	precipitation rate	**x**	distance from source to receptor
Y	source dimension width	**(RO + EV)**	run-off and evaporation	**n**	contaminated soil porosity
Z	source dimension depth	**D1/2US**	days when ground surface temp is below 0° C.	**nu**	water-filled porosity
				ne	effective porosity
				foc	soil org. C. fraction
				V	Darcy velocity in saturated zone
				d	depth to unconfined aquifer
				da	depth of unconfined aquifer
				ρb	soil dry bulk density
				pH(s)	soil pH
				pH(gw)	groundwater pH

Source: CSST, 1996.

The modification of assumptions about the first suite (source volume) or second suit (climatic conditions) of linked variables does not present in technical challenges. The modification of soil texture for the subsurface environment (suite 3) requires that adjustment to the Darcy Velocity, for example, be accompanied by corresponding changes to assumptions about soil porosity, since both would be expected to be lower in fine grained soils.

Assumptions used for a coarse-grained (<30% of soil passing through a 75 μm sieve) or fine-grained site, as used for the derivation of the Canada-Wide Standards for Petroleum Hydrocarbons (PHC CWS) are provided in Table 3.

Table 3: Generic site assumptions used to derive Canada-Wide Standards for Petroleum Hydrocarbons

Parameter	Units	Default CSST Value	PHC CWS Values, as recommended by PIWG and HHFT TAG[A]	
			coarse textured	fine textured
Contaminant Source Width	m	30		
Contaminant Source Depth	m	3	(3) [B]	(3) [B]
Contaminant Source Length	m	5	10 [C]	10 [C]
Distance to Receptor	m	10		
Precipitation	m/yr	1.000		
Runoff & Evaporation	m/yr	0.454	(0.72) [D]	(0.80) [D]
Precip. minus Runoff and Evap.	m/yr		0.28	0.20
Depth to Groundwater (water table)	m	3.0	(3) [E]	(3) [E]
Half-life in unsaturated zone	days	substance specific	infinite (set at 1E+09) [H]	infinite (set at 1E+09) [H]
Partition Coefficient, K_{oc}	mL/g	substance specific		
Weight fraction of organic carbon in soil, f_{oc}	[/]	0.006	0.005	0.005
H_2O-filled porosity (unsaturated)	[/]	0.1	0.119	0.168
Air filled porosity (unsaturated)	[/]	0.2	0.281	0.132
Henry's Constant = H*42.3	[/]	substance specific		
Days with surface temp. < 0 deg. C	days	0		
Darcy velocity in saturated zone	m/yr	12.6	16	0.79
Depth of unconfined aquifer	m	5	(5) [F]	(5) [F]
Total porosity (saturated)	[/]	0.3	0.4	0.3
Effective porosity (saturated)	[/]	0.2	(0.4) [G]	(0.3) [G]
Soil bulk density	g/cm^3	1.74	1.7	1.4
Maximum solubility of contaminant	mg/L	substance specific		
Half-life in saturated zone	days	substance specific	infinite (set at 1E+09) [H]	infinite (set at 1E+09) [H]

Shaded values indicate a chemical specific parameter for which CSST has not supplied default values.

Additional Notes: (A) Where no determination was made by HHFT TAG and/or PIWG, the CSST defaults were applied provided that they were reasonable estimates; (B) Not explicitly defined as part of HHFT modeling calculations; however, the model assumed intimate contact between the top of the groundwater table and the PHC contaminated soil mass. This is also consistent with CSST default assumptions; (C) Not required for HHFT modeling calculations. However, the original publication by Domenico on which the PIRI toolkit model is based makes mention of a 'width' of 10 m in the direction of groundwater flow; (D) Not set as part of human health-based calculations; however, "precip. minus runoff and evaporation" was set at 0.28 m/yr for coarse-grained soils and 0.20 m/yr for fine-grained soils; (E) 3 m based on discussions between Mah-Paulson and Bright, in recognition of the nature of the generic site scenario; (F) set at 2 m by PIWG as a conservative assumption which would minimize dilution; however, set a 5 m herein, since this was the minimum allowable distance within the BCE model; (G) For purpose of the PHC CWS, it was assumed that the effective porosity is 100% of the total porosity; (H) The issue of biodegradation in the subsurface environment was subsequently revisited. After extensive deliberations, the t1/2 for CWS F1 and F2, respectively was established as 712 d and 1,750 d for both the saturated and unsaturated zones. These were chosen as being highly conservative values.

Keys (pers. com.) suggesting using the categorization of soil types based on "Water Holding Capacity" (WHC) as opposed to percent fines. WHC is required as a measure enroute to analyzing salt ion concentrations using the saturated paste method. The application of soil texture under the PHC CWS requires an additional analytical procedure. This is worth pursuing provided that WHC can be confidently related to Darcy Velocity and soil porosity.

