Concentration and fluxes of dissolved organic carbon (DOC) in three Norway spruce stands along a climatic gradient in Sweden

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Abstract. Leaching of dissolved organic carbon (DOC) from the forest floor and transport in soil solution into the mineral soil are important for carbon cycling in boreal forest ecosystems. We examined DOC concentrations in bulk deposition, throughfall and in soil solutions collected under the O and B horizons in three Norway spruce stands along a climatic gradient in Sweden. Mean annual temperature for the three sites was 5.5, 3.4 and 1.2 °C. At each site we also examined the effect of soil moisture on DOC dynamics along a moisture gradient (dry, mesic and moist plots). To obtain information about the fate of DOC leached from the O horizon into the mineral soil, ¹⁴C measurements were made on bulk organic matter and DOC. The concentration and fluxes of DOC in O horizon leachates were highest at the southern site and lowest at the northern. Average DOC concentrations at the southern, central and northern sites were 49, 39 and 30 mg l⁻ respectively. We suggest that DOC leaching rates from O horizons were related to the net primary production of the ecosystem. Soil temperature probably governed the within-year variation in DOC concentration in O horizon leachates, but the peak in DOC was delayed relative to that of temperature, probably due to sorption processes. Neither soil moisture regime (dry, mesic or moist plots) nor seasonal variation in soil moisture seemed to be of any significance for the concentration of DOC leached from the O horizon. The ¹⁴C measurements showed that DOC in soil solution collected below the B horizon was derived mainly from the B horizon itself, rather than from the O horizon, indicating a substantial exchange (sorption-desorption reactions) between incoming DOC and soil organic carbon in the mineral soil.

Introduction

Forest soils of northern latitudes cover extensive areas and contain significant amounts of carbon. According to a recent inventory of organic carbon pools in boreal forest soils in Scandinavia, 70–80% of the organic carbon in the upper 100 cm is normally found in the mineral soil (Callesen et al. 2003). Dissolved organic carbon (DOC) leached from the forest floor is a major source of the carbon input to the mineral soil (McDowell and Likens 1988; Qualls et al.

1991; Guggenberger et al. 1994; Neff and Asner 2001). Typical concentrations in soil solution leaving the forest floor are in the range 20–90 mg l⁻¹, whereas the concentration normally found in the mineral soil is about 2-35 mg l⁻¹ (Michalzik et al. 2001). This corresponds to a net removal of DOC from forest floor solution by the upper mineral soil layers (A/E plus B horizons) of $10-30~{\rm g~carbon~m^{-2}~yr^{-1}}$. The retention is widely thought to be caused mainly by physico-chemical processes such as sorption and/or precipitation (summarised by Kalbitz et al. 2000). In contrast, the production of DOC in the forest floor is thought to be controlled mainly by biological processes (decomposition of litter and humus, root exudation), implying a high sensitivity to changes in soil temperature and moisture (Kalbitz et al. 2000). Knowledge of factors and processes regulating the production and transport of DOC in soils is therefore important for the prediction of soil carbon stocks under a varying climate. The role of temperature and soil moisture in regulating DOC production in O horizon materials has been demonstrated in several laboratory studies. For example, Christ and David (1996) and Gödde et al. (1996) showed that DOC production increased with both increasing soil temperature and moisture. This is in accordance with results from field studies, which frequently show a seasonal pattern with the highest DOC concentration in late summer (Michalzik and Matzner 1999; Solinger et al. 2001; Kaiser et al. 2002). Other studies have shown that drying and rewetting also affects DOC leaching (e.g. Tipping et al. 1999; Schaumann et al. 2000). To date, however, most studies have focused on the short-term dynamics and relatively little is known about the long-term response to climatic factors such as temperature and precipitation. In other words, only few published field studies have dealt with the comparison of concentrations and fluxes of DOC in ecosystems under different climates. The complexity of the processes controlling DOC production and transport at ecosystem level was illustrated in a recent review by Michalzik et al. (2001). Data from 42 ecosystem studies performed mainly in Europe and the USA were compiled and no correlation was found between mean annual temperature and DOC concentrations or fluxes in soil leachates collected below the O horizon, although the former varied in the range 1–16 °C. Thus, at ecosystem level other factors are likely to interact with temperature in controlling DOC concentrations and fluxes in forest floor leachates.

