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Cl distribution in different terrestrial habitats along hill slope gradients in Forsmark

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Update notice

The original report, dated December 2021, was found to contain factual errors which have been corrected in this updated version. The corrected factual errors are presented below.

Updated 2023-08

Location	Original text	Corrected text
Page 6, Introduction	New paragraph added	This report is focused...
Page 35, References	New reference added	Svensson et al. 2023

Contents

1 Introduction

The view of chlorine in nature is undergoing a major change as a result of the recent decades of research. It is now clear that chloride (Cl^-), that was previously considered non-reactive and the totally dominating Cl species, instead is reactive and does not always constitute the predominant form of chlorine (Cl) (e.g. Bastviken et al. 2013, Svensson et al. 2007). Extensive natural chlorination of organic matter occurs in many terrestrial ecosystems (Gustavsson et al. 2012, Redon et al. 2011). Experiments with radioactive Cl (^{36}Cl) as tracer have confirmed natural chlorination rates corresponding to as much as 50–300 % of the annual wet deposition of Cl in several types of soils (Bastviken et al. 2007, 2009). Substantial chlorination of organic matter occurs in a wide range of agricultural and forest soils (Gustavsson et al. 2012, Redon et al. 2013). The estimates are based on bulk soil excluding roots, which is likely an underestimation of the chlorination potential as it was later shown that the majority of chlorination occurs in the rhizosphere (Montelius et al. 2019). Available evidence indicates that 29–100 % of Cl in soil is organically bound (Cl_{org}) (Bastviken et al. 2013, Redon et al. 2011). Therefore, it is not surprising that Cl_{org} in soils frequently exceeds the level of Cl^-. This dominance of Cl_{org} in soils has been observed in several different types of soils. A study in France reported that > 80 % of the total chlorine was Cl_{org} across grassland, arable and forest soils (Redon et al. 2013).

Earlier studies have primarily focused on Cl_{org} and Cl^- in forest soils and a large variation in the concentrations of organically bound Cl has been reported in humus; Johansson et al. (2003a) found Cl_{org} concentrations of 32–2 100 µg g^{-1} in Southern Sweden, whereas Redon et al. (2011) reported a range of Cl_{org} concentrations of 34–687 µg g^{-1} in France. The variation at large and coarse spatial scales has been correlated to atmospheric Cl^- deposition, and soil organic matter content (Gustavsson et al. 2012, Johansson et al. 2003a). However, at local levels correlations to other factors may be more important. For example, Johansson et al. (2003b) and Redon et al. (2011) observed higher Cl_{org} concentrations in forest soil dominated by coniferous trees than in areas with deciduous trees, and this pattern was later experimentally confirmed by Montelius et al. (2015).

Chloride is rapidly taken up by plants in amounts that may exceed what is needed for growth, though the reason for this excess uptake is unknown (White and Broadley 2001). Cl^- tends to accumulate in plant tissue and the concentrations in plant biomass may be 1.5 to 305-fold higher than soil-water concentrations for agricultural plants (Kashparov et al. 2007a, b, Marschner 2012). Henner et al. (2013) suggested that the vegetation (agricultural species such as potato, spring wheat, radish, green bean) uptake of Cl^- was primarily driven by an increase in biomass. Montelius et al. (2015) investigated Cl cycling in different tree species and found very high Cl^- leaching rates for some tree species, in particular for Norway spruce. As this chloride was clearly not incorporated in biomass, it indicates excess uptake. Soil planted with coniferous trees in this study showed a particularly high chlorine accumulation and it was clear that internal circulation of chlorine was large in some of the experimental forest plots. Thus, extensive tree uptake and subsequent leaching back to the forest floor contributed to retaining Cl in the system. There are also other indications of a rapid accumulation of chlorine in green plant parts (Tröjbom and Grolander 2010). Moreover, the existing studies indicate very large differences in Cl storage among different plant communities and habitats (Johansson et al. 2003b, Montelius et al. 2015, Redon et al. 2011).

The above variability has consequences for Cl residence times at the landscape scale, as Cl can be retained as Cl_{org} in surface soils or Cl^- in biomass and there seems to be a continuous cycling between these Cl pools (Bastviken et al. 2013). Montelius et al. (2015) could link the 30 years accumulation of Cl_{org} in forest soils directly to the type of forest vegetation. Thus, to estimate overall landscape Cl cycling and residence times, it is necessary to characterize Cl^- and Cl_{org} uptake and storage in the most common habitats and vegetation communities. This is relevant for several reasons, including the use of Cl^- for estimating catchment hydrology (Kirchner et al. 2010) and for risk assessment modelling associated with ^{36}Cl in nuclear waste (Limer et al. 2009).

Studies published to date have primarily focused on relatively dry recharge areas, such as upland forests and agricultural land, and little is known of Cl cycling in productive discharge areas and other wetland habitats. Such areas are of particular interest to determine the fate of chlorine reaching surface ecosystems via discharge of deep groundwater.

The aim of this study is to analyze, based on two field campaigns, the distribution of Cl^- and Cl_{org} in vegetation and soil in the Forsmark area along elevation gradients, ranging from upland coniferous forest to wet alder forest and mires. This design thereby incorporates habitats with discharge areas, while also allowing comparisons with the nearby upland areas. The Cl characteristics and related biotic and environmental factors, such as the composition of dominating plant species, biomass, distance to groundwater and soil type, were assessed to enable a search for relationships and discussions of possible mechanisms behind the chlorine cycling and plant uptake.

This report is focused on description of raw Cl data. Additional statistical analysis and an in-depth discussion of the results are presented in Svensson et al. (2023). The article also includes a mass-balance modelling and presents habitat specific Cl fluxes and transfer rates between soil and biomass pools.

2 Material and methods

2.1 Study area

The area chosen is located in the Forsmark area, Eastern Sweden. The corrected average yearly precipitation (2003–2010) is 584 mm and average yearly temperature is +6.7 °C (Werner et al. 2014). The landscape is dominated by conifer forest (primarily Scots pine, *Pinus sylvestris*, and Norway spruce, *Picea abies),* and also includes mires and shallow lakes (Löfgren 2010). The covered land area (water excluded) is characterized by 70–75 % forest, 10–20 % wetlands, 5 % arable land and 5 % pasture and meadow (Löfgren 2010). The estimated vegetation period in the area is April – September (Löfgren 2010).

2.2 Site characterization and survey designs

2.2.1 Field study I

Field study I was conducted in 2015 in early September covering three locations in the Forsmark area; moderately rich fen (Labboträsk), poor fen (Stenrösmossen), and a Norway spruce and alder wetland (Alsumpskogen) (Figure A1-1 and Appendix 1). At each location, sampling was conducted at two separate sites within the identified ecosystem (Figure A1-1, Appendix 1). One composite sample of vegetation and soil, respectively, were collected within a circle with a diameter of 12 m at each site. The green parts of the vegetation were sampled from the dominating species of the bottom, field, shrub and tree layer by collecting approximately 1.5–2 dm^3 biomass from many locations (7–10) within the circle to have a representative sample for the circle area (38 m^2). If trees were present in the circle, wood was sampled as well. Four soil corings were taken 3 m from the centre of the circle in each of the main compass directions (N, E, S, W). Soil was sampled at two depths, at 0–10 cm and at 50 cm soil depth. pH of the water was measured in the field for each soil coring. The same type of chemical analysis was made as in Field study II, as described below.

2.2.2 Field study II

Field study II was conducted in the first half of September 2017 close to the period when the biomass peaks. On each of the four locations, a gradient with three habitat sampling sites was established along an elevation gradient (Figure 2-1). The three sites along each gradient were chosen based on hillslope elevation to represent recharge (dry) and discharge (wet) areas as well as zones with intermittent recharge and discharge (typically moister soils and therefore denoted as moist) (Figure A2-1 to A2-4 in Appendix 2). These sites also had different vegetation. Wet sites were located closest lake shores and had alder forest with lush herb vegetation. Dry sites had spruce forest with blueberry (*Vaccinium myrtilis*) as dominant ground cover. The moist sites had spruce forest with more herbs and grasses than the dry site (Figure A2-1 to A2-4 in Appendix 2). The four gradient locations are hereafter referred to with the name of the lake closest to the wet habitat sampling site, namely, Lillfjärden, Gällsboträsk, Labboträsk, Eckarfjärden (Figure 2-1). The elevations of the habitat sampling sites, and the height over lake level, were estimated using a laser-based digital elevation map of the area (Petrone and Strömgren 2020) and the height over lake level was estimated for each plot (Table A2-1 in Appendix 2). The time since the habitat sampling site emerged from the sea due to land uplift was estimated based on the current elevation above the sea level (RH2000) and a model describing the historic sea-level change in Forsmark (Påsse 2001, Brydsten and Strömgren 2013).

Figure 2-1. *Sampling locations in Forsmark for Field study I: Labboträsk (young mire, LI and LII), Alder-Spruce wetland forest (AI and AII), Stenrösmossen (older mire, S1 and SII) and Field study II hill-slope gradients: Dry (upland dry coniferous forest, bilberry type), Moist (moist coniferous forest), Wet (wet alder forest wetland, wet herb type).*

2.2.3 Forest stand and vegetation characteristics – Field study II

The forest stand structure and vegetation characteristics were assessed at each habitat sampling site at the scale of a 100 m^2 circular plot. Habitat sampling squares of 1 m^2 were chosen at the centre of each habitat sampling site close to a tree. Bottom-, field-, and shrub-layer vegetation and soil were characterised within the square whereas tree samples were collected close to the square.

Stem density was determined by counting trees in the 100 m^2 plot and basal area of the stand (m^2 ha^{-1}) was estimated with a relascope (aperture-chain ratio of 1:5). For 4 to 6 trees of the dominating tree species, tree height was measured with a digital Clinometer (EC II D, Haglöf) and the diameter at breast height (DBH, breast height: 1.3 m above ground-level) was determined with a girthing tape. To estimate the age of the stand, the tree with largest diameter was sampled with a tree corer (5 mm

diameter). In addition, a central core was collected from the trees closest to each habitat sampling square (central tree). The distance between the last five-year growth rings, and the width of the last full year growth ring were recorded for each sampled tree using a slide calliper for the measurements.

Biomass for tree compartments (tissue, stem and living branches), was calculated for each measured from tree height and DBH. For conifer trees (*Picea abies* and *Pinus sylvestris*) and birch (*Betula* sp.) equations from Marklund (1988) were used, and for alder (*Alnus glutinosa*), the equations from Johansson (2000) were used. Deciduous leaf biomass was estimated using a function derived for birch (*Betula lenta*) (Martin et al. 1998). The derived average tree estimates were adjusted to the stand level of the site by dividing by the average tree cross-sectional area (m^2) and multiplying by the average basal area (m^2 ha^{-1}), resulting in a basal-area-weighted mean.

Biomass for the shrub and field layer was sampled in a 1 × 1 m square whereas the bottom layer (total bulk sample) was sampled in a 0.25 × 0.25 m sub-square, within the larger square. Senescent tissue and attached litter were sampled and assigned to this year production. Fresh weight and weight after freeze drying were noted for all samples and the biomass per square metre was calculated for the shrub, field and bottom layer based on the quantified biomass in each square plot. The dominating species in respective vegetation layer (shrub, field, bottom) was noted and the relative abundance (% cover) in field and bottom layer was estimated at the square plot level at each site.

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2.2.4 Soil texture analysis and soil density

The soils in the investigated area contain considerable amounts of rocks and boulders, which need to be accounted for when estimating soil pool excluding these fractions. The volumetric proportion of stones and boulders was estimated by the steel rod method (Stendahl et al. 2009, Viro 1952). The soil depth down to a stone closer to surface than 30 cm (30–60 cm was noted) was measured at each meter with a solid sond (10 mm diameter) along four perpendicular transects with the 1×1 m sampling square as the central point (n = 24) (Lundin et al. 2004). The volumetric content of stones and boulders in the soil (Z) was then calculated from the average penetration depth (x);

$$Z = 71.7 - 2.39x$$

where Z is volumetric content down to 30 cm (%) and x the penetration depth (cm).

Soil density, gravimetrical soil water content and texture of soil were estimated by collecting soil with a soil corer (10 cm diameter) next to the habitat sampling square. The soil was collected in PE-bags and transported to the lab where samples were air-dried (45 °C) for 24 – 48 h and weighed. The air-dried samples were passed through two different sieves, to separate the soil into three fractions (> 4 mm, 2– 4 mm, and < 2 mm) and then further dried at 105 °C for < 24 h. Loss-on-Ignition (LOI; the loss of weight during combustion) of the dried fine fraction (< 2 mm) was determined by combustion at 500 °C for four hours and the weight loss was calculated.

2.3 Sampling for chemical analysis (Study II)

Vegetation. The tree representing the dominating tree species closest to the square was chosen for sampling of foliage and phloem (Table 2-1). Wood was sampled from both the central tree (young tree) and the largest tree (old tree) at the site (tree core from the inner bark to the centre of the stem, the same sample as used for the tree age determination). Samples for foliage, annual shoot for spruce, phloem (inner bark), and wood were collected in PE-bags and stored in refrigerator until further analysis. For each vegetation layer (excluding trees) a bulk sample of all biomass in the sample square was collected

for further chemical analyses (specified below). If possible, in terms of enough sample biomass of the field layer being present, sub-samples were also collected separately for 1–2 dominating species. Green leaves and stems for shrubs and dwarf shrubs were not separated. For the bottom layer, the recent biomass of moss from the summer, was collected separately from the total bulk sample.

Soil. Soil samples for chemical analyses were collected by a soil corer (2.5 cm diameter, 40 cm) at five randomly selected spots within the square and mixed to one composite sample (Table 2-1). Soil was collected for the litter layer, the organic soil layer, and the mineral soil (≤ 40 cm).

Table 2-1. Summary of sampling for chemical analysis in Field study II. A habitat representative sampling square of 1 m^2 was chosen close to a tree at the center of each 100 m^2 sampling site). See text for details on sampling scheme.

	Type of sample		Sampling
Vegetation	Tree	Foliage, current year shoots	< 2 m from the habitat square (sample tree)
		Phloem	< 2 m from the habitat square (sample tree)
		Wood (tree core)	< 2 m from the plot (sample tree)
	Shrub	Stem and foliage	Square (1 × 1 m)
	Field layer	Herbs, grasses and dwarf shrubs (stem and foliage)	Square (1 × 1 m)
	Bottom layer	Biomass and annual shoot	Square (1 × 1 m)
Soil	Litter layer		Square (1 × 1 m)
(5 random sub-samples per square)	Organic layer		Square (1 × 1 m)
	Mineral layer		Square (1 × 1 m)

2.4 Chemical analyses (Study II)

2.4.1 Soil samples

Soil pH. Water extracts of the original soils were used for pH analysis. Soil pH was measured by adding fresh soil to media consisting of MilliQ water, KCl (1 mol L^{-1}), and $CaCl_2$ (0.01 mol L^{-1}) (1:5, soil:solution) (SIS 1994).

Freeze drying. The soil sampled for chemical analyses was initially freeze dried (−54 °C, 48 h) and thereafter homogenized by mortar before chemical analysis.