The groundwater model was run after re-visiting assumptions about site characteristics and chloride ion K_d. The implications of the previous assumptions are itemized below.

Case 1:

Assumptions

Site-properties:	CSST Default
Number of days with frost:	0 (CSST Default)
Chloride adsorption cofficient:	Kd = 0 ml/g
Drinking water guideline:	250 mg/L Cl⁻
Predicted soil conc.:	**48 mg/kg**

Case 2:

Assumptions

Site-properties:	PHC CWS fine-textured (Table 3)
Number of days with frost:	0 (CSST Default)
Chloride adsorption cofficient:	Kd = 0 ml/g
Drinking water guideline:	250 mg/L Cl⁻
Predicted soil conc.:	**81 mg/kg**

As can be seen from case 2, a large decrease in the groundwater velocity is not predicted to substantially decrease the allowable chloride soil concentration. The model is run iteratively until an equilibrium solution is achieved. While this would be expected to take far longer for contaminated plume migration in fine grained soils, similar downgradient concentrations would nonetheless be expected to be achieved in fine-textured and coarse-textured soils, since chloride ion is conserved and since the potential for dilution through dispersion at the plume margins would be decreased in proportion to the decreased groundwater flow velocity. For contaminants that can be biodegraded, the additional travel time would allow for a much greater attenuation along the transport pathway, and hence higher acceptable concentrations at the soil source area.

The relative unimportance of Darcy Velocity is further illustrated in Figure 1. For this example, the CSST default assumptions were used along with a K_d of zero, except that the Darcy Velocity was varied.
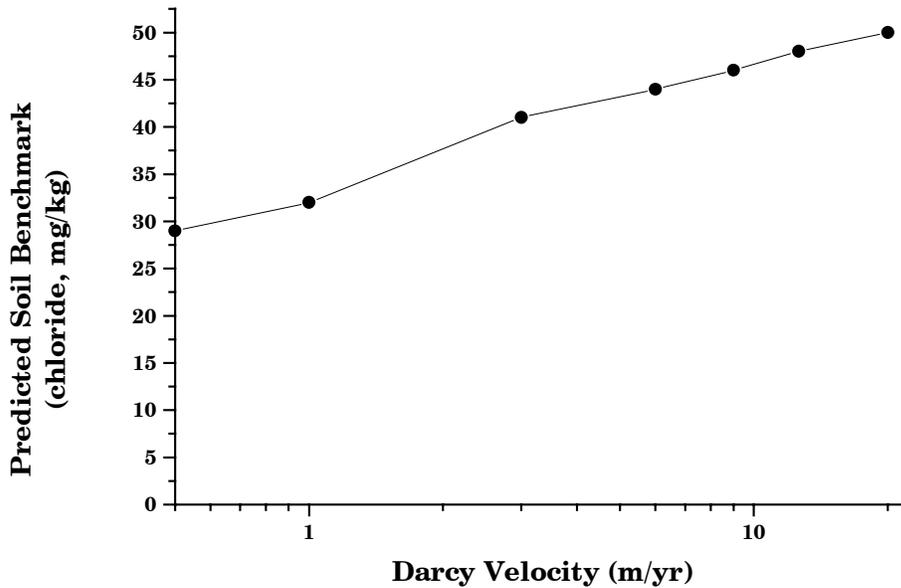
Figure 1: Sensitivity of chloride modeling predictions to Darcy Velocity

For the CSST as opposed to PHC CWS assumptions, the predicted acceptable soil chloride concentration actually decreased in relation to decreasing assumed Darcy Velocity, although it should be noted that effective porosity and other soil aspects were not adjusted to reflect actual fine-grained scenarios. This has further implications for the viability of using site-specific calculations for groundwater related issues at salt contaminated sites.

In a fine-grained soil, it may be reasonable to assume a chloride K_d that departs from zero:

Case 3:

      Assumptions

Site-properties:	PHC CWS fine-textured (Table 3)
Number of days with frost:	0 (CSST Default)
Chloride adsorption cofficient:	Kd = 0.1 ml/g
Drinking water guideline:	250 mg/L Cl⁻
Predicted soil conc.:	**149 mg/kg**

The difference between this prediction and the CSST default case is almost entirely attributable to differences in the assumed chloride K_d.

Given the relative insensitivity of the groundwater model to hydraulic conductivity in the case of chloride ion, a sensitivity analysis was carried out for infiltration (Figure 2) and for the number of frost-free days (Figure 3), within a reasonable range.