The peak of ¹⁴CO₂ in the atmosphere in the mid 1960 s, caused by the upper atmospheric testing of thermonuclear weapons ('bomb carbon'), offers a unique possibility of tracing the origin and fate of DOC entering the mineral soil. DOC derived from carbon fixed prior to 1950 is depleted in ¹⁴C due to the radioactive decay of the isotope, whereas DOC derived from carbon fixed after 1950 is enriched in ¹⁴C due to the bomb carbon effect. To date, only a few studies have dealt with the ¹⁴C content of DOC sampled from intact soil profiles. In an early study, Trumbore et al. (1992) reported DO¹⁴C values for solutions sampled from O, Ah B1, B2 and B3 horizons of a Spodosol in Ontario, Canada, indicating a significant amount of bomb carbon in DOC sampled from all horizons. This contrasts with the results of a recent study on

¹⁴C content of DOC and solid phase organic matter in a Norwegian Podzol (Michalzik et al. 2003), showing a decreasing ¹⁴C content with depth in both DOC and solid phase organic matter. Data on ¹⁴C content in DOC and in various pools of solid phase organic matter can also be used quantitatively in model parameterisations in order to constrain the model (Michalzik et al. 2003).

Here we present data on concentrations and fluxes of DOC in bulk deposition, throughfall and soil solutions collected from O and B horizons at three sites located along a climatic gradient (57–64°N). The sites were chosen to keep other variables (geology, pedology, vegetation, stand age, site history, etc.) as equal as possible between sites. Each site is characterised by a hydrological gradient from moist to dry conditions. This allows us to study not only the response of DOC to variations in temperature and soil moisture depending on weather conditions and season, but also the effects of different climates (i.e. long-term effects) on DOC leaching. We also use ¹⁴C determinations on DOC and solid-phase organic matter to obtain information on the origin and fate of DOC leached from the forest floor at two of the sites.

Materials and methods

Description of sites

Field measurements were conducted at three sites along a transect from south to north in Sweden: Asa (57°08′N), Knottåsen (61°00′N) and Flakaliden (64°07′N). The sites are identical to the common experimental field sites of the LUSTRA (Land Use Strategies for Reducing Net Greenhouse Gas Emissions) research programme, which were chosen with the intention of establishing a climatic gradient, whilst maintaining other environmental parameters as

Table 1. Site characteristics (Berggren et al. 2004).

	Asa	Knottåsen	Flakaliden
Latitude	57°08′N	61°00′N	64°07′N
Longitude	14°45′E	16°12′E	19°27′E
Altitude (m.a.s.l.)	200	320	320
Mean annual temperature (°C) ^a	5.5	3.4	1.2
Length of vegetation period (days) ^b	190	160	120
Mean annual precipitation (mm) ^a	688	613	523
Nitrogen deposition (kg ha ⁻¹ yr ⁻¹)	10	4	2
Average C/N ratio in O horizon	29.7	34.0	39.6
Major tree species	Picea abies	Picea abies	Picea abies
Stand age (years)	38	38	41
Soil type according to FAO (1990)	Podzols	Podzols	Podzols

^aLong-term averages (1961–1990) from nearest official meteorological station (Asa: Berg. Knottåsen: Åmotsbruk. Flakaliden: Kulbäcksliden).