Total chlorine (Cl_{tot}) and organic chlorine (Cl_{org}). Measurements of chlorine were made by measuring TX (total halogens) as well as TOX (total non-leachable organic halogens) according to Asplund et al. (1994) using an ECS3000 analyzer (Euroglas). TX and TOX were assumed to primarily reflect chlorine (Cl_{tot} and Cl_{org}, respectively) because chlorine is the by far the most abundant halogen in soils. The soil was analysed for (TX) by weighing in approximately 20 mg of sample followed by combustion under a stream of oxygen at 1 000 °C (Euroglas AOX Analyzer). During combustion, the organic chlorine is converted into hydrogen chloride and trapped in acetic acid. Microcolumetric titration is used to quantify the chlorine content.

For TOX, in short, 20 mg sample was added to an acidic nitrate solution (0.2 KNO_3, 0.02 M HNO_3) and shaken one hour on a rotary shaker (180 rpm) to extract leachable chlorine from the soil. The solution was then filtered through a polycarbonate filter and rinsed with a nitrate solution (0.01 KNO_3, 0.001 M HNO_3) followed by acidified MilliQ-water (pH < 2). The filter and the soil sample were then combusted and analysed following the procedure for TX. The results are expressed as µg Cl g^{-1} dry mass.

The TOX concentration, as described above, may also include mineral bound Cl ($Cl_{mineral}$). Thus, to quantify the amount of $Cl_{mineral}$ in soil, a separate analysis was done. In this analysis, soil samples were precombusted (at 500 °C for 4 h) to remove organic matter, and then leached with acidic nitrate solution to remove all non-mineral chloride. The $Cl_{mineral}$ was then subtracted from the TOX results to yield

Cl_{org} in these samples. Cl^- (along with small amounts of leachable Cl_{org}) was calculated by subtracting values of Cl_{org} and $Cl_{mineral}$ from the TX results (Montelius et al. 2015). The fraction of water leachable chlorine has previously been shown to consist of > 90 % chloride (> 99 % in the O-horizon), the rest being organic chlorine (Asplund et al. 1994).

CNH analysis. Determination of elemental content of carbon (C), nitrogen (N), and hydrogen (H) – referred to as CNH analysis – was conducted using an Elemental analyser (Perkin Elmer EA2400). 0.002–0.015 g of soil was weighed in a tin container and the sample was combusted in a temperature of 925 °C and the CNH content was determined by separating the formed gases (CO_2, H_2O, N_2) and measuring their conductivity.

2.4.2 Vegetation samples

The vegetation samples were freeze dried in 48 h and thereafter homogenized until further analyses. The vegetation samples were analyzed for total organic halogens (TOX)[1] and total halogens (TX) with the same procedure as for soil samples. The chloride concentrations were estimated by subtracting TOX from TX. Determination of CNH was conducted using an Elemental analyser with the same procedure as for the soil samples.

2.5 Chlorine pool estimates

The area specific pools of total Cl, and the fractions of Cl_{org} and Cl^-, in vegetation and soil were determined for each sample site as follows:

The Cl pool in overstory was calculated by multiplying the measured biomass per m^2 of foliage and stem (basal area weighted) with the measured Cl concentrations of stem and foliage sample of the central tree (described above). The Cl pool in understory was calculated separately for the shrub-, field-, and bottom layers. For each layer the measured Cl concentrations of the bulk samples were multiplied with the measured biomass per m^2 (see above). At all four sites, the field layer pool of Cl was calculated from the biomass per m^2 and Cl concentrations of the four dominant species, rather than from a bulk sample.

Soil Cl pools were calculated by multiplying the average Cl concentrations (from five replicate soil samples) with the mass per m^2 for each soil layer. The soil mass per square meter was calculated by multiplying the soil bulk density with the average thickness of the soil layer, accounting for the content of stones and boulders:

$$Cl_{soilpool} = Cl_{conc} \cdot BD \cdot depth \cdot (1 - Z)/\,100$$

Where,

$Cl_{soilpool}$ (g m^{-2}) is the storage of total Cl in the different soil layers

Cl_{conc} (µg g^{-1} dry matter, *DM*)

BD (g m^{-3}) is the bulk density

Depth (m) is measured soil depth of the soil layer at each site

Z (%) volumetric content of stones and boulders

[1] Note that as the mineral fraction of chlorine ($Cl_{mineral}$) is insignificant in biomass, TOX for biomass corresponds to the organic fraction; Cl_{org}.

3 Results

3.1 Description of soil and vegetation

3.1.1 Field study I

Stenrösmossen and Labboträsk are two open mires influenced by calcite-rich water. Of these. Stenrösmossen is situated on a higher elevation (Table A1-1 in Appendix 1,), which means that it was isolated from the sea earlier than Labboträsk. The age of the mires is reflected in clear vegetation differences. That is, the older mire, Stenrösmossen, occasionally has Sphagnum species present in the bottom layer among the otherwise dominating *Bryales species* (e.g. *Warrnstorfia sp)*. In Labboträsk no sphagnum species are present but the presence of *Scorpidium sp* indicates an early successional stage (Table A1-2 in Appendix 1). The difference in age between the two mires is also reflected in the composition of vascular plant community. That is, the cover of *Myrica gale* in Stenrösmossen and is relatively high, whereas the cover of *Carex sp* and herbs are relatively low in Stenrösmossen, as compared to that in Labboträsk. Moreover, *Phragmites australis* is very common in Labboträsk but is completely missing in the older mire. The alder swamp forest is less influenced by calcite-rich water and is only occasionally flooded by a high-water table (situated close to a small pond). Forest species are common in the bottom, field and shrub layers of the alder swamp, though species indicating wet or moist conditions (*Filipendula ulmaria, Lysimachia thyrsiflora*) are also present.

The soils at the two mires (the young fen Labboträsk and the older mire Stenrösmossen) had a carbon content of approximately 45 % C at both sampled depths (at 0–10 and 50 cm) (Table A1-3 in Appendix 1). However, the soil of the wetland forest (Alsumpskogen) had substantially lower carbon content (25 %), but only slightly lower nitrogen content (1.7 % compared to 2.4 % in the two mires) and the C/N ratio at this site was lower than at the two mires (15 as compared to 20).

3.1.2 Field study II

Soil characteristics

The litter layer was 1–2.5 cm at dry and moist coniferous forest sites and 50 % thicker in the alder wetlands (Table A2-2 in Appendix 2). The organic layer was also thicker in the alder wetlands than in the dry forest (~30 %), whereas the thickness in the moist sites resembled that in the alder wetlands (rather than the thickness of the dry sites).

At the upland dry coniferous forest, the stone and boulder content (down to 30 cm depth) ranged from 36 % to 56 %, which is similar to previous estimates for upland sites in Forsmark (44 %, Lundin et al. 2004). The moist coniferous forest had a somewhat lower content of stones (17–31 %), whereas stones and boulders were less frequently encountered at the wet sites. That is, at three of the wet alder forest sites no stones or boulders were observed above a depth of 60 cm (Table A2-3 in Appendix 2). However, the wet site at Lillfjärden had a stone content of 34 %.

The average bulk density of the organic soil layer was 0.11 g cm^{-3} (0.04 – 0.24 g cm^{-3}) for dry coniferous forest, 0.21 g cm^{-3} (0.07– 0.31 g cm^{-3}) for moist coniferous forest, and 0.15 g cm^{-3} (0.03 – 0.27 g cm^{-3}) for wet alder forest. The average bulk density of the litter layer was 0.20 g cm^{-3} (0.13 – 0.30 g cm^{-3}) in dry coniferous forest, 0.26 g cm^{-3} (0.16 – 0.36 g cm^{-3}) for moist coniferous forest, and 0.22 (0.17– 0.27 g cm^{-3}) for wet alder forest. The observed bulk density was lower than previously observed in organic soil layer at upland sites in the area (0.6 – 1.4 g cm^{-3}; Leptosol and Regosol/Gleysol, Lundin et al. 2004).

Soil pH, gravimetric soil water content, LOI, C/N

The average soil water content of the organic soil was similar at the dry and moist sites (~57 %), and substantially higher at the wet sites (73 %). However, the water content varied considerably between habitat sampling sites, especially between the four moist sites (Table 3-1).

The soil pH increases successively from the upland coniferous forest to the wet alder forest sites, with averages of 4.5 (pH_{H2O}) and 6.7 for the dry and wet sites, respectively (Table 3-1). This pattern corresponds well to the general pattern previously studied in the area (Lundin et al. 2004). That is, most soils (80 %) have a relatively high pH (> 5.4), and there is a gradient of increasing pH from shallow upland soils (5.2), over moder forming soils in fresh and moist locations (5.6), towards relatively wet peaty mor soils (6.9).

The highest C content in the soil was found in the litter layer, on average 46.8 % (44.6 – 48.7 %) with similar percentages on all sites along the hill-slope. The C content of the organic soil layer was slightly lower with on average 35.7 % C at all sites. The C content in the mineral soil layer was less than 12 % (data not shown).

The C:N ratio was on an average 33.2 (23.8 – 44.3) in the litter layer and 21.5 (12.3–29.2) in the organic soil layer and 20.4 (11.7–28.9) in the mineral soil layer. There was also a tendency of the C/N ratio to decrease with soil depth in the two examined mires in Field study I (Table A1-3 in Appendix 1).

The C/N ratio in the organic soil layer decreased successively from an average of 27 at the upland coniferous forest to an average of 16 in the wet alder forest (Table 3-1). We also noted a similar low C/N ratio of 15 in the wetland forest sampled in Field study I (Table A1-3 in Appendix 1).

The dominant tree species in the dry and moist sites were spruce and pine, while alder dominated at all wet sites. A general description of the vegetation of the four different sites are presented in (Table A2-4 in Appendix 2).

Table 3-1. C/N-ratio, pH, and soil water content at the investigated sites in Field study II. Dry: upland dry coniferous forest, Moist: moist coniferous forest, Wet: wet alder forest wetland. Values show average (min–max).

		Position along hill-slope		
Soil layer		**Dry**	**Moist**	**Wet**
Litter	C:N ratio	34.0 (26.9 – 44.3)	37.0 (31.2 – 44.3)	28.8 (23.8 – 33.9)
Organic soil	Soil water content (%)	56.7 (47.8 – 61.9)	57.7 (38.2 – 84.8)	72.9 (52.3 – 85.5)
	pH (H_2O)	4.5 (4.0 – 5.1)	5.6 (4.8 – 6.5)	6.7 (6.1–7.1)
	pH (KCl)	3.2 (2.9 – 3.5)	4.8 (3.8 – 5.7)	6.2 (5.9 – 6.6)
	pH ($CaCl_2$)	3.5 (3.0 – 4.1)	5.3 84.1 – 6.4)	6.2 (5.8 – 6.5)
	C:N ratio	27.1 (23.8 – 29.2)	21.6 (17.4 – 27.2)	15.8 (12.3 – 20.8)
Mineral soil	Soil water content (%)	27.0[1]	25.6 (17.5 – 33.6) [1]	57.6 (47.1 – 68.2)
	pH (H_2O)	4.7 (4.6 – 4.8) [1]	7.4 (7.0 – 7.8)[1]	6.7 [2]
	pH (KCl)	3.5 (3.4 – 3.5) [1]	7.3 (6.9 – 7.7) [1]	6.1 [2]
	pH ($CaCl_2$)	3.6 (3.6 – 3.7) [1]	6.9 (6.8 – 7.1) [1]	6.4 [2]
	C:N ratio	22.6 (19.0 – 28.9) [1]	21.5 (19.5 – 23.6) [1]	11.7 [2]

1) N = 2
2) N = 1

The median stem biomass ranged from 12 to 62 kg m^{-2} between the habitat sampling sites (Table A2-5 in Appendix 2). The biomass tended to increase downwards the hill slope gradient, with the median values being 25 for upland dry sites, and 32 and 36 kg m^{-2}, for the moist and wet sites, respectively. The foliage biomass was an order of magnitude smaller than the stem biomass with a median value (and range) of 2.2 (0.5 to 4.3) kg m^{-2}. Moreover, the foliage biomass was higher in the in the spruce forest types than in the alder wetland. That is, the median values (and ranges) for upland dry sites, moist sites and alder wetlands were 2.4 (1.9–3.8), 3.4 (1.5–4.3) and 1.1 (0.5–2.3) kg m^{-2}, respectively.

There was a tendency of the understory biomass to decrease along the hill-slope from the dry to the wet sites; the median biomass (and range) was 0.32 kg m^{-2} (0.07–0.64 kg m^{-2}) in the upland sites, 0.25 kg m^{-2} (0.17–0.29 kg m^{-2}) in the moist coniferous forest sites, and 0.14 kg m^{-2} (0.11–0.26 kg m^{-2}) in the wet alder forest sites. However, the difference in biomass along a hill-slope of a site was small compared to the variation between specific hill-slope habitats of different sites. The biomass of the bottom layer was clearly lower at the wet sites (median 0.05 kg m^{-2}) compared to the dry and moist sites (median 0.24 and 0.17 kg m^{-2} respectively, Table A2-6 in Appendix 2).

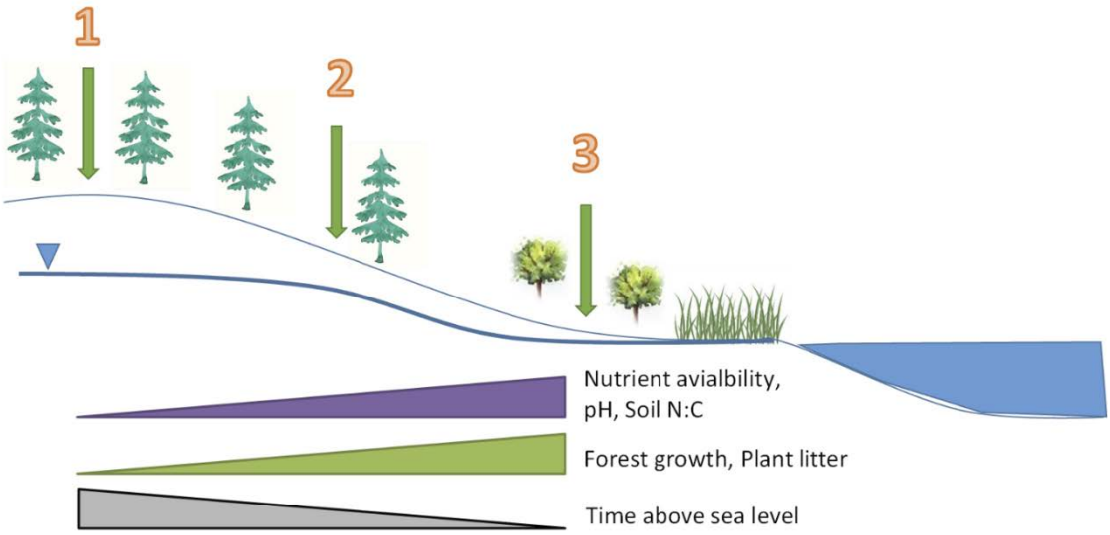

Figure 3-1. *Schematic figure of the three sampling sites along a hill-slope gradient where 1) upland dry coniferous forest (bilberry type), 2) moist coniferous forest (less bilberry/more grass and/or herbs), and 3) wet alder forest wetlands (wet herb type). The depth to the groundwater table (blue line) is expected to decrease with decreasing elevation along the hillslope (from left to right), whereas the productivity is expected to increase due to increased nutrient and water availability. The upland soils are expected to be much older than the soils close to the lake margin.*

Table 3-2. General forest stand characteristics of the three different sites for Field study II. See methods for definition of dry, moist and wet, respectively. dbh = diameter at breast height.