In the case of infiltration, the ratio of precipitation to runoff and evaporation was maintained at the same ratio of the default CSST assumptions (= 1 m/yr precip:~0.5 m/yr ro+evap), but the actual infiltration rate was varied from 0.05 to 2 m/yr by adjusting the precipitation rate.
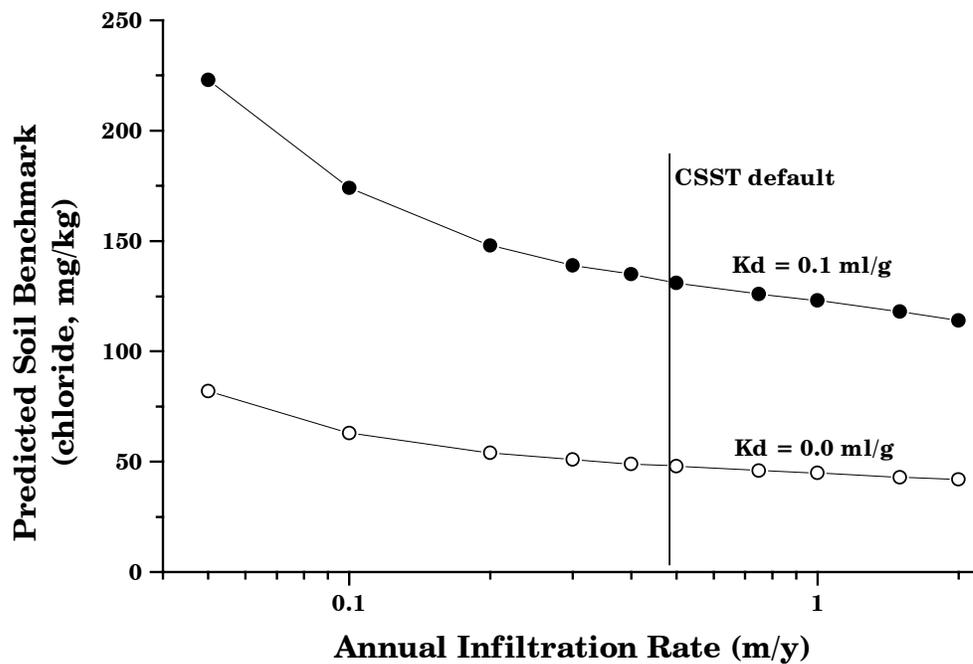

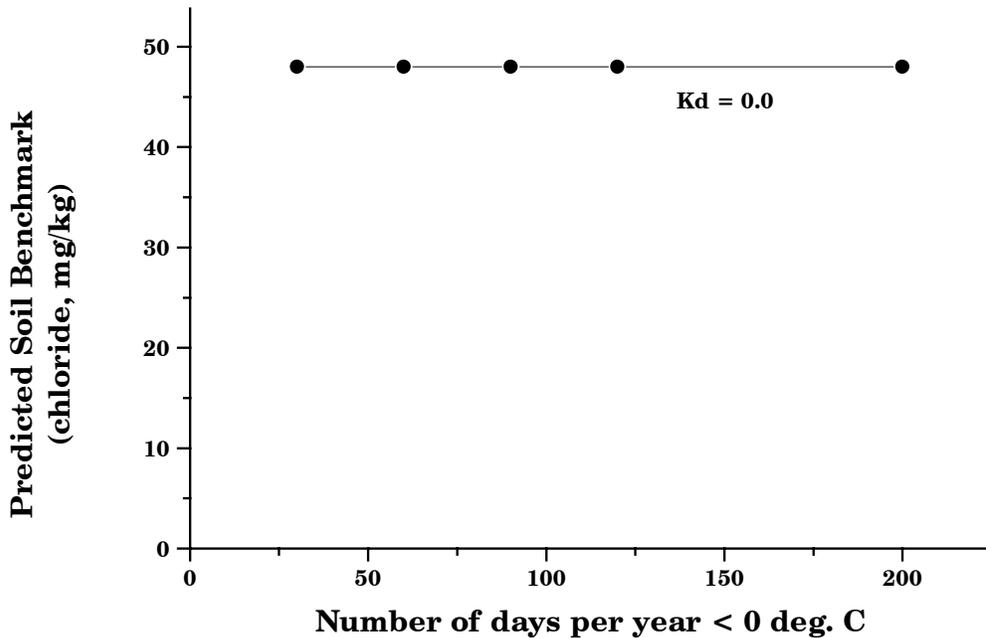**Figure 2: Model sensitivity analysis: Annual Infiltration Rate**

Figure 3: Model sensitivity analysis: No. of Days Annually with Frost

As illustrated in Figure 2, the predicted soil concentration for the protection of drinking water is relatively insensitive to changes in the assumed annual infiltration rate.