^bDays with average temperature > 5 °C.

constant as possible. Properties of the sites are summarised in Table 1. The range in mean annual temperature is 1.2-5.5 °C and in mean annual precipitation 523-688 mm. Norway spruce (Picea abies (L.) Karst.) is the dominating tree species at all sites and the stands are 38-41 years old. Base saturation and pH of the soils are given in Table 2. All sites are situated on acidic bedrock and the soils are Podzols developed on sandy loamy tills. According to FAO (1990), soils at the moist plots are Gley Podzols and at the mesic and dry plots Haplic Podzols. The O horizon at all sites have a mor-type organic layer, typically with a depth of 3–10 cm. There is more carbon in the O horizon and in the upper 50 cm at Asa than at the two northern sites (Table 3), which is consistent with the general national pattern (Callesen et al. 2003). Estimated mean forest production (total volume over bark from stump to tip) at Asa, Knottåsen and Flakaliden is 10.1, 6.3 and 3.2 m³ yr⁻¹ ha⁻¹, respectively (Berggren et al. 2004). Nitrogen deposition (NO₃⁻-N plus NH₄⁺-N) based on the county levels is 10 kg ha⁻¹ yr⁻¹ at Asa, 4.1 kg ha⁻¹ yr⁻¹ at Knottåsen and 2.3 kg ha⁻¹ yr⁻¹ at Flakliden (data from IVL, Swedish Environmental Institute, http:// www.ivl.se). Sulphur deposition is also higher at Asa than at the two northern sites.

Table 2. Base saturation (BS) and pH(H₂O) in soil profiles at Asa, Knottåsen and Flakaliden.

Site Horizon		pH (H ₂ O)	BS (%)	
Asa	O	3.55	66	
	Mineral soil (5–25 cm)	4.33	5.9	
Knottåsen	0	4.15	83	
	B (0–20 cm)	4.78	28	
Flakaliden	O	4.18	80	
	B (0–20 cm)	5.01	42	

Sampling is based on horizon at Knottåsen and Flakaliden and on depth in mineral soil at Asa due to a lack of well-defined A and E horizons. Data from Berggren et al. (2004).

Table 3. Average carbon stocks per moisture class in kg carbon m^{-2} , (n=3) corrected for stone and boulder frequency.

Horizon/depth	Site	Moisture regime			
		Dry	Mesic	Moist	
0	Asa	2.5 Ab	2.2 Ab	12.6 Aa	
	Knottåsen	1.3 ABb	1.2 Bb	3.7 Ba	
	Flakaliden	0.9 Bb	0.8 Bb	2.1 Ba	
0-50 cm	Asa	9.1 Ab	8.6 Ab	21.2 Aa	
	Knottåsen	5.0 Bb	5.5 Bb	9.0 Ba	
	Flakaliden	5.1 Bb	5.4 Bb	10.2 Ba	

^{0–50} cm refers to carbon stocks down to a depth of 50 cm (O horizon included). Different letters refer to statistically significant differences within the same depth. Upper case letters refer to differences between sites and lower case letters to differences between moisture regimes. Data from Berggren et al. (2004).

The major tree species at all sites is Norway spruce (*P. abies* (L.) Karst) occasionally mixed with Scots pine (*Pinus sylvestris* L.). The sites were clear-cut in 1963 (Flakaliden and Knottåsen) or 1966 (Asa). Present stands were planted in 1963 (Flakaliden), 1965 (Knottåsen) or 1967 (Asa) with Norway spruce seedlings. The former forest generations were dominated by Norway spruce. Land-use history of one of the mesic plots in Asa differed from the others. Prior to the present Norway spruce stand, it was an arable field.

Experimental design

At each site there is a moisture gradient from dry via mesic to moist conditions, i.e. from deep lying to shallow groundwater levels. Three plots $(30 \times 30 \text{ m})$ per moisture class – dry, mesic and moist – were established at each site. When selecting location of plots, transects along the slopes were selected in order to cover the slope areas. The positions of plots along the transects were selected subjectively based on (i) position along the slope, and (ii) the composition of field and bottom layer vegetation. The species used for indicating dry conditions were *Cladonia* sp. (L.) and moist conditions *Sphagnum* sp. (L.) The dwarf shrubs *Vaccinium myrtillus* (L.) and *Vaccinium vitis-idaéa* (L.) were generally frequent at both dry and mesic plots, but less frequent at moist plots. The denomination of the moisture classes follows the one used by the National Survey of Forest Soils and Vegetation and is based on average depth to the groundwater level during the vegetation period; dry > 2 m, mesic 1–2 m and moist < 1 m.