Properties	Gradient	Dry	Moist	Wet
Vegetation type[1]	**All**	**Spruce forest of bilberry type**	**Spruce forest of low herb type**	**Wet alder forest of herb type**
Dominant tree closest to sampling plot	*Lillfjärden*	*Picea abies/Betula…*	*Picea abies/Pinus sylvestris*	*Alnus glutinosa*
	Gällsboträsk	*Picea abies Pinus Sylvestris*	*Picea abies Pinus Sylvestris*	*Alnus glutinosa…*
	Labboträsk	*Pinus Sylvestris Picea abies*	*Picea abies Pinus Sylvestris*	*Alnus glutinosa Pinus Sylvestris*
	Eckarfjärden	*Picea abies Pinus Sylvestris*	*Picea abies Pinus Sylvestris*	*Alnus glutinosa Betula…*
Tree density (no trees/100 m^{-2})	*Lillfjärden*	11	7	26
	Gällsboträsk	10	27	24
	Labboträsk	12	19	15
	Eckarfjärden	10	10	58
Average tree height (m)[2]	*Lillfjärden*	16	16	14
	Gällsboträsk	23	21	15
	Labboträsk	17	17	13
	Eckarfjärden	18	21	10
Average dbh (m)[2]	*Lillfjärden*	0.19	0.20	0.18
	Gällsboträsk	0.34	0.32	0.22
	Labboträsk	0.25	0.20	0.25
	Eckarfjärden	0.26	0.30	0.14
Basal area (m^2/ha)	*Lillfjärden*	3 100	2 100	2 300
	Gällsboträsk	3 100	3 400	3 400
	Labboträsk	2 500	4 100	3 100
	Eckarfjärden	3 200	4 200	4 500
Tree age (years) 1 tree	*Lillfjärden*	59[a]	88[a]	45
	Gällsboträsk	88	85	62
	Labboträsk	155[a]	101	139[a]
	Eckarfjärden	132	118	62
Typical field layer species[3]	*Gällsboträsk*	*V. myrtillus, V. vitis-idaea*	*M. nutans, D. glomerata*	*C. appropinquata, F. ulmaria*
	Lillfjärden	*V. vitis-idaea, V. myrtillus*	*H. nobilis, C. majalis*	*E. canabinum, F. ulmaria*
	Labboträsk	*V. myrtillus, V. vitis-idaea*	*V. myrtillus, C. canescens*	*P. australis, C. diandra*
	Eckarfjärden	*V. vitis-idaea, V. myrtillus*	*L. vulgaris, V. vitis-idaea*	*F. ulmaria, T. palustris*
Dominating bottom layer species	*Lillfjärden*	*H. splendens, P. schreberi*	*H. splendens, P. schreberi*	-
	Gällsboträsk	*H. splendens, P. crista-castrensis*	*D. majus, H. splendens*	*H. seligeri, Mnium sp.*
	Labboträsk	*H. splendens, P. schreberi*	*P. crista-castrensis, H. splendens*	*C. cuspidate,* -
	Eckarfjärden	*H. splendens, R. triquetrus*	*R. triquetrus, P. schreberi*	*H. seligeri, Mnium sp.*

1. (Påhlsson 1998)

2. N = 4–6

3. See specific description if the species abundance in Appendix 5.

a = tallest tree non-dominat species (*Pinus sylvestris*)

3.2 Cl in soil

The total Cl concentrations in all soil layers at the investigated sites spanned almost two orders of magnitude, ranging from 30 µg g^{-1} DM (in mineral soil) to 1 800 µg g^{-1} DM (in peat at 50 cm depth) (Table A1-3 in Appendix 1). The average total Cl concentration in the organic soil layer (or humus) in Field study II was 570 µg g^{-1}, whereas the concentration in the litter layer was 370 µg g^{-1}. Mineral soil exhibited the lowest total Cl concentrations, with an average 120 µg g^{-1} soil. Despite a large variation among sites, there were no clear tendencies for the total Cl concentration to vary systematically along the hill-slope gradient in any of the soil layers. In the two sampled mires (Field study I) the Cl concentration increased with sampling depth, and the concentration was approximately twice as high at 50 cm depth, as compared to the upper layer (0–10 cm). The average concentration (across the two depths) for both of the mires (500 and 1 350 µg g^{-1}) fell within the range of the organic soils sampled along the hill-slopes (190–1 350 µg g^{-1}), but the relatively thin organic layer in the alder swamp forest (0–10 cm) had a noticeably lower concentration (120 µg g^{-1}).

Cl_{org} was detected in all soil samples and the percentage of Cl_{org} (of total Cl) was higher in the organic layer (74 %) than in the litter layer (39 %) (Table 3-3). Moreover, the percentage Cl_{org} in the litter layer was clearly affected by the position along the hill-slope. Thus, the percentage decreased from 50 % at the dry sites, to over 39 % at the moist sites, to 29 % in the wet site at the bottom of the hill-slope. The pattern was somewhat different in the organic layer, where the average percentage at the dry and moist forest sites was clearly higher than at the wet alder sites (82 % as compared to 39 %). The percentage Cl_{org} varied little with depth in the two mires, but the percentage Cl_{org} was significantly higher in the older mire compared to the younger mire (95 % as compared to 35 %).

The median concentration of mineral-bound Cl was estimated to be 14 µg g^{-1} soil in the mineral soil layer, and the percentage of mineral-bound Cl (of Cl_{tot}) ranged between 7 % and 45 % (Table 3-3).

Interestingly, it was noted that the absolute amounts of organic chlorine in organic soils was relatively stable across all wet sites (100–300 µg g^{-1}) compared to the range of total chlorine levels (120–1 350 µg g^{-1}). However, this was not true for the dry and moist sites, in which the amounts of organic chlorine appeared to be closely linked to the total chlorine levels ($r^2 = 0.99$).

There was a significant positive correlation between Cl_{org} concentration in organic soil and % C ($r = 0.629$, $p = 0.028$). The highest Cl_{org} concentrations were found at % C > 40 %. Cl_{org} was varying 2-fold at the dry and wet site, which is similar as for % C (Figure 3-2). At the moist sites, the Cl_{org} concentrations were varying more, approximately 10-fold, and the Cl_{org} concentrations increased with increasing % C. The same pattern was shown for % N, with a higher variation of Cl_{org} at moist sites compared to the dry and wet sites.

Table 3-3. Total Cl concentrations and percentage of Clorg in soil samples in Field study II. DM denotes dry matter soil. Clmin denotes the percentage mineral-bound Cl of total Cl. See methods for definition of dry, moist and wet, respectively. n/d = no mineral soil was observed with the method used. n/d = not determined.

		Total Cl (µg g^{-1} DM)			Clorg [Cl_{min}] (%)		
		Dry	Moist	Wet	Dry	Moist	Wet
Litter	Lillfjärden	371	386	459	48	29	19
	Gällsboträsk	252	478	221	41	52	28
	Labboträsk	351	757	392	69	26	41
	Eckarfjärden	266	355	230	43	48	28
Organic soil	Lillfjärden	369	188	609	88	69	38
	Gällsboträsk	591	393	452	92	94	63
	Labboträsk	712	326	1340	100	76	18
	Eckarfjärden	707	967	236	90	100	62
Mineral soil	Lillfjärden	80	123	907	47 [15]	86 [12]	16 [7]
	Gällsboträsk	n/a	n/a	n/a	n/a	n/a	n/a
	Labboträsk	150	32	n/a	75 [n/d]	30 [45]	n/a
	Eckarfjärden	n/a	n/a	n/a	n/a	n/a	n/a

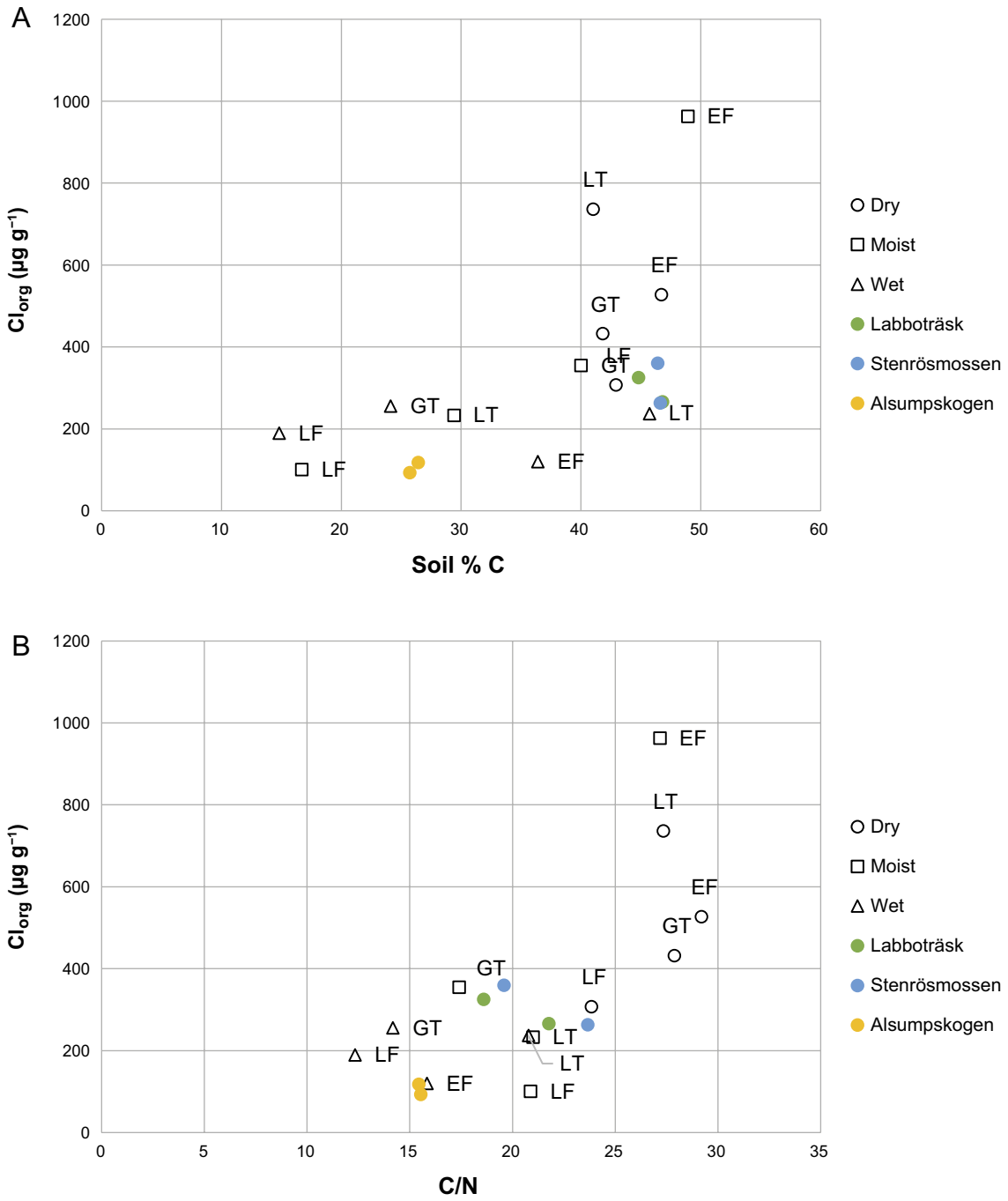

Figure 3-2. *The concentration of Clorg in organic soil layer plotted against soil C content in organic soil layer (A) and C to N ration (B). Dry: upland dry coniferous forest, Moist: moist coniferous forest, Wet: wet alder forest wetland. LF = Lillfjärden, GT = Gällsboträsk, LT = Labboträsk, EF = Eckarfjärden. Labboträsk (moderately rich fen), Stenrösmossen (poor fen), and Alsumskogen (a Norway spruce and alder wetland).*

There was also a positive correlation between Cl_{org} concentration in organic soil and the C/N ratio ($r = 0.693$, $p = 0.013$) suggesting that the nitrogen content in soil have a hampering effect on Cl_{org} or that N-poor organic matter is more easily chlorinated. The observed highest Cl_{org} concentrations were found at pH < 5.5. However, there were no significant correlations between the observed variations of total Cl or Cl_{org} concentrations in the organic soil layer with soil pH (Table 3-4).

Table 3-4. Correlation coefficients of Spearmans correlations for Cltot and Clorg against % C, C/N-ratio and pH in the organic soil layer.

	Cl_{tot} (µg g^{-1})	Cl_{org} (µg g^{-1})
% C	0.525	0.629*
C/N	0.326	0.693*
pH (H_2O)	0.220	−0.476
pH (KCl)	0.203	−0.363
pH ($CaCl_2$)	0.177	−0.433

* Correlation is significant at the 0.05 level (2-tailed).

3.3 Cl in vegetation

3.3.1 Total Cl in biomass

Total Cl varied substantially among the different types of biomass samples. However, in vascular plants the highest total Cl concentrations were found in biomass fractions which have a fast turnover rate, such as foliage and annual herbs compared to perennial biomass such as wood. For example, in the hill-slope study, the total Cl concentration in the tree layer was highest in foliage with an average of 820 µg g^{-1} DM, followed by phloem with 134 µg g^{-1} DM, and wood with 118 µg g^{-1} DM (Table 3-5). The same pattern was found at the forested site in Field study I, with higher total Cl in foliage than in wood for both spruce and alder trees (Table A1-3 in Appendix 1).

Table 3-5. Cl chemistry in sampled trees. See methods for definition of dry, moist and wet, respectively.

		Total Cl (µg g^{-1} DM)			% Cl_{org} of Cl_{tot}		
		Dry	**Moist**	**Wet**	**Dry**	**Moist**	**Wet**
Foliage	Lillfjärden	699	843	1 631	3.3	2.6	n/d
	Gällsboträsk	527	405	906	0.9	1.0	n/d
	Labboträsk	644	603	2 117 (alder) 383 (birch)	2.5	3.8	0.4 (alder) n/d (birch)
	Eckarfjärden	548	989	365	2.0	0.5	n/d
Phloem	Lillfjärden	119	166	276	6.7	n/d	3.6
	Gällsboträsk	88	205	107	1.1	11.2	32
	Labboträsk	97	162	100 (alder) 110 (birch)	6.2	9.9	16.4 (alder) 1 (birch)
	Eckarfjärden	100	152	61	14	9.9	25
Stem	Lillfjärden	34	59	59	27	n.d.	12
Young tree	Gällsboträsk	51	119	362	5.9	9.2	6.6
	Labboträsk	25	148	72 (alder) 49 (birch)	28	4.1	n/d 12.2 (birch)
	Eckarfjärden	197	134	222	3.6	6.7	4.1
Wood	Lillfjärden	22	14	15	n.d.	7.1	n.d.
Old tree	Labboträsk	22	38	n/a	46	5.3	n/a
	Gällsboträsk	20	20	47	5.0	25	2.1
	Eckarfjärden	48	30	71	17	6.7	11

At the forested site in Field study I, the total Cl concentration in alder foliage (417 µg g^{-1} DM) was approximately half of that observed in spruce foliage (861 µg g^{-1} DM) (Appendix 1, Table A1-3). In Field study II, where spruce and alder grew in different habitats such a clear difference was not evident. That is, the total Cl concentrations in spruce foliage ranged between 527–644 µg g^{-1} DM in the dry upland sites, and it was similar (though more variable) in the moist site (405–989 µg g^{-1} DM). However, the concentration in alder foliage at the bottom of the hill-slope tended to be higher, and it was typically above 900 µg g^{-1} DM (three out of four sites).