Predicted protective chloride concentrations in soils did not vary at all in relationship to the assumed number of days in an average year when the ground is frozen. *Why?* The number of days in a year, like the Darcy Velocity, primarily affects the number of days or years it would take for the maximum contaminant concentration in the leachate plume to reach a specified point (in these cases, a distance of 10 m laterally to the salt-contaminated soil mass). In the case of biodegradable contaminants, this would also be expected to influence extent of degradation, since this is a time-dependent phenomenon. In the case of salt, however, few mechanisms exist that would contribute to a decrease in concentration along the flow path of the contaminated groundwater plume, and the influence of all of these (e.g. dispersive mixing at the plume boundary) would be expected to be equivalently affected when groundwater movement is restricted in frozen ground, or when overall groundwater transport rates are modified.

Modification of assumptions regarding the depth of the unconfined aquifer did not alter model predictions, an expected result since this would not be expected to influence plume dispersion or the mixing zone depth, except in very shallow aquifers which are poorly described by the majority of theoretical models.

Similarly, any changes in the assumed depth of the unsaturated zone do not result in changes in the back-calculated soil protective number. The major controlling influences are likely to be leachate concentration in and immediately beneath the soil source area. Any separation between the point of leachate generation and surface of the water table will undoubtedly increase the vertical travel time, but not the final equilibrium concentration along the vertical migration path, enroute to the saturated zone, assuming no source depletion (an infinite source term). Again, there are no mechanisms in the model that would allow for loss of chloride ions in the unsaturated zone, nor any major theoretical reasons based on current scientific knowledge to believe that might be important. The concentration of salt ions introduced directly to groundwater when the contaminated soils directly overly it and are in direct contact with it would be expected to be the same if there were greater vertical separation, given enough time.

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Recommendations

In light of the preceding analysis, it is recommended that the provisional matrix standard for salt include a fine-grained generic case for the purpose of calculating risk-based soil chloride concentrations based on groundwater-mediated transport and/or exposure.

The sole reason for considering a generic fine-grained case is that there is limited reason to believe that chloride adsorption coefficients (K_d) could reasonably depart from zero.

In the case of fine grained soils, and assuming that for these types of sites a K_d of 0.1 ml/g adequately describes chloride associations with soil particles in the saturated zone, the following groundwater-based soil thresholds are estimated:

> **Soil Quality Threshold for Protection of Drinking Water (chloride –aesthetic guideline = 250 mg/L):**　　　　　　　　**150 mg/kg Cl⁻**
>
> **Soil Quality Threshold for Protection of Aquatic Life (chloride –provisional water quality guideline = 460 mg/L):**　　　　　　**3,800 mg/kg Cl⁻**

A necessary pre-condition for the implementation of such site categorizations under the BC CSR matrix standards would be the creation of a scientifically defensible definition of fine-textured versus coarse-textured sites, along with specific data collection requirements during site characterization to allow the supported use of categorical designations.

Above all, the least certain of all assumptions used in deriving risk-based soil standards for chloride based on groundwater-mediated exposure revolve around predictions of the partitioning behaviour of the chloride ion. As a minimum, real site data (with both well characterized soil and groundwater chloride concentrations) should be assessed as a measure of the validity of this derivation exercise.

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Annex A: CSST Default Groundwater Modeling Assumptions

Parameter		Definition (units)	Default
S	=	maximum solubility	chemical specific1
n	=	total porosity (dimensionless)	0.3
nu	=	water filled porosity	0.1
na	=	air filled porosity (dimensionless):	calculated value
		na = n - nu	0.2
Pb	=	dry bulk density of soil (g/cm3)	1.75
H	=	Henry's Law constant	chemical specific1
H'	=	dimensionless Henry's Law constant	chemical specific
	=	dispersivity in unsaturated zone	calculated
foc	=	fraction of organic carbon in soil	0.006
V	=	Darcy velocity in saturated zone (m/yr)12.6	
Z_d	=	thickness of mixing zone (m)	0.5
K_d	=	distribution coefficient for a chemical (cm^3/g)	chemical specific
K_{oc}	=	organic carbon partitioning coefficient (cm^3/g)	chemical specific
dx	=	dispersivity in x-direction	calculated
dy	=	dispersivity in y-direction	calculated
d	=	unconfined groundwater aquifer thickness (m)	5
b	=	thickness of the unsaturated zone below source (m)	0
d	=	depth from surface to uncontaminated groundwater surface (m)	3
x	=	distance from source to receptor (m)	10
	=	effective porosity (dimensionless)	0.2
t1/2 US	=	decay (biodegradation) half-life at unsaturated sites	chemical specific
t1/2 S	=	decay (biodegradation) half-life at saturated sites	chemical specific
I	=	infiltration rate (m/yr) Note: (1-0.45)	0.55
P	=	precipitation rate (m/yr)	1
(RO + EV)	=	runoff rate plus surface evapotranspiration rate (m/yr)	0.45
X	=	source dimension length (m)	5
Y	=	source dimension width (m)	30
Z	=	source dimension thickness (m)	3
D1/2US	=	frost free days	365