At each of the nine plots per site, three lysimeters were installed below the O horizon and two below the B horizon, i.e. 9 lysimeters below the O horizon and 6 below the B horizon were used per moisture class and site. Each plot had an inner area of 15×15 m for non-destructive sampling such as soil solution, soil temperature and soil moisture measurements. At Asa, the mesic plot that had been subjected to a different land-use was omitted in all analyses.

Sampling and analyses of solutions

Sampling of O horizon leachates was carried out with zero tension lysimeters (30 cm²), made of plexiglass and polyethene (PE) netting. The lysimeters were horizontally installed directly beneath the O horizon with the aim of minimizing the disturbance of the forest floor. Soil solution samples were collected in 1 l polyethene bottles, which were placed in PVC tubes below ground to keep samples cool. Soil solution from the lower part of the B horizon (40–50 cm soil depth) was continuously sampled with tension lysimeters (teflon/quarts, Prenart super quarts, vacuum 50 kPa). Solutions were collected in

2 l borosilicate glass bottles, which were also placed in PVC tubes below ground. To allow the lysimeters to equilibrate with the soil solution, solutions obtained within the first 6 months after installation were discarded. Bulk deposition was sampled in a forest clearing adjacent to the plots using four continuously open PE funnel collectors (20 cm diameter) with PE sieves at the bottom, placed 1.7 m above ground. Solutions were collected in shaded PE bottles placed on the ground. At one plot per moisture class and site, ten collectors of throughfall were randomly distributed below the canopy but > 1 m away from tree trunks. Sampling of bulk deposition, throughfall and soil solutions was carried out bi-weekly during the period June 2000 to November/December 2001.

All handling of field equipment and samples was performed in a '14°C clean' laboratory (Jädraås experimental forest). Water volume (lysimeter solutions) and pH were determined in the laboratory immediately after each collection. Bulk deposition, throughfall and soil solutions collected from the same plots were pooled by volume prior to analysis. An aliquot for DOC analysis was filtered through 0.2 μ m filters (Acrodisc PF, Gelman Sciences, MI) and acidified to pH 3 with HCl. DOC measurements were made by a Shimadzu TOC-5000 A analyser, usually within 1 week of sampling. The samples were stored at +2 °C until analysis.

Determining 14C content of DOC and solid soil organic matter

Soil samples for 14 C analyses were collected at one of the mesic plots at Asa and Knottåsen. At Asa samples were taken from Oe, Oa, Ah and B horizons, and at Knottåsen from Oe, Oa and B horizons. Samples from mineral soil horizons were taken from the walls of four pits, which were dug one at each side of the plot. Samples from the O horizon were taken with a metal cylinder (diameter 5 cm) in a transect along the plot (n = 10). To remove stones and roots, samples were passed through a 4 mm (O horizon samples) or 2 mm (mineral soil horizons) sieve. Prior to 14 C analyses, the samples were pooled to one sample per horizon. Soil solutions for DO 14 C analysis were collected on one sampling occasion (October 2001) from the lysimeters at the same plots.

Measured volumes of filtered lysimeter solutions were rotary-evaporated in acid-washed, sample-rinsed glassware to within a few ml of solution. This solution was freeze-dried before being combusted to CO_2 (Boutton et al. 1983). A sub-sample of CO_2 was used to measure stable carbon isotope ratios using a dual-inlet gas source mass spectrometer (VG OPTIMA) calibrated with international reference materials (Craig 1957) and with an overall precision of $\pm 0.1\%$, based on repeated measurements of standards. A further sub-sample of CO_2 was converted to a graphite/iron mixture by iron/zinc reduction (Slota et al. 1987) and sent to the NSF-AMS Facility, University of Arizona, USA for ^{14}C analysis (Donahue 1990). Accelerator mass spectrometry results are reported as % modern absolute ^{14}C , taking into account the ongoing radioactive

decay of the international reference standard (oxalic acid) since 1950 AD (Stuiver and Polach 1977). ¹⁴C enrichment > 100% modern absolute indicates unambiguously the presence of ¹⁴C from atomic weapons testing (i.e. post 1955 AD).