3.3.2 Cl_{org} in biomass

Even though Cl^- was the dominating form for chloride in biomass, Cl_{org} was observed in all analyzed fractions. Wood and mosses exhibited the highest Cl_{org} levels. The percentage Cl_{org} was on average 17 % in wood sampled along the hill-slope gradient, whereas the percentage was somewhat larger in the alder-spruce wetland forest (22 % for alder and 38 % for spruce, Table 3-5). The median % Cl_{org} in the bottom layer was 19 % in Field study II and 10 % in field study I. The phloem also had a relatively high percentage of Cl_{org} (10 %), but the fractions in tree foliage were substantially lower (< 4 %). The Cl in the field layer was strongly dominated by Cl^- (> 98 %), though there were still detectable amounts of Cl_{org} (< 65 µg g^{-1} DM).

3.3.3 Total Cl in ground vegetation

In both field studies the field layer had significantly higher total Cl concentration than the shrub and bottom layers. In the hill-slope habitats the average concentration in the field layer was 6 900 µg g^{-1} DM, whereas the concentrations in the shrub and bottom layers were 1 700 and 640 µg g^{-1} DM, respectively (Table 1 in Appendix 3). Similar concentrations were observed in Field study I, where the field layer had consistently higher concentration than the bottom layer in the alder-spruce wetland forest as well as at the two mires (Table A3-2 in Appendix 3).

The total Cl concentration in the field layer along the hill-slope ranged between 500 and 14 000 µg g^{-1} DM (Figure 3-1). The concentrations were significantly lower in dry upland sites compared to the moist and wet habitats. Upon a closer examination, the lower concentrations in the dry habitat coincided with differences in the species composition (Table A3-2 in Appendix 3). That is, the low concentration in the dry habitat was associated with dwarf shrubs, which were rare in the moist habitats, and absent in the alder stands at the bottom of the hill-slope. However, when dwarf-shrubs were present in the moist habitats (two sites) the concentration was similar to that in the dry upland habitat. Thus, it seems like the differences in total Cl concentrations is determined by the vegetation composition rather than the hill-slope position. It was also noted that the total Cl concentrations in lingonberries, *Vaccinium vitis-idaea*, tended to be lower than in blueberries, *Vaccinium myrtillus* (Table A3-2 in Appendix 3). Thus, the pattern of lower Cl concentrations in perennial than in annual tissues was also evident among dwarf-shrubs, as blueberries shed their leaves annually, but lingonberries do not.

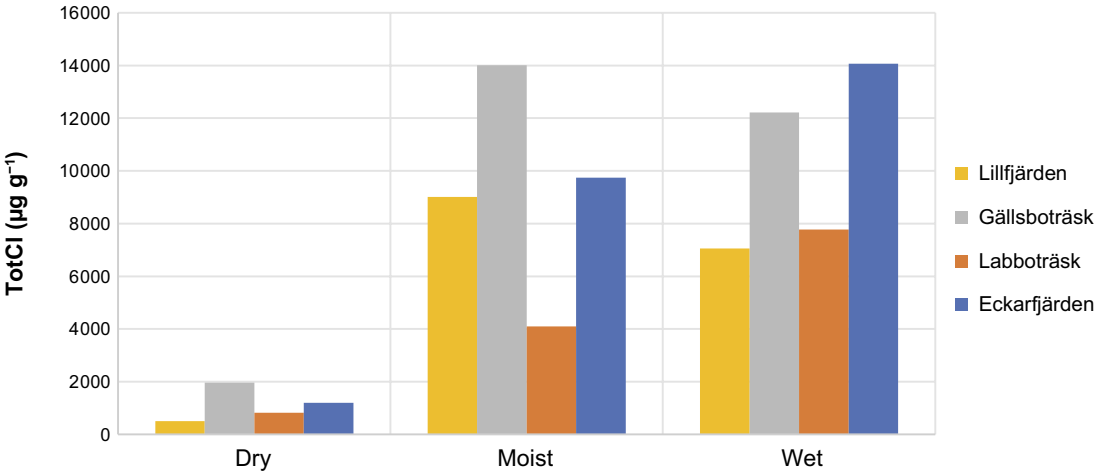

Figure 3-3. *Total Cl concentrations in the field layer (composite samples). See specific concentrations in Table A3-1 and A3-2 in Appendix 3. See methods for definition of dry, moist and wet, respectively.*

3.4 Pool estimates of Cl

The total Cl pool in the litter layer of the hill-slope sites ranged between 0.5 and 5.8 g m^{-2}, of which 40 % was typically in organic form. However, as the total concentrations and the thickness of the litter layer did not vary substantially between habitats, the pool was stable along the hill-slope. The total Cl stored in the organic soil layer was typically five times larger than the litter pool; the median of the two pools were 7.5 and 1.4 g m^{-2}, respectively. The between site variation of the Cl pool in the organic soil layer was considerably larger than in the litter layer, and Cl pools in organic soil varied between 1.6 and 72.9 g m^{-2}. Moreover, the amounts of Cl were considerably smaller in the dry upland habitat (5.5 g m^{-2}) compared to the moist and wet habitats (21 and 24 g m^{-2} respectively). As the Cl concentration in organic soil layer did not differ notably between habitats (Table 3-3) the difference was primarily due to a thinner and more boulder rich soil layer at the upland site (Table 2-3 in Appendix 2). The median total Cl pool in the mineral soil in the investigated areas was 23.3 (6–28.4) g m^{-2}. However, the estimate of these soil Cl pool is highly uncertain due to lack of mineral soil samples from Gällsboträsk and Eckarfjärden.

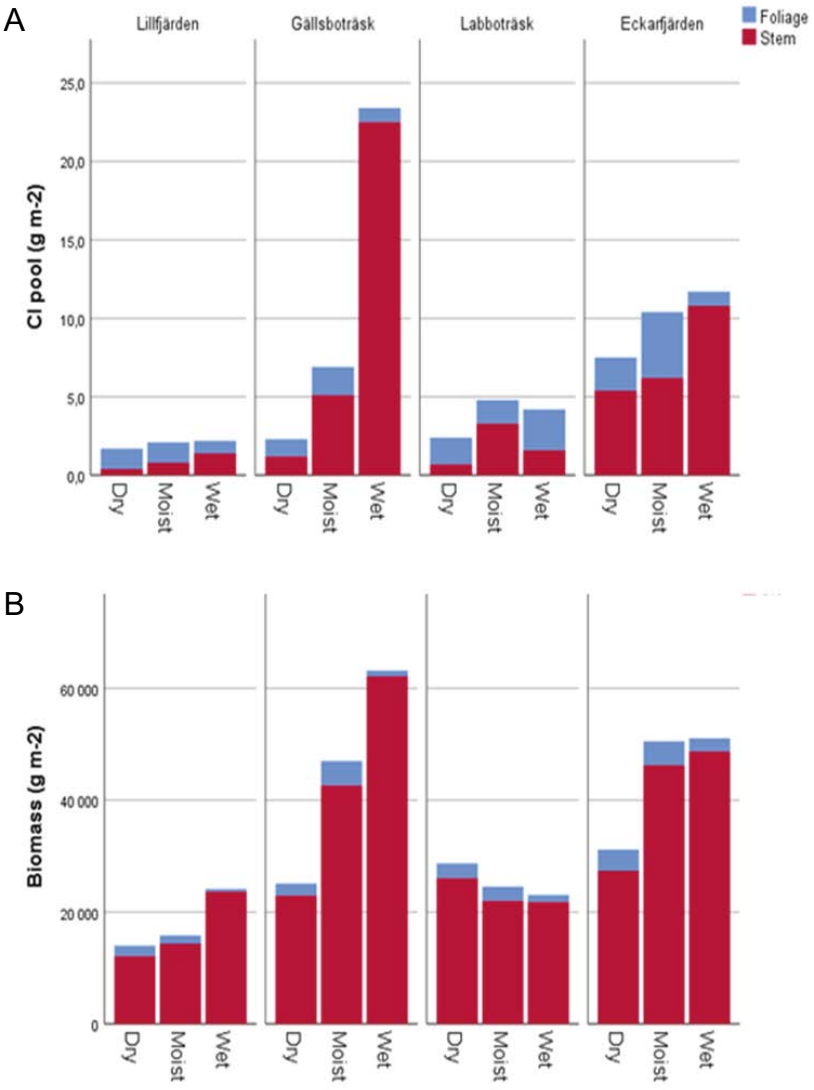

Figure 3-4. *Estimated Cl pool in tree compartments (foliage and stem) of each sampled sites (A) and corresponding biomass (B). See methods for definition of dry, moist and wet, respectively.*

Based on the measurements, the estimated biomass of foliage was relatively small compared to the biomass in stems (Table A2-5 in Appendix 2). However, as the Cl concentration in foliage was considerably higher than in stem tissue the storage of Cl in foliage still made up a significant fraction of the total tree Cl pool (Table 3-4). For three of the sites the foliage pool was even in the same order of magnitude as storage in wood (Figure 3-2). The Cl foliage pool was relatively stable along the hill-slope gradient, though there was a tendency for somewhat larger pools at the dry conifers dominated, and moist upland sites habitats (1.5 and 2.2 g m^{-2}) as compared to the wet alder stands (1.3 g m^{-2}).

Across all sites, the stem wood was the largest pool for storage of chlorine in tree biomass, and the average pool of Cl in stems was 4.9 g m^{-2}. However, the amount stored in wood varied systematically along the hillslope gradient. Thus, this chlorine pool increases from 0.9 m^{-2} in the coniferous upland site to 4.2 g m^{-2} in moist habitat, and the largest pool, 6.2 g m^{-2}, was found in the alder stands at the bottom of the slope. The change in the size of Cl pool along the hill-slope primarily reflects the increase in stem biomass from dry to wet habitats, but at the dry site the low pools are further accentuated by relatively low chlorine concentrations in the wood.

The median total Cl pool in the understory biomass in the investigated area was 0.7 g m^{-2} (0.04–1.4 g m^{-2}) (Figure 3-5). The pool of Cl in the understory increased along the gradient from an average of 0.19 g m^{-2} at the upland dry coniferous forest sites to an average of 1.13 g m^{-2} at the wet alder forest wetland sites (Figure 3-5). The increase was due to the combined effect of the increasing Cl concentration and biomass in the field layer along the gradient (Table A2-6 in Appendix 2 and Table A3-1 in Appendix 3).

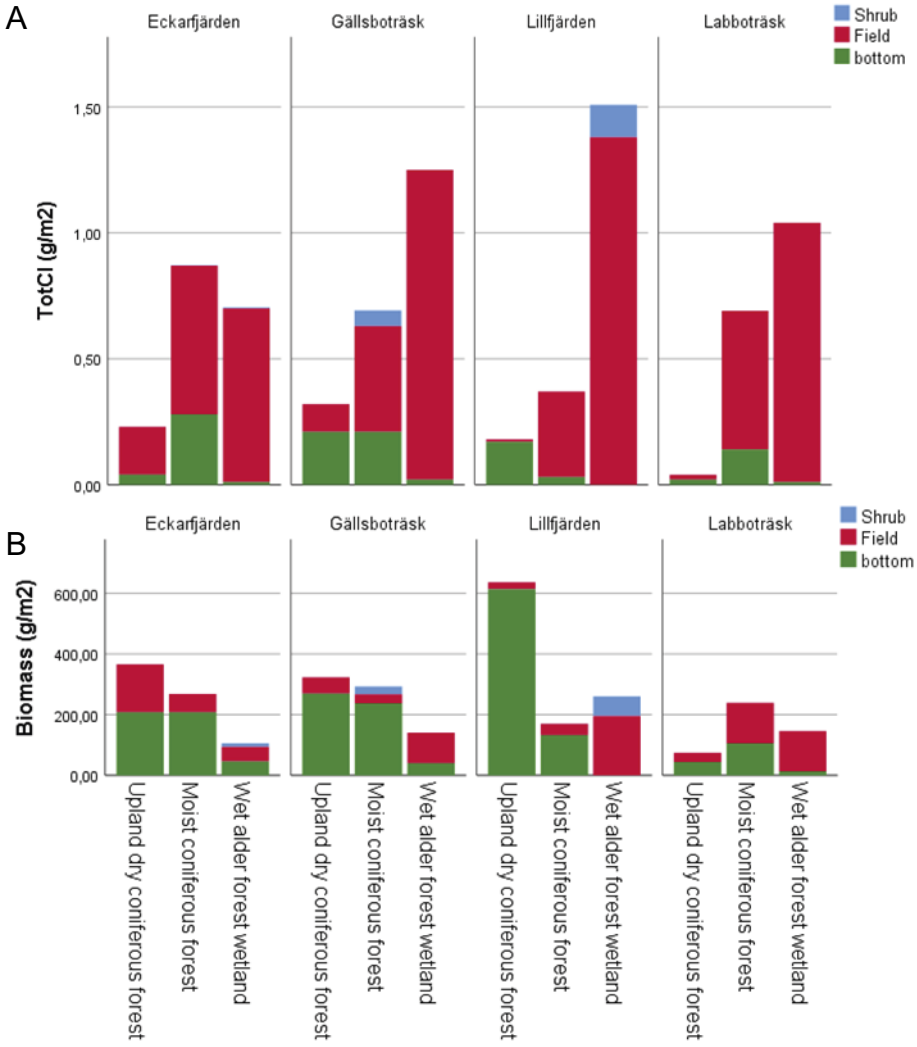

Figure 3-5. *Estimated Cl pool in understory layers (shrub, field and bottom) of each sampled sites (upper figure) and corresponding biomass (lower figure). See methods for definition of dry, moist and wet, respectively.*

3.4.1 Comparing Cl in soil and vegetation

The relationship between Cl in soil and in green plant tissue was examined for the tree, shrub, field and bottom layers (see below). No clear positive relationship between the Cl (Cl_{tot} or Cl^-) concentration in the organic soil layer and the plant Cl (Cl_{tot}) concentration could be found in any of the examined vegetation layers (see below).

Thus, the Cl concentration in foliage was not significantly correlated to that in soil ($r = 0.068$, $p > 0.721$, and $r = -0.110$, $p = 0.721$ for soil Cl_{tot} and Cl^- respectively) (Figure 3-6). However, the alder foliage concentrations on the two sites with the highest soil concentrations (> 400 µg g^{-1}) also had the highest foliage concentrations (> 1 500 µg g^{-1}). In the shrub layer there was a tendency in the opposite direction, as the Cl concentrations in plants decreased with increasing soil Cl^- concentrations at the moist sites ($r = -0.487$, $p = 0.268$). However, the species at the sites were all different, and thus the pattern may have been associated with shrub species rather than with soil Cl^- concentrations.