Bulk soil samples were oven-dried and prepared to benzene for ¹⁴C analysis by liquid scintillation counting using standard procedures at the NERC Radiocarbon Laboratory (RCL) (Harkness and Wilson 1972). Stable carbon isotope ratios were measured on benzene combusted to CO₂ using the dual-inlet mass spectrometer (VG OPTIMA) described above.

Climatic and soil physical measurements

Meteorological measurements such as air temperature, wind speed, solar radiation (used for modelling) etc. were made at towers used for micrometeorological measurements. Precipitation was collected in rain gauges in open fields near the tower. At Knottåsen the precipitation log was incomplete and measurements from the nearest meteorological station of the Swedish Hydrological and Meteorological Institute (Åmotsbruk) were used to fill in the gaps.

Soil temperature (all sites) and moisture (Knottåsen) were measured at one randomly chosen plot per moisture class. For soil temperature measurements 4 thermistors (Campbell Scientific, 107 Temperature Probe) were installed at 3 cm depth. The volumetric water content (soil moisture) was measured using TDR probes (Campbell Scientific, CS615 Water Content Reflectometer) installed vertically from the soil surface (0–30 cm). Two probes were installed adjacent to two randomly selected lysimeter positions. At Asa, the position of the groundwater level was registered manually bi-weekly at each plot.

Modelling of water flux and soil temperature

Vertical soil water fluxes and soil temperature gradients were calculated using the COUP model (Jansson and Karlberg 2001). The model simulates soil water and heat processes in different types of soils. The calculations of water and heat flows are based on soil properties such as the water retention curve, functions for unsaturated and saturated hydraulic conductivity, the heat capacity including the latent heat at thawing/melting and functions for the thermal conductivity. Air temperature, vapour pressure, wind speed, precipitation, global radiation and net radiation are driving variables in the model.

In this work the model was only used to calculate the vertical water flux and soil temperature at the dry and mesic plots, because lateral water transport was negligible at these plots (recharge areas at upper and mid-slope positions). The model was calibrated using average values for stand properties representing all the dry and mesic plots. Thus, water flux data and soil temperatures represent

site-specific rather than plot-specific conditions. Modelled and measured soil temperatures were in good agreement, typically within 1 °C. DOC fluxes in the soils at the dry and mesic plots were calculated by multiplying simulated water fluxes and average DOC concentrations per sampling period.

Statistical analysis

All measurements were grouped into three seasons: 'summer' (May–July), 'autumn' (August–November), and 'winter' (December–April). For O and B horizon leachates a statistical evaluation was made with a three-way ANOVA to test for differences between sites, moisture classes and seasons. A one-way ANOVA was used to test for differences between sites in throughfall.

Pearson correlation analysis was applied to test for relationships between DOC concentration and soil temperature and water flux, respectively. Statistical analysis was performed with SAS 8.01 statistical software (SAS Institute Inc).

Results and discussion

DOC concentration decrease with latitude

There were statistically significant differences in DOC concentration in the O horizon leachates between the three sites (Table 4). The southernmost site, Asa, had the highest concentration (average 49 mg l^{-1}), followed by the site in central Sweden, Knottåsen (average 39 mg l^{-1}). The lowest average DOC

Table 4. DOC concentration (mg l^{-1}) in bulk deposition, throughfall and leachates from the O and B horizon grouped by site, season and moisture class.