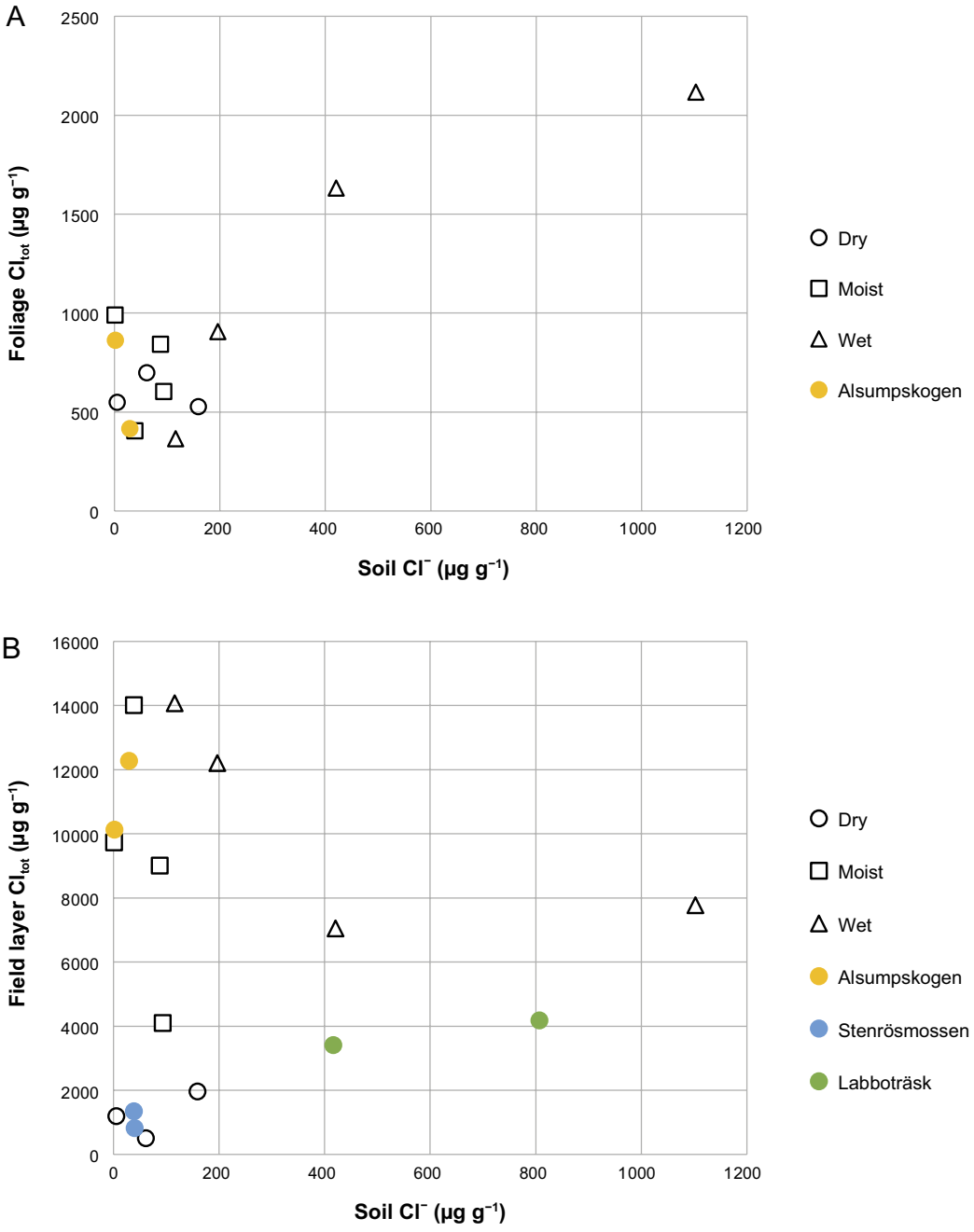

Figure 3-6. *Total Cl concentration in tree foliage (A) and in the dominating species in the field layer (lower panel) and Cl^- concentration in the organic soil layer. Results from Field study I: Alsumpskogen, Stenrösmossen, Labboträsk. Results from Field study II: Dry, Moist and Wet, (See methods for definition of the three habitats).*

The Cl concentration of the composite field layer samples was not positively related to neither the Cl_{tot} nor the Cl^- concentration in organic soil ($r = -0.288$, $p > 0.247$, and $r = -0.184$, $p = 0.480$). Neither were the concentrations in the dominating species correlated to the soil Cl concentration. However, opposite to the pattern in alder foliage, the field layer Cl concentration tended to be negatively related to soil concentrations in the wet sites at the bottom of the hill-slope. Finally, the bottom layer (total biomass) exhibited a large variation in Cl concentrations, but this variation was not related to the soil Cl concentration ($r = -0.202$, $p > 0.421$, and $r = 0.001$, $p = 0.966$ for soil Cl_{tot} and Cl^- respectively).

4 Discussion

4.1 Cl concentrations are similar among different habitats, but the fraction of Cl_{org} is higher in the drier habitats

It is well-known that chlorine is abundant in soils, primarily based on studies that have been conducted in well-drained soils (such as in the upper forest type along the hill-slope gradient). The total Cl concentrations in the upland dry forest site and moist forest were in general higher, 369–712 µg g^{-1} than the earlier studies in coniferous forest soils further south in Sweden, 99–458 µg g^{-1} (Gustavsson et al. 2012, Johansson et al. 2003a). One possible reason for that could be that the soils of Forsmark are located beneath the highest coastline and thus the soils are rather young soils compared to Southern Sweden and contain typically high Cl. In addition, Forsmark location is beneath the highest coastline from after the recent ice age and is situated close to the Baltic Sea, having a maritime impact including an increased influence of atmospheric deposition of Cl from sea spray. Johansson et al. (2003a) studied the spatial distribution of Cl in humus from the west coast to the east coast in Southern Sweden and saw higher Cl_{org} and Cl^- at the west coast than at the east coast suggesting an influence of Cl^- deposition on the soil Cl concentrations. The local variations in wet Cl deposition among the sites in Forsmark are considered to be negligible. If we compare with previous measurements of Cl in soil in the area, the previous estimate of Cl in soil, < 281 µg g^{-1} (Hannu and Karlsson 2006) is lower compared to an average of 574 (188–1 340, min–max) µg Cl_{tot} g^{-1} in the current study. This can be attributed to different analytical methods, where the previous measurements were conducted with ICP excluding Cl_{org}, while in the current study, we used a sum parameter of total chlorine including the Cl_{org} content.

There is increasing evidence that a substantial part of the terrestrial Cl is organically bound in upland forest soils (Bastviken et al. 2013). In the organic soil in the current study, the proportion of organically bound chlorine averaged more than 80 % in the upper habitats and approximately 41 % in the wet alder forest. Thus, even if the total Cl concentrations are similar among the sites, there is a large variation in the total Cl chemical composition. Earlier data have shown that the Cl_{org} content is correlated to carbon content in forest soils (Redon et al. 2011). In this study, the highest carbon content was observed at the dry sites and this thus coincided with the highest amounts of Cl_{org} along the hill-slope gradient. However, the carbon content of the peat soils in the two sampled mires was much higher > 70 % and the Cl_{org} concentrations was less than 400 µg g^{-1}, and thus do not reach the very high Cl_{org} concentrations observed in the upland soils. Thus, the amount of carbon cannot alone explain the variation of Cl_{org} in forest soils. The role of carbon for the formation of Cl_{org} in discharge areas with a thick organic soil layer needs more studies. It may well be that the quality of the organic matter, i.e. its tendency to be degraded more or less rapidly, might be as important for chlorine immobilization as its quantity. Whether some Cl_{org} is more refractory than other types of soil organic matter remain to be investigated.

Other previously discussed factors that may influence soil Cl_{org} concentrations are pH and nitrogen. In the current study, the highest Cl_{org} concentrations were found in soils with pH < 5.5. This is corresponding to previous studies showing that pH correlated negatively to Cl_{org} in soil (Johansson et al. 2001). The dominating formation processes of organochlorines are known to be biotic, however the specific formation processes are still not understood. A common explanation is that Cl_{org} is formed during degradation of organic matter through (enzymatic) formation of reactive chlorine. The two main substrates for this process are organic matter and Cl^-. In the pH-interval 3–5, the enzymatic process increases with decreasing pH (Asplund et al. 1993, Öberg et al. 1996). However, as the study by Johansson et al. (2003a) was conducted on a landscape scale the deposition of chloride was regarded to be the major factor explaining soil Cl_{org} concentrations and thus pH is likely an important factor on a small spatial scale as in the current study with similar Cl^- deposition.

The total Cl concentrations were similar along the hillslope gradients, but the fraction of Cl_{org} was higher in the upland soils. This could be a result of pH (see above), but more likely a combination of several factors. From upland soil to the wet alder forests the relative amount of nitrogen increased, and the soil C/N ratio was much higher at the dry sites than at the sites closer to the lake. In several studies, nitrogen has been suggested to be an important factor for the Cl_{org} levels in soil. Montelius et al. (2016) and Svensson et al. (2017) showed that specific chlorination was hampered by addition of nitrogen in experimentally incubated soil. Thus, a combination of relative high pH and nitrogen levels will likely lead to relative lower fraction of Cl_{org} in soil.

It has been suggested that a local variation in soil is caused by vegetation and more specifically tree leaching, reaching soils as throughfall, and vary among different forest types and tree species (Montelius et al. 2015, Öberg et al. 1998). Despite the change in vegetation habitat along the hill-slope from coniferous forest in the upper recharge areas to alder forest close to the lakes, the variation in Cl_{tot} concentrations was similar in the current study.

4.2 High Cl_{tot} concentrations in biomass with short turnover time

Cl was abundant in all types of vegetation investigated, but to a varying degree. The highest concentrations were found in the field layer. The concentrations in the field layer were even higher than in tree foliage that previously has been shown to accumulate high Cl concentrations (Gielen et al. 2016, Montelius et al. 2015, Van den Hoof and Thiry 2012). Vegetation with annual above-ground plant parts, e.g. grass and herbs species, show high Cl concentrations while plants with perennial plant parts, such as *Vaccinium vitis-idaea*, show lower concentrations. The Cl concentrations of *Vaccinium myrtillus* and *Myrica* (which has a perennial stem) were higher than in *Vaccinium vitis-idaea* having perennial parts only. This suggest an active regulation and limited Cl accumulation in perennial plant biomass, as compared to annual, where surplus Cl possibly is translocated to the annual biomass of the perennial plants.

Cl^- is the dominating chlorine form in both tree and understory biomass. The number of previous studies that have examined the form of chlorine in plant tissue is limited, but overall, they support a dominance of Cl^- over Cl_{org} in plant biomass (Table 4-1). Cl^- is an essential micronutrient that has a direct role in e.g. photosynthesis and stomatal regulation, and it is needed for ion balance and osmotic regulation (White and Broadley 2001). Thus, it is possible that the observed variation in biomass concentrations may be linked to conditions affecting one or several important plant functions.

Biomass with fast turnover and photosynthetic active parts, like leaves, needles, and annual vegetation parts had higher Cl concentrations than “supporting tissue” such as stem. Chlorine is ubiquitous in plant biomass but varies among species, as well as among different parts of the plants (Table 4-1). Among trees the reported total Cl concentrations in wood varies over an order of magnitude between 10 and 92 µg g^{-1} DM (Flodin et al. 1997, Gielen et al. 2016), with one study from Forsmark reporting concentrations an order of magnitude above the high end of the observations in the current study (Hannu and Karlsson 2006). There are more studies that have investigated the Cl concentration in foliage (over a broader range of species and ecosystems), and consequently it is not surprising that the reported variation of total Cl concentrations is larger in tree foliage than in wood. That is, the reported Cl concentration in foliage range from 183 to 15 000 µg g^{-1} DM (Flodin et al. 1997, Gielen et al. 2016, Hannu and Karlsson 2006, Holmes and Baker 1966, Montelius et al. 2015) (Table 4-1).

Table 4-1. Reported average (min–max) concentrations of Cl in different types of vegetation. Data in rows in bold originates from Forsmark area. CT = Central tree for chemical analysis (young tree) and the tree with largest diameter (old tree, for age estimation). DM = dry matter.

Vegetation	Cl_{tot} (µg g^{-1} DM)	% Cl_{org} of Cl_{tot}	References
Tree layer	**485–989** **365–2 117** **385**	**2.1 (0.5–3.8)** **0.4**	***Current study*** **(spruce)** **(alder)** **(birch)**
Foliage (*Picea abies*)	10 367 (6 100–15 000)		*1*
Foliage[a]	1 242 (183–14 264)		*2, 3, 4, 5, 6, 10*
Foliage[b]	315 (183–600)	8 (2–15)	*3, 5*
Wood (*Picea abies*)	**25–197 [14–48]** **49–362 [15–71]** 3 700 (1 900–5 600)	**23 (3.6–100) [16 (5.0–46)]** **7.5 (4.1–12) [6.7 (2.1–11.3)]**	***Current study*** **Young tree [Old tree]** **(spruce)** **(alder)** *1*
Wood[c]	14 (6–27)		*5.6*
Wood[d]	15 (10–27)	28 (22–36)	*5*
Branches[c]	96 (19–278)		*5, 6*
Branches[d]	63 (19–133)	18 (9–32)	*5*
Bark[c]	98 (26–296)		*5, 6*
Bark[d]	96 (26–296)	17 (5–31)	*5*
Phloem	136 (88–205) 148 (61–276)	8.4 (1.1–14) 20 (3.6–32)	***Current study*** (spruce) (alder)
Field layer	**131 –> 30 000** 31 360 (12 000–68 000)	**4.6 (0.1–34)**	***Current study*** *1*
	1 237 (116–4 420)	35 (0.3–75)	*3, 7, 8, 9*
Bottom layer	**191–1 344**	**13** **(2.5–36)**	***Current study (total bulk sample)***
	5 063 (890–9 600)		*1*
	757 (541–1 030)	26 (8.6–47)	*3, 7, 9*

1. (Hannu et al. 2006), 2. (Holmes et al. 1966), 3. (Flodin et al. 1997), 4. (Lovett et al. 2005), 5. (Montelius et al. 2015), 6. (Gielen et al. 2016), 7. (Asplund et al. 1994), 8. (Nkusi et al. 1995), 9. (Zlamal et al. 2017), 10. (Edwards et al. 1981)

[a] Birch, Oak, European beech, Sugar maple, Norway spruce, Black pine, Douglas fir, Scots pine *Fraxinus Pennsylvania*, *Beula pendula*, *Caragana arboscens*, *Prunus virginiania*, *Cornus stolonifera*, *Ulmus americana*, *Ulmus pumila*, *Lonicera tartarica*, *Acer negundo*, *Quercus macrocarpa*, *Elaeagnus angustifolia*, *Pinus contorta*, *Pinus contorta*, *Pinus resinosa*, *Pinus sylvestris*, *Polpulus balsmafera*, *Populus tremuLOIdes*, *Populus sp*, *Picea pungens*, *Picea glauca*, *Salix pentandra*.

[b] Oak, European beech, black pine, Douglas fir, Norway spruce , Birch.

[c] Oak, European beech, black pine, Douglas fir, Norway spruce, Scots pine.

[d] Oak, European beech, black pine, Douglas fir, Norway spruce.

4.3 Species composition determines the Cl concentration in field layer rather than soil Cl concentrations

In radio-ecology the most common method to model transfer of elements from soil into plants is the use of empirical transfer factors, under the assumption that there is a linear relationship between plant and soil concentrations (IAEA 2010, Sheppard and Evenden 1988, 1990, Vera Tome et al. 2003). In Field study I, the young mire exhibited relatively high soil Cl concentrations and high totCl concentrations in the field layer compared to the older mire with lower soil totCl concentrations. However, the species composition was different and thus we cannot elucidate if the higher totCl concentrations in the young mire were a result of soil Cl. Field study II suggests that the field layer concentrations are not related to soil Cl concentration (i.e. the organic layer), but as the species composition changes along our gradient so do the Cl_{tot} concentration in the field layer.

4.4 Cl_{org} in biomass exhibited the highest percentage in perennial structures

The totCl concentrations are much higher in foliage than in wood. This finding is consistent with Montelius et al. (2015) who found the highest concentrations in foliage followed by bark > branches > wood suggesting a transport of Cl^- within the trees from root uptake to foliage. The transport of substances within the plant (translocation) is regarded to be very high for Cl. In general, it is assumed that a major part of vegetation uptake is by moving Cl^- through the root and into the xylem and further to the shoot, where it accumulates or is redistributed throughout the plant via the phloem (Atwell et al. 1999, MacAdam 2009).

The concentrations of Cl_{org} in trees of Field study I are ranging from 4–23 µg g^{-1} DM in foliage (which is approximately 2 % of total Cl. The concentrations of Cl_{org} are similar in wood, resulting in a larger relative content of Cl_{org} in wood (Table 4-1). Similar pattern of relatively high Cl_{org} of totCl was found in wood by Montelius et al. (2015) for all investigated tree species (oak, European beech, black pine, Douglas fir, Norway spruce).

In trees, the observed Cl concentrations of phloem (61 to 276 µg g^{-1} DM) are similar to what has previously been reported for Scots pine (app 55 years old), where the average Cl concentration in phloem was 159 µg g^{-1} DM (Gielen et al. 2016). In the current study we separated the inorganic part from the organic and found on average 11 % Cl_{org} in phloem and approximately 19 % in wood. As phloem conducts the transport from foliage and the photosynthesis production of carbohydrates and other organic molecules towards roots, the observed higher Cl_{org} concentrations in phloem than in foliage might suggest that Cl_{org} can be produced within the tree as suggested by Montelius et al. (2015). However, there are unfortunately no earlier studies on Cl_{org} in phloem.

The observed concentrations for wood and foliage in the current study are within the range of reported values from earlier studies (Table 4-1). For wood the Cl concentration in old spruce trees was similar to those reported for Norway spruce wood by Flodin et al. (1997) and for pine wood by Gielen et al. (2016). The concentration in young trees was approximately twice as high, but still an order of magnitude below the concentrations previously reported from Forsmark (Hannu and Karlsson 2006). The analytical procedures differed between the earlier data from Forsmark and current study, total extraction followed by ICP analysis compared to total Cl analysis with combustion followed by microcoulometric titration, however it is unclear how this could explain the variability. Earlier studies indicate that the Cl concentration in deciduous foliage is lower than that in coniferous foliage (Montelius et al. 2015). In the current study, this pattern is not as clear as alder foliage ranged from 365 µg g^{-1} DM up to as high as 2 117 µg g^{-1} DM. At the site with the highest alder foliage total Cl level, total Cl concentration in birch foliage was much lower, 383 µg g^{-1} DM. Our results indicate that deciduous trees can have varying total Cl levels in foliage depending on the location, and that there can be large differences among foliage from different tree species at the same locations. There are no earlier studies on Cl concentrations in alder trees known by the authors. For birch, foliage Cl^- levels of approximately 500 µg g^{-1} DM were observed by Edwards et al. (1981).

4.5 Pool estimates

Despite the large variation in total Cl pools, there was no strong gradients in Cl pools among the investigated habitats and total soil and vegetation Cl concentrations were not affected by the forest hill-slope gradient in this study (Table 3-3 and 3-5). The size of chlorine pools was instead clearly linked to the thickness of soil layers and the tree biomass, and thus the highest Cl pools were found at moist and wet sites. Moreover, the dry habitats tended to have less Cl stored in ground vegetation than moist and wet habitats, and this shift was presumably due to the relatively high abundance of perennial dwarf shrubs in the dryer habitats (as opposed to annual herbs and grasses). The contribution of organic Cl to the total soil pools also tended to be lower in the wet sites compared to the dry and moist habitats. For example, Cl_{org} accounted for more than 70 % of the humus layer in the two upper habitats, whereas Cl^- dominated the humus pool in the wet alder stands.

There have only been a few attempts to estimate the standing stock of Cl in biomass. Öberg et al. (2005) estimated the Cl pool in biomass, dominated by *Pinus sylvestris* to 2.1 g m^{-2} of which 0.1 g m^{-2} is Cl_{org} and Montelius et al. (2015) estimated the total tree Cl biomass for Norway spruce to approximately 2.5 g m^{-2}, which is lower than the estimated pools in current study of 3.6 – 10.4 g m^{-2} in tree biomass. Tröjbom and Grolander (2010) estimated that the terrestrial biomass (including below-ground biomass) Cl pool contained 60 % of the total catchment Cl content in Forsmark (Sweden). The high enrichment of Cl in biomass, much higher than biomass in another similar area (Laxemar, Sweden), called for further studies to confirm the results. The aboveground biomass Cl pool in the current study was 33 %, 31 %, 40 % at dry, moist and wet sites, respectively, of total Cl pool of aboveground biomass and litter and organic soil (Figure 4-1). It is known that Cl is an essential element, but this level of enrichment indicates that the role of Cl for organisms may not be fully understood. A large part of potential contaminant ^{36}Cl accumulating in terrestrial parts of the landscape could potential be taken up by biota.

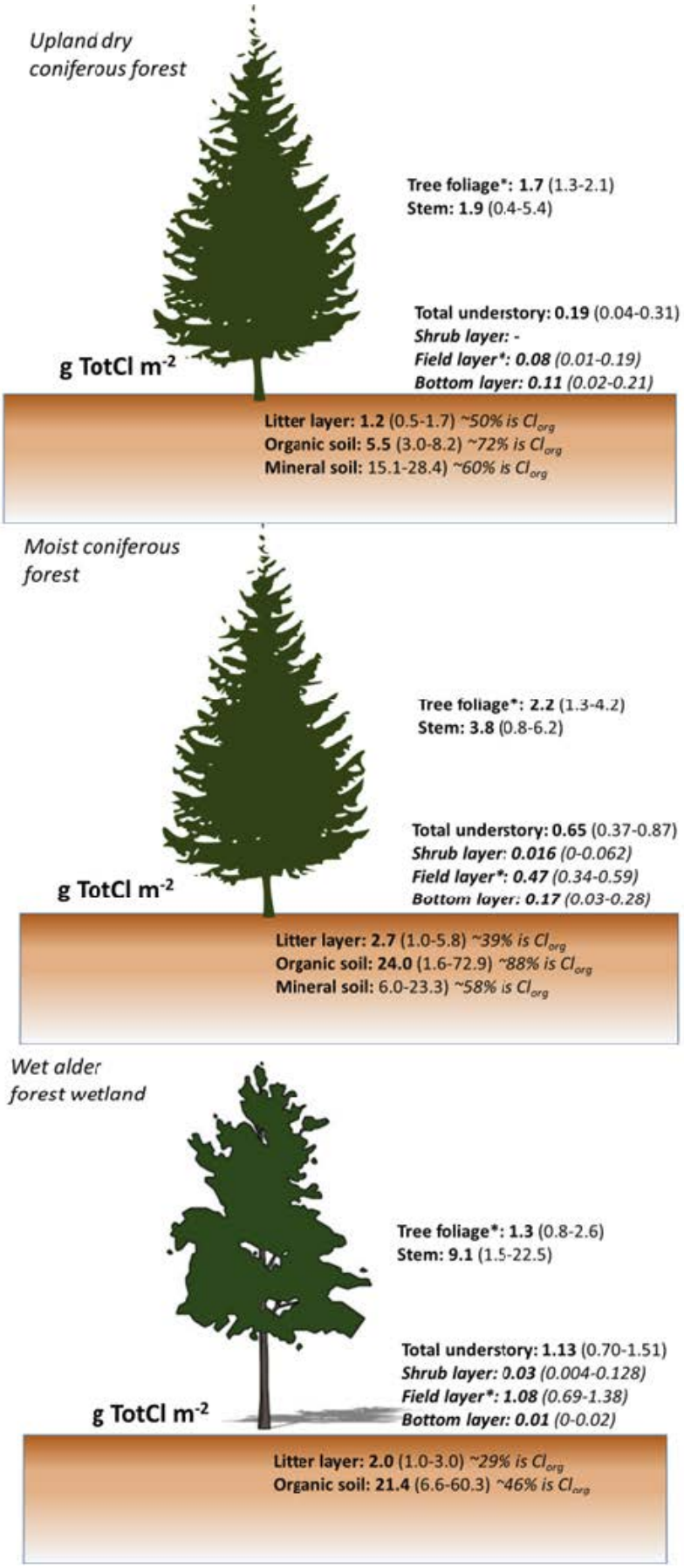

Figure 4-1. *Average total Cl pool (min–max among the different sites in brackets) at three different habitats along a hill-slope (see methods for definition of dry, moist and wet, respectively). The % Cl_{org} in vegetation was low < 4 % in foliage, < 1 % in field layer, but higher in bottom layer < 24 %. Wet deposition in the area is 0.45 g Cl_{tot} m^{-2} y^{-1}.*

5 Conclusions

The dominant form of chlorine in plants is chloride (Cl^-). In this study, this was confirmed for all tree fractions and for the understory vegetation. The concentrations of Cl_{org} were typically less than 2 % in foliage and field vegetation but could be higher in wood (< 30 %) and in the bryophytes of the bottom layer (< 36 %). Biomass with fast turnover and photosynthetic active parts, like leaves, needles, and annual shoots had higher Cl concentrations than "supporting" tissue. Thus, in trees, the highest Cl concentrations were found in foliage, followed by phloem and wood. In addition, very high Cl concentrations were found in annual herbs and graminoid species in the field layer. The high Cl concentration in understory was most pronounced at the wet and moist sites and seems to be related to the species composition (rather than the soil Cl concentration). Further studies are needed to elucidate the hitherto unknown processes that regulates Cl uptake in vegetation, and to determine to what extent plant driven Cl^- cycling may affect the residence times of chlorine in terrestrial ecosystems.

Organic Cl (Cl_{org}) is a quantitatively important fraction of chlorine in soil. Cl_{org} was abundant also in the soils examined in this study, and it was the dominant Cl form in both mineral soil and the organic layer at dry and moist sites. In this study Cl storage in the landscape was primarily driven by variation in tree biomass and thickness of soil layers, and more than twice as much Cl was stored in biomass and organic soil pools at moist and wet sites as compared to dry habitats among hill-slopes. Moreover, there was a positive correlation between Cl_{org} and % C (and soil C/N ratios) in the organic soil layer and the highest Cl_{org} concentrations were observed in soils with carbon content above 40 % (and a pH below 5.5). If this is related to a gradual accumulation of Cl_{org} remains to be studied. Thus, to estimate overall landscape Cl cycling and residence times it is necessary to characterize Cl^- and Cl_{org} uptake and storage in the most common habitats and vegetation communities.

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Appendix 1

A description of soil and vegetation characteristics for the three localities investigated in field study I and, some soil and vegetation chemistry.

Table A1-1. The three sampling sites, ecosystem type, their position and estimated height above sea level, groundwater level, and number of years above sea level. (Coordinates in SWEREF 99 18 00).

Locations	Site	North–South SWEREF 99 18 00	East–West SWEREF 99 18 00	Height above sea level (m)	Distance between circles (m)	Years above sea level
Young rich fen	LI	6697393.6	158771.2	3.6		600
(Labboträsk)	LII	6697336.4	158778.3	3.6	60	600
Older mire	SI	6695725.5	159989.1	8.6		1 360
(Stenrösmossen)	SII	6695618.8	160068.5	8.6	130	1 360
Norway spruce and alder wetland	AI	6696237.8	162449.9	2.3		390
(Alsumpskogen)	A2	6696245.9	162446.2	2.3	10	390

Table A1-2. Vegetation description of the moderately rich fen (Labboträsk), poor fen (Stenrösmossen) and Norway spruce and alder wetland (Alsumpskogen).

Locations	Site	Vegetation description
Rich fen (Labboträsk)	LI	Vegetation consists of bogbean, a lot of reed, marsh grass of Parnassus, tufted loosestrife, purple marshlocks, birch, alder, cranberry, willow, pointed spear-moss. Noteworthy that both locations at Labboträsk include *Warnstorfia* sp. och *Scorpidium* sp. as well as species indicative of groundwater flow, such as pointed spear-moss.
Rich fen (Labboträsk)	LII	Vegetation reed dominated with early marsh-orchid, tufted loosestrife, marsh spurge, purple marshlocks, bogmyrtle, tormentil, bogbean, cranberry, marsh grass of Parnassus, dwarf willow, birch, Scotch pine, willow, and pointed spear-moss. A lot of *Bryales* mosses at the bottom layer.
Poor fen (Stenrösmossen)	SI	Vegetation consists of wire sedge and a bit of carnation sedge, bogbean and a lot of bog-myrtle., *Bryales* dominates the bottom layer. Generally, only a little *Sphagnum*, but some tussocks of *Sphagnum magelanicum/rubrum. Warnstorfia* sp.
Poor fen (Stenrösmossen)	SII	*Sphagnum* more common than at the previous site, a little bit of *Warnstorfia* sp., wire sedge,and carnation sedge, bogbean, Water horsetail, cranberry, and a lot of bog-myrtle.
Norway spruce and alder wetland (Alsumpskogen)	AI	Thick topsoil on sand (20 cm). Partly stony. Spruce and alder on the edges. *Carex elata* tussocks with vegetation free areas in between. Vegetation consists of meadowsweet, tufted loosestrife, narrow buckler-fern, and wood sorel. Mosses mostly as tussocks; *Mnium* sp., Stairstep moss, and big shaggy-moss.
Norway spruce and alder wetland (Alsumpskogen)	AII	Mineral rich topsoil, sand at about 15–20 cm. Vegetation consists of Spruce, alder, ash shrubs, birch. meadowsweet, yellow loosestrife, stone bramble, fern, common self-heal, broad-leaved helleborine, wood sorel, false lily of the valley, marsh violet. big shaggy-moss, tree climacium moss, *Mnium* sp., and silesian feather-moss. Greater featherwort.

Table A1-3. Vegetation and soil chemistry at the three investigated sites in Field study I, where Labboträsk and Stenrösmossen are without a tree layer while Alsumpskogen has a tree layer.

Site		Soil	Cl_{tot} (µg g^{-1})	% Cl_{org} of Cl_{tot}	% C	% N	C/N ratio
Labboträsk	L1	0–10 cm	1 072	25	46.8	2.2	21.7
	LII		741	44	44.8	2.4	18.6
	LI	50 cm	2 397	36	43.2	3.1	13.9
	LII		1 229	23	46.5	2.4	19.6
Stenrösmossen	SI	0–10 cm	398	90	46.4	2.4	23.7
	SII		303	87	46.6	2.0	21.7
	SI	50 cm	1 005	100	45.6	3.6	12.8
	SII		287	100	47.3	2.0	23.5
Alsumpskogen	AI	0–10 cm	122	79	25.7	1.7	15.5
	AII		119	99	26.4	1.7	15.5
		50 cm	Not possible to sample due to stones at this depth				
Site		**Vegetation**	**Cl_{tot} (µg g^{-1})**	**% Cl_{org} of Cl_{tot}**	**% C**	**% N**	
Labboträsk	L1	Field layer	4 180	1.1	44.7	0.9	48.0
	LII		3 417	0.9	44.8	0.9	56.0
	L1	Bottom layer	1 490	16	43.5	1.7	26.0
	LII		1 476	15	41.4	1.3	31.1
Stenrösmossen	SI	Shrub-foliage	648	2.9	50.4	1.9	26.2
	SII		645	1.1	50.4	2.0	25.9
	SI	Shrub-wood	200	6.7	48.5	1.0	48.9
	SII		148	7.2	48.9	1.0	48.0
	SI	Field layer	1 349	1.4	45.6	1.0	45.2
	SII		819	1.1	45.0	1.1	39.4
	SI	Bottom layer	658	23	43.4	1.1	41.3
	SII		561	19	42.0	0.7	56.8
Alsumpskogen[1]		Tree-foliage[1]	417	4	48.3	2.5	19.4
		Tree-foliage[2]	862	2	48.5	0.9	51.8
		Tree-wood[1]	57	22	47	0.5	91.1
		Tree-wood[2]	62	38	48	< d.l.	
	AI	Field layer	12 283	0.8	22.9	1.9	22.9
	AII		10 128	1.1	22.6	1.9	22.6
	AI	Bottom layer	659	16	38.7	1.2	38.7
	AII		706	22	27.5	1.6	27.5

1. Alnus glutinosa

2. Picea abies

Figure A1-1. *Sampling sites for Field study I: moderately rich fen (Labboträsk; LI and LII), Norway spruce and alder wetland (Alsumpskogen; AI and AII), and poor fen (Stenrösmossen; SI and SII).*

Appendix 2

A description of soil and vegetation characteristics for the twelve localities investigated in field study II.