Grouped by		Asa	n	Knottåsen	n	Flakaliden	n
Site	Bulk deposition	2.5	1	4.8	1	5.1	1
	Throughfall	15.9 (0.4) a	3	7.7 (3.3) b	3	6.2 (0.7) b	3
	O horizon	49.3 (3.6) a	24	38.6 (3.3) b	18	30.3 (2.9) c	18
	B horizon	5.9 (0.9) a	24	7.0 (2.3) a	18	2.2 (0.2) b	18
		May-July		August-November		December-Aprila	
Season	O horizon	27.6 (1.8) a	26	52.0 (3.1) b	26	44.0 (3.4) b	8
	B horizon	4.4 (1.1) a	26	5.4 (1.4) a	26	6.4 (1.8) a	8
		Dry		Mesic		Moist	
Moisture class	O horizon	40.4 (4.0) a	21	37.4 (3.6) a	18	42.9 (3.8) a	21
	B horizon	3.1 (0.4) a	21	2.4 (0.2) a	18	9.5 (1.9) b	21

The evaluation for soil leachates was performed with a three-way ANOVA. For throughfall, only site effects were tested (n=3). Letters refer to significant differences within each group. Average and standard error are based on average concentration per plot and season.

^aData from Asa only.

concentration of the three sites was measured at the northernmost site, Flakaliden (average 30 mg l⁻¹). In comparison with other boreal and temperate forests, DOC concentrations in O horizon leachates are low at Knottåsen and Flakaliden and intermediate at Asa (Michalzik et al. 2001). Under the B horizon the average DOC concentrations were between 2 and 7 mg l⁻¹ (Table 4), which is low compared to measurements in other boreal and temperate forest soils (Michalzik et al. 2001). Flakaliden had a significantly lower DOC concentration in the B horizon, which is due to relatively low DOC concentration at the moist plots compared to the other sites.

The LUSTRA sites were chosen with the ambition of keeping parameters other than temperature (tree species, stand age, soil type, precipitation etc., see Table 1), as constant as possible. However, the temperature gradient coincides with a gradient in nitrogen status, as indicated by the C/N ratio of the O horizon and nitrogen deposition rates (Table 1). Both temperature and nitrogen status are higher in the south. We suggest that the different DOC concentrations in O horizon leachates at the three sites are related to differences in mean annual temperature, in combination with different nitrogen status.

Temperature might affect DOC concentrations in O horizon leachates in two ways, either directly via an effect on the microbially mediated processes governing the production of DOC, or indirectly affecting the amount of litter (substrate) produced and decomposed. Laboratory incubations have shown that the DOC production rate increases with increasing temperature (e.g. Christ and David 1996; Gödde et al. 1996; Andersson et al. 2000). However, the difference in average temperature between the sites during the vegetation period (average daily temperature >5 °C) is less than 1 °C, which with the temperature sensitivity for DOC production published in the literature (Christ and David 1996; Gödde et al. 1996), would give less than 10% difference in DOC concentration between the sites. The differences in DOC concentration can therefore not be attributed solely to a direct effect of temperature. Instead we suggest that a major link between mean annual temperature and DOC leaching from the O horizon is related to the effect of temperature on the net primary production of the ecosystem. A higher mean temperature means a longer growing season (Table 1), which in turn means a higher net primary production and a higher litter production rate. Since litter is the primary source of DOC-producing substrate, a higher input rate of litter – at least with time – should result in higher DOC production. For example, Park and Matzner (2003) showed that the addition of extra litter increased the leaching rates of DOC from the Oe and Oa horizons. At present, no litter fall data are available for the three sites. However, litter production is related to the aboveground tree biomass (Berg et al. 1999), which shows a clear south-north gradient and is well correlated with DOC concentrations (Figure 1a). Furthermore, the gradient in DOC concentration between the sites coincided with a south-north gradient in carbon stocks in the O horizon (Table 3). This carbon stock trend is a general phenomenon in this region according to a recent inventory of organic carbon pools in well-drained boreal forest soils in Scandinavia (Callesen et al. 2003)