Table A2-1. Geographical position and elevation of sampling sites in field study II. The distance to the groundwater level and the time since the site emerged from the sea were estimated from the present height above the water surface of the closest lake and the height above the sea level, respectively. Dry: upland dry coniferous forest, Moist: moist coniferous forest, and Wet: wet alder forest wetland (Sweref 99 18 00).

Locations	Sampling Site	North–South SWEREF 99 18 00	East–West SWEREF 99 18 00	Elevation RH70 (m)	Depth to water (m)	Time since isolation (y)
Lillfjärden	Dry	6696915.4	162354.7	7.8	9.8	1 287
	Moist	6696770.1	162484.5	1.8	1.5	346
	Wet	6696712.8	162544.1	−0.2	0.2	11
Gällsboträsk	Dry	6697136.4	159339.2	5.9	5.6	1 002
	Moist	6697095.9	159372.1	2.9	3.8	535
	Wet	6696989.1	159490.1	1.7	0.0	339
Labboträsk	Dry	6697775.2	158879.6	7.0	5.2	1 180
	Moist	6697748.2	158944.9	4.0	3.8	700
	Wet	6697592.9	158921.9	3.7	1.8	660
Eckarfjärden	Dry	6695773.5	160584.6	7.8	1.9	1 296
	Moist	6695632.9	160648.4	6.2	1.0	1 054
	Wet	6695645.4	160695.6	5.2	0.1	897

Table A2-2. Measured soil depth at sampling, stone and boulder content at each plot. See methods for definition of dry, moist and wet, respectively.

		Soil layer depth (cm)		Stone/boulder volumetric content in 0–30 cm (%)
Locations	**Site**	**Litter**	**Org**	**Org**
Lillfjärden	Dry	2.0	16.0	56
	Moist	1.0	6.0	31
	Wet	3.0	11.0	34
Gällsboträsk	Dry	1.0	12.0	49
	Moist	2.0	19.0	24
	Wet	3.0	17.5	0
Labboträsk	Dry	2.5	12.5	36
	Moist	3.0	18.0	19
	Wet	3.0	30.0	
Eckarfjärden	Dry	2.5	18.0	44
	Moist	1.5	43.5	17
	Wet	2.0	19.0	

Table A2-3. The soil depth until a stone or a boulder was encountered. The depth was measured each meter along two perpendicular transects at each sampling site (n = 25). See methods for definition of dry, moist and wet, respectively.

		Soil depth (cm) (n = 25)
SiteLocations	**Site**	**Mean (min–max)**
Lillfjärden	Dry	7 (0–31)
	Moist	16 (0–29)
	Wet	19 (5–55)
Gällsboträsk	Dry	9 (0–16)
	Moist	20 (13–35)
	Wet	58 (32–90)
Labboträsk	Dry	15 (1–28)
	Moist	29 (12–50)
	Wet	65 (38–75)
Eckarfjärden	Dry	12 (2–23)
	Moist	57 (23–90)
	Wet	> 90

Table A2-4. Description of the vegetation and the species contained within the sampling squares for the twelve habitat sampling sites.

Lillfjärden – Upland dry coniferous forest

Spruce and moss dominated, even-aged, stony peatland forest with lingonberry, bilberry, stone bramble and eagle fern.

Species and their coverage in the sampling square

	Species		**Coverage (%)**
Shrub layer	Spruce	*Picea abies*	2
Field layer	Lingonberry	*Vaccinium vitis-idaea*	7
	Bilberry	*Vaccinium myrtillus*	2
	Wood violet	*Viola riviniana*	< 1
	Small cow-wheat	*Melampyrum sylvaticum*	< 1
	Sedge	*Carex sp.*	< 1
	Wavy hair-grass	*Avenella flexuosa*	< 1
Bottom layer	Stairstep moss	*Hylocomium splendens*	50
	Red-stemmed feather moss	*Pleurozium schreberi*	40
	Thyme moss	*Mnium sp.*	< 1
	Big shaggy-moss	*Rhytidiadelphus triquetrus*	< 1
Fungis		*Cudonia sp.*	< 1
	Marasmioid sp.	*Micromphale perforans*	< 1

Lillfjärden – Moist coniferous forest

Spruce dominated forest with trases of grazing, including some common hazel, ash, rowan and Scotch pine. Relatively rich in herbs, bottom layer moss covered., broad-leaved helleborine, mezereum.

Species coverage in the sampling square

	Species		Coverage (%)
Shrub layer	Rowan	*Acer platanoides*	< 1
Field layer	Wood violet	*Viola riviniana*	1
	Lilly of the valley	*Convallaria majalis*	4
	Hepatica	*Hepatica nobilis*	6
	Wood sorrel	*Oxalis acetosella*	3
	Herb-paris	*Paris quadrifolia*	1
	Mountain melic	*Melica nutans*	< 1
	Wild strawberry	*Fragaria vesca*	2
	Germander speedwell	*Veronica chamaedrys*	< 1
	False lily of the valley	*Maianthemum bifolium*	1
Bottom layer	Stair step moss	*Hylocomium splendens*	40
	Red-stemmed feather moss	*Pleurozium schreberi*	25
	Thyme moss	*Mnium sp.*	5
	Rose moss	*Rhodobryum roseum*	5

Lillfjärden – Wet alder forest wetland

Tree layer dominated by alder, shrub layer with some ash, and field layer dominated by hemp-agrimony, grass and meadowsweet.

Species coverage in the sampling square

	Species		Coverage (%)
Shrub layer	Ash	*Fraxinus excelsior*	20
	Raspberry	*Rubus idaeus*	5
Field layer	Hemp-agrimony	*Eupatorium cannabinum*	50
	Meadowsweet	*Filipendula ulmaria*	10
	Bearded Couch	*Elymus caninus*	10
	Meadow buttercup	*Ranunculus acris*	< 1
	Common valeriana	*Valeriana sambucifolia*	< 1
	Heath dog-violet	*Viola canina*	< 1
Fungi	Galerina	*Galerina sp.*	< 1
	Paxilllus	*Paxillus sp.*	< 1

Eckarfjärden – Upland dry coniferous forest

Spruce dominated stony forest with some oak, birch, rowan, Scotch pine, common hazel, European beech, and rosehip, Some ferns and a lot of bilberry and lingonberry.

Species coverage in the sampling square

	Species		Coverage (%)
Field layer	Bilberry	*Vaccinium myrtillus*	25
	Lingonberry	*Vaccinium vitis-idaea*	15
	Wavy hair-grass	*Avenella flexuosa*	7
	Segde	*Carex digitata*	1
Bottom layer	Stair step moss	*Hylocomium splendens*	60
	Big shaggy moss	*Rhytidiadelphus triquetrus*	25
	Red-stemmed feather moss	*Pleurozium schreberi*	8
	Knights plume moss	*Ptilium crista-castrensis*	1
Fungi	Goatcheese webcap	*Cortinarius camphoratus*	1
	-	*Cortinarius renidens*	1

Eckarfjärden – Moist coniferous forest

Spruce dominated stony forest with some alder, birch, Scotch pine, common hazel, pteridophytes, horsetail, yellow loosestrife, herb-paris, outer parts delineated by wetter area, Scotch pine stands in the dryer part of the area.

Species coverage in the sampling square

	Species		Coverage (%)
Shrub layer	Rowan	*Sorbus aucuparia*	< 1
Field layer	Lingonberry	*Vaccinium vitis-idaea*	5
	Bilberry	*Vaccinium myrtillus*	2
	Stone bramble	*Rubus saxatilis*	3
	False lily of the valley	*Maianthemum bifolium*	< 1
	Yellow loosestrife	*Lysimachia vulgaris*	7
	Meadow horsetail	*Equisetum pratense*	4
	Small cow-wheat	*Melampyrum sylvaticum*	< 1
	Chickweed-wintergreen	*Trientalis europaea*	< 1
	-	*Poaceae sp.*	1
	Mezereum	*Daphne mezereum*	< 1
Bottom layer	Big shaggy-moss	*Rhytidiadelphus triquetrus*	89
	Red-stemmed feather moss	*Pleurozium schreberi*	1

Eckarfjärden – Wet alder forest wetland

Alder dominated swamp at the edge of a lake. Some tussocks of carex, marsh horsetail and larger herbs, such as yellow loosestrife. 20 m to reedbelt.

Species coverage in the sampling square

	Species		Coverage (%)
Shrub layer	Alder	*Alnus glutinosa*	1
	Spruce	*Picea abies*	< 1 %
Field layer	Marsh fern	*Thelypteris palustris*	8
	Meadowsweet	*Filipendula ulmaria*	20
	Water horsetail	*Equisetum fluviatile*	1
	Common marsh bedstraw	*Galium palustre*	< 1
	Veronica species	*Veronica sp.*	< 1
	Bogbean	*Menyanthes trifoliata*	2
	Yellow loosestrife	*Lysimachia vulgaris*	6
	Sedge	*Carex sp.*	6
	Marsh violet	*Viola palustris*	1
Bottom layer	Thyme moss	*Mnium sp.*	3
	Silesian feather-moss	*Herzogiella seligeri*	15
	Dented silk-moss	*Plagiothecium denticulatum*	1
Fungi	-	*Conocybe sp.*	

Gällsboträsk – Upland dry coniferous forest

Alder dominated swamp with tussocks at the edge of a lake. Vegetation includes marsh horsetail and larger herbs, such as yellow loosestrife. 20 m to reedbelt.

Species coverage in the sampling square

	Species		Coverage (%)
Field layer	Bilberry	*Vaccinium myrtillus*	15
	Lingonberry	*Vaccinium vitis-idaea*	3
	Stone bramble	*Rubus saxatilis*	2
	Wavy hair-grass	*Avenella flexuosa*	2
	Small cow-wheat	*Melampyrum sylvaticum*	< 1
	Hairy wood-rush	*Luzula pilosa*	< 1
Bottom layer	Knights plume moss	*Ptilium crista-castrensis*	22
	Stairstep moss	*Hylocomium splendens*	50
	Rose moss	*Rhodobryum roseum*	3
	Red-stemmed feather moss	*Pleurozium schreberi*	20
Fungi	Amethyst deceiver	*Laccaria amethystina*	< 1
	-	*Mycena sp.*	< 1
	-	*Conocybe sp.*	< 1

Gällsboträsk – Moist coniferous forest

Spruce dominated forest with some common hazel, elm, ash, rowan and honeysuckle. Includes a batch with smaller spruce, Calamagrostis arundinacia, mountain currant, herb-paris, stone bramble and narrow buckler-fern.

Species coverage in the sampling square

	Species		Coverage (%)
Shrub layer	Mountain currant	*Ribes alpinum*	1
Field layer	Mountain melick	*Melica nutans*	5
	Cock's Foot	*Dactylis glomerata*	5
	Hairy wood-rush	*Luzula pilosa*	1
	Heath dog-violet	*Viola canina*	1
	Wood sorel	*Oxalis acetosella*	3
	Small cow-wheat	*Melampyrum sylvaticum*	< 1
	False lily of the valley	*Maianthemum bifolium*	< 1
Bottom layer	Greater Fork-moss	*Dicranum majus*	60
	Stairstep moss	*Hylocomium splendens*	18
	Rose moss	*Rhodobryum roseum*	15
	Big shaggy-moss	*Rhytidiadelphus triquetrus*	1
	Red-stemmed feather moss	*Pleurozium schreberi*	1
Fungi	-	*Mycena sp.*	< 1

Gällsboträsk – Wet alder forest wetland

Area between spruce forest and reedbelt. Tree layer dominated by alder. Some tussocks. Field layer dominated by sedges, Talictrum flavum, water horsetail, iris and gypsywort.

Species coverage in the sampling square

	Species		Coverage (%)
Field layer	Fibrous tussock-sedge	*Carex appropinquata*	25
	Meadowsweet	*Filipendula ulmaria*	7
	Tufted loosestrife	*Lysimachia thyrsiflora*	2
	Sedge	*Carex sp.*	2
	Common marsh bedstraw	*Galium palustre*	< 1
Bottom layer	Silesian feather-moss	*Herzogiella seligeri*	10
	Thyme moss	*Mnium spp.*	2
Fungis	Milkycap	*Lactarius sp.*	

Labboträsk – Upland dry coniferous forest

Spruce, Scotch pine, and bilberry dominated forest with some lingonberry and mosses (stairstep moss and red-stemmed feather moss). Moss covered stones occupy 6 % of the area.

Species coverage in the sampling square

	Species		Coverage (%)
Field layer	Bilberry	*Vaccinium myrtillus*	70
	Lingonberry	*Vaccinium vitis-idaea*	5
Bottom layer	Stairstep moss	*Hylocomium splendens*	45
	Red-stemmed feather moss	*Pleurozium schreberi*	25
	cypress-leaved plaitmoss	*Hypnum cupressiforme*	1
	Greater Fork-moss	*Dicranum majus*	1
	Knights plume moss	*Ptilium crista-castrensis*	2
	Big shaggy-moss	*Rhytidiadelphus triquetrus*	< 1
Fungi	Gassy webcap	*Cortinarius traganus*	2
	-	*Conocybe sp.*	< 1
Moss covered stones			6

Labboträsk – Moist coniferous forest

Species coverage in the sampling square

	Species		Coverage (%)
Field layer	Bilberry	*Vaccinium myrtillus*	5
	Lingonberry	*Vaccinium vitis-idaea*	2
	Yellow loosestrife	*Lysimachia vulgaris*	1
	Purple small-reed	*Calamagrostis canescens*	5
	Heath dog-violet	*Viola canina*	3
	Tormentil	*Potentilla erecta*	2
	Twinflower	*Linnaea borealis*	5
	Blue sedge	*Carex flacca*	< 1
	False lily of the valley	*Maianthemum bifolium*	< 1
	Wild strawberry	*Fragaria vesca*	< 1
	Wavy hair-grass	*Avenella flexuosa*	3
Bottom layer	Greater featherwort	*Plagiochila asplenioides*	2
	Bog groove-moss	*Aulacomnium palustre*	< 1
	Greater fork-moss	*Dicranum majus*	2
	Stairstep moss	*Hylocomium splendens*	2
	Knights plume moss	*Ptilium crista-castrensis*	50

Labboträsk – Wet alder forest wetland

Reedbelt, dominating trees Scotch pine and alder. Also birch and some ash, rowan, and buckthorn. Field layer dominated by reed and sedge with some bogmyrtle and meadowsweet.

Species coverage in the sampling square

	Species		Coverage (%)
Field layer	Reed	*Phragmites australis*	8
	Lesser tussock-sedge	*Carex diandra*	8
	Tufted loosestrife	*Lysimachia thyrsiflora*	< 1
	Common marsh bedstraw	*Galium palustre*	< 1
	Purple small-reed	*Calamagrostis canescens*	< 1
	Water horsetail	*Equisetum fluviatile*	< 1
Bottom layer	Pointed Spear-moss	*Calliergonella cuspidata*	75
Fungi	Milkcap	*Lactarius sp.*	1

Table A2-5. Basal area weighted estimate of tree biomass and general description of sample trees in Field study II (the tree, also the dominating tree species, next to the plot for sampling of foliage, phloem and wood). Stand tree age was estimated from the tree with largest diameter at breast height (DBH), from which a tree core (5 mm diameter) was sampled. Dry: spruce foliage in upland dry coniferous forest, Moist: spruce foliage in moist coniferous forest, Wet: alder and birch forest in wet alder forest wetland.

						Basal area weighted estimates	
Location	**Site**	**Age**	**Height (m)**	**Circumference (m)**	**Tree growth (mm year^{-1}) average [last year]**	**Stem biomass (kg dry weight m^{-2})**	**Foliage biomass (kg dry weight m^{-2})**
Large trees (old tree)							
Lillfjärden	Dry	59	20.2	1.0	1.27 [1.25]	12.1	1.9
	Moist	88	28.2	1.6	1.65 [1.20]	14.4	1.5
	Wet	49	15.6	0.9	1.74 [1.90]	23.7	0.5
Gällsboträsk	Dry	88	28.2	1.5	1.60 [1.70]	23.0	2.1
	Moist	85	29.4	1.7	0.93 [0.95]	42.6	4.3
	Wet	62	19.6	1.2	1.68 [1.85]	62.1	1.0
Labboträsk	Dry	155	25.8	1.3	1.42 [1.35]	26.1	2.7
	Moist	101	26.2	1.2	0.64 [0.75]	22.0	2.5
	Wet	139	14.6	1.0	1.2 [1.35]	21.8	1.2
Eckarfjärden	Dry	132	20.2	1.1	1.70 [1.60]	27.3	3.8
	Moist	118	24.5	1.3	0.40 [0.45]	46.3	4.3
	Wet	62	15.6	0.8	1.12 [1.35]	48.7	2.3
Central tree (young tree)							
Lillfjärden	Dry	37	13.7	0.5	0.90 [0.55]		
	Moist	22	4.0	0.2	0.48 [0.55]		
	Wet	35	11.5	0.5	Missing		
Gällsboträsk	Dry	99	21.0	0.9	0.70 [0.60]		
	Moist	15	2.0	0.1	1.14 [1.20]		
	Wet	31	6.3	0.2	0.89 [0.75]		
Labboträsk	Dry	112	19.3	0.9	0.93 [1.15]		
	Moist	75	4.7	0.2	0.12 [0.14]		
	Wet (*birch*)	21	8.7	0.3			
	Wet (*alder*)	47	14.0	0.9 1.10 [1.30]	1.61 [1.5]		
Eckarfjärden	Dry	99	10.8	0.4	1.61 [1.50]		
	Moist	64	6.6	0.3	0.35 [0.35]		
	Wet	43	5.2	0.3	0.96 [0.80]		

Table A2-6. Area-estimated biomass (dry matter, DM) in the different biomass layers of the localities in field study II. Dry: spruce foliage in upland dry coniferous forest, Moist: spruce foliage in moist coniferous forest, Wet: alder and birch forest in wet alder forest wetland. (-) indicates that no vegetation of that type was present in habitat sampling square.

Site	Gradient	Shrub layer (kg DM m^{-2})	Field layer (kg DM m^{-2})	Bottom layer (kg DM m^{-2})	Total understory (kg DM m^{-2})
Lillfjärden	Dry	-	0.023	0.613	**0.636**
	Moist	-	0.038	0.132	**0.170**
	Wet	0.065	0.196	-	**0.261**
Gällsboträsk	Dry	-	0.054	0.269	**0.323**
	Moist	0.026	0.030	0.237	**0.293**
	Wet	-	0.101	0.040	**0.142**
Labboträsk	Dry	-	0.030	0.044	**0.074**
	Moist	-	0.133	0.105	**0.239**
	Wet	-	0.133	0.013	**0.147**
Eckarfjärden	Dry	-	0.158	0.208	**0.374**
	Moist	-	0.060	0.208	**0.268**
	Wet	0.012	0.049	0.045	**0.106**

Figure A2-1. *Sampling sites and vegetation types (dominating species) at Lillfjärden. Dry: spruce forest of bilberry type (Picea abies, V. vitis-idaea, V. myrtillus, H. splendens, P. schreberi), Moist: spruce forest of low herb type (Picea abies/Pinus sylvestris, H. nobilis, C. majalis, H. splendens, P. schreberi), Wet: wet alder forest of herb type (Alnus glutinosa, E. canabinum, F. ulmaria).*

Figure A2-2. *Sampling sites and vegetation types (dominating species) at Gällsboträsk. Dry: spruce forest of bilberry type (Picea abies, Pinus Sylvestris, V. myrtillus, V. vitis-idaea, H. splendens, P. crista-castrensis), Moist: spruce forest with more herbs (Picea abies, Pinus Sylvestris, M. nutans, D. glomerate, D. majus, H. splendens), Wet: wet alder forest of herb type (Picea abies, Pinus Sylvestris, C. appropinquata, F. ulmaria, H. seligeri, Mnium sp.).*

Figure A2-3. *Sampling sites and vegetation types (dominating species) at Labboträsk. Dry: spruce forest of bilberry type (Pinus Sylvestris, Picea abies, V. myrtillus, V. vitis-idaea, H. splendens, P. schreberi), Moist: spruce forest with more herbs (Picea abies, Pinus Sylvestris, V. myrtillus, C. canescens, P. crista-castrensis, H.), Wet: wet alder forest of herb type (Pinus Sylvestris, Betula pubescens, P. australis, C. diandra, C. cuspidate).*

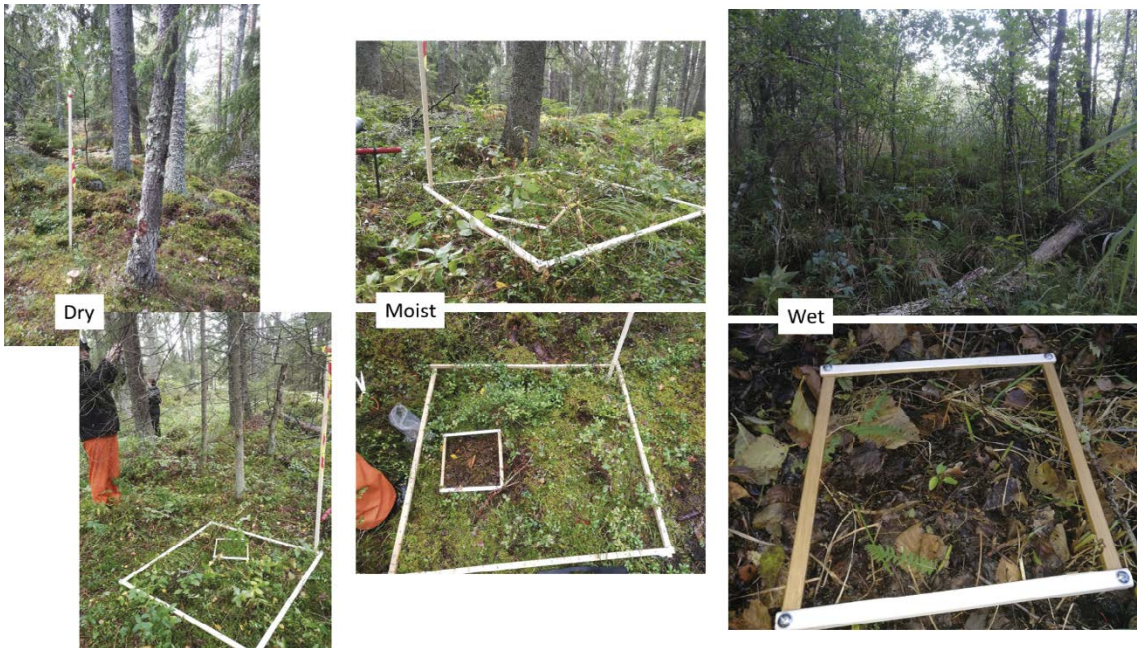

Figure A2-4. *Sampling sites and vegetation types (dominating species) at Eckarfjärden. Dry: spruce forest of bilberry type (Picea abies, Pinus Sylvestris, V. vitis-idaea, V. myrtillus, H. splendens, R. triquetrus) Moist: spruce forest with more herbs (Picea abies, Pinus Sylvestris, L. vulgaris, V. vitis-idaea, R. triquetrus, P. schreberi) Wet: wet alder forest of herb type (Alnus glutinosa, Betula pubescens, F. ulmaria, T. palustris, H. seligeri, Mnium sp.).*

Appendix 3

Total Cl in dominating field layer species of field study II.

Table A3-1. Total Cl (Cl_{tot} = organic + inorganic Cl) in bulk samples shrub, field and bottom layer of field study II.

		Cl_{tot} (µg g^{-1})		
		Dry	**Moist**	**Wet**
Shrub layer	Lillfjärden	-	668	1969
	Gällsboträsk	-	2398	-
	Labboträsk	-	-	-
	Eckarfjärden	-	3051	359
Field layer	Lillfjärden	504	9017	7056
	Gällsboträsk	1963	14010	12213
	Labboträsk	816	4099	7700
	Eckarfjärden	1193	9740	14073
Bottom layer	Lillfjärden	264	751	751
	Gällsboträsk	770	900	579
	Labboträsk	371	306	415
	Eckarfjärden	191	1344	218

Table A3-2. Cl concentrations among the dominating field layer species in the plots. Dry: upland dry coniferous forest, Moist: moist coniferous forest, and Wet: wet alder forest wetland.

			Cl_{tot} (µg g^{-1} DM)		
Dominating species		**Site**	**Dry**	**Moist**	**Wet**
Shrubs	*Vaccinium vitis-idaea*	Lillfjärden	206		
	Vaccinium vitis-idaea	Gällsboträsk	131		
	Vaccinium vitis-idaea	Labboträsk	185	233	
	Vaccinium vitis-idaea	Eckarfjärden	253	820	
	Vaccinium myrtillus	Lillfjärden	1629		
	Vaccinium myrtillus	Gällsboträsk	1337		
	Vaccinium myrtillus	Labboträsk	864		
	Vaccinium myrtillus	Eckarfjärden	1094		
Grass/Sedge	*Carex*	Gällsboträsk			10772
	Carex	Labboträsk			8283
	Carex	Eckarfjärden			3617
	Calamagrostis canescens	Labboträsk		5956	
	Elymus caninus	Lillfjärden			> 20000
	Phragmites australis	Labboträsk			7257
Herbs	*Eupatorium cannabinum*	Lillfjärden			5587
	Filipendula ulmaria	Lillfjärden			1679
	Filipendula ulmaria	Gällsboträsk			14270
	Filipendula ulmaria	Eckarfjärden			10667
	Thelypteris palustris	Eckarfjärden			12867
	Linnaea borealis	Labboträsk		1474	
	Lysimachia vulgaris	Eckarfjärden		15886	

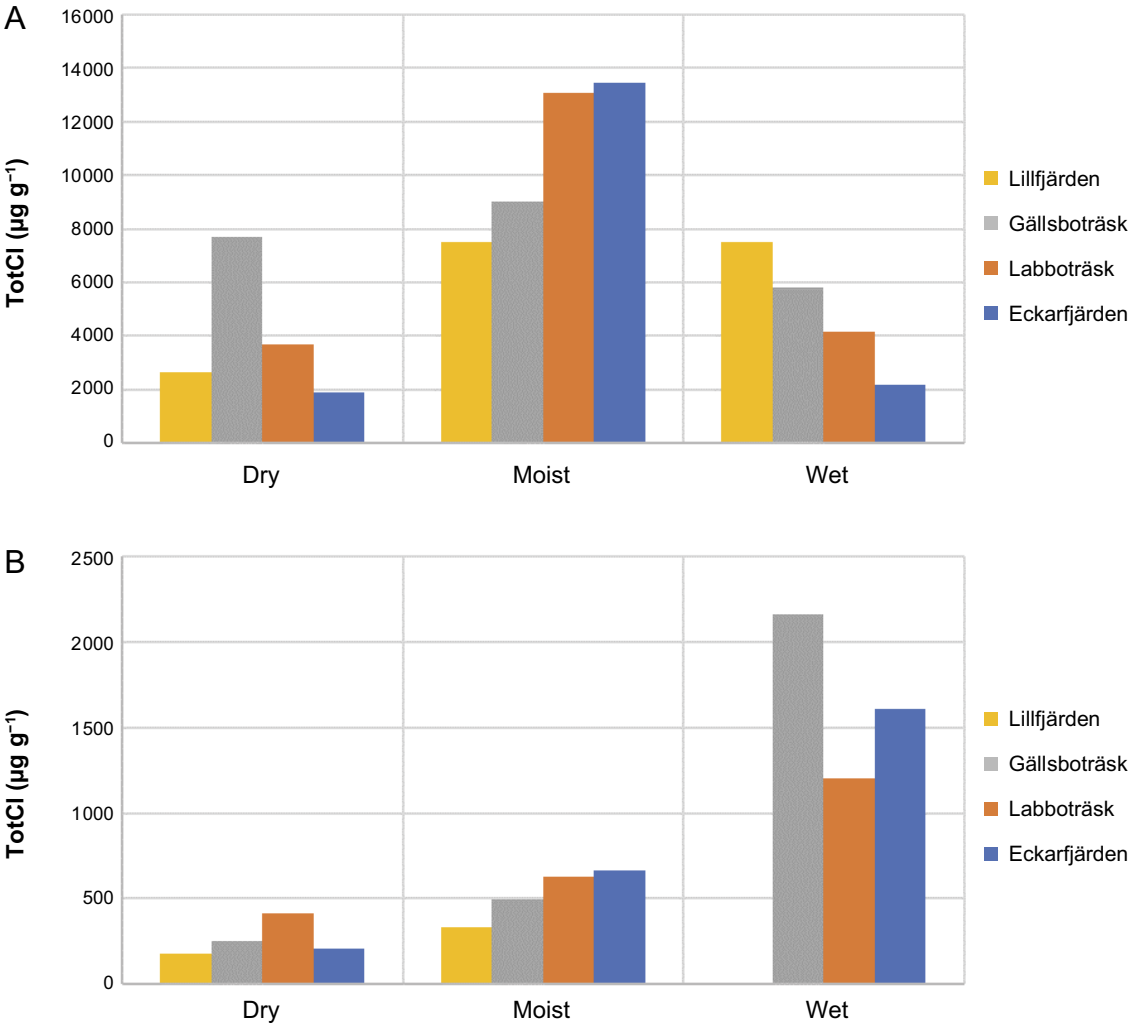

Figure A3-1. *Total Cl concentrations in the bottom layer. Bottom layer bulk sample (Panel A) and Bottom layer Shoot (Panel B).*

Arkitektkopia AB, Bromma, 2021

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