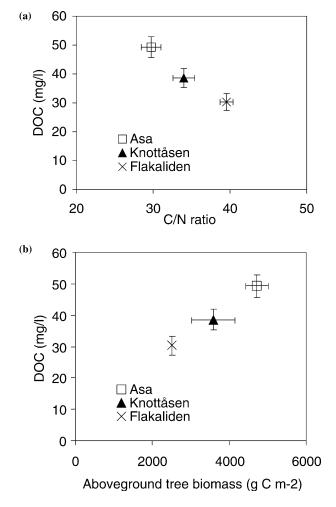


Figure 1. Average DOC concentrations in O horizon leachates (2000–2001) versus aboveground tree biomass (a) and C/N ratio of the O horizon (b). Error bars represent standard errors. Data for aboveground biomass from Berggren et al. (2004) (Asa, Knottåsen) and Strömgren (2001) (Flakaliden). Tree age at the time of measurements was 38 years at Asa and Knottåsen and 40 years at Flakaliden.

and the authors ascribed the decreasing carbon stocks from south to north to differences in net primary production. Not only the mean annual temperature, but also nitrogen status influences the net primary production at our sites. This is evident from numerous nitrogen fertilization experiments made in Swedish forests (e.g. Tamm 1991). The effects of temperature and nitrogen status cannot be separated with the data available and they both act in the same direction, i.e. favouring a high net primary production in the south. It is

important to note that all our sites have low nitrogen status compared to areas in central Europe subjected to high loads of nitrogen deposition (MacDonald et al. 2002). There is strong evidence in the literature that high nitrogen deposition rates hamper decomposition rates of soil organic matter (Nohrstedt et al. 1989; Magill and Aber 1998), and probably also DOC production (Gödde et al. 1996; Michel and Matzner 2002). However, the mechanism behind this observation is not vet known. Both chemical stabilisation of organic matter through nitrogen incorporation (Nömmik and Vahtras 1982), a suppressed lignin degradation (Berg and Matzner 1997) and a changed carbon use efficiency of the microorganisms (Agren et al. 2001) have been proposed as explanations. Thus, in nitrogen-rich ecosystems there might be a decoupling between net primary production and the DOC leaching rate from the O horizon. This was illustrated in a long-term nitrogen fertilisation experiment in Stråsan, central Sweden, where nitrogen applications increased tree growth considerably (Tamm 1991) but DOC concentration in O horizon leachates remained unaffected (Berggren et al. 1997). One possible explanation for the fact that we found a negative relationship between the DOC concentration in O horizon leachates and the C/N ratio in the O horizon (Figure 1b), whereas others have found a positive relationship (Aitkenhead and McDowell 2000) or no relationship (Michalzik et al. 2001), could be due to this dual effect of nitrogen, on one hand favouring net primary production in nitrogen-limited ecosystems and on the other reducing soil organic matter decomposition rates.

At present, there are relatively few published studies of DOC concentrations along a climatic gradient. However, the data in this study are in accordance with the results of Liechty et al. (1995), who found differences in DOC concentrations between two northern hardwood stands in the USA, with the highest DOC concentration at the warmest site. In a review, Michalzik et al. (2001) compiled data from 42 studies covering a wider range in mean annual temperature (1–16 °C) than in this study and found no significant correlation between temperature and DOC concentration or fluxes in O horizon leachates. As discussed above, a high mean annual temperature should favour high production of DOC, but sufficient precipitation is also needed to transport DOC out of the O horizon. Furthermore, high temperature combined with low or moderate precipitation may impose water stress on vegetation, limiting tree growth and litter production. In addition, nitrogen effects as discussed above and other environmental factors may influence DOC leaching. Accordingly, care should be taken when extrapolating the findings in this study to other environmental conditions.

Seasonal variations in DOC

Seasonal variations in DOC concentrations have been found in several studies, normally with a peak during summer—autumn (e.g. Michalzik and Matzner 1999; Yano et al. 2000; Solinger et al. 2001; Kaiser et al. 2002). In this study, a

seasonal effect was found at all three sites, with peaks in DOC concentrations in O horizon leachates in September-October (Figure 2). In spite of the seasonality, no simple correlation between air or soil temperature and DOC could be found (Table 5). Nevertheless, as illustrated in Figure 2, there seems to be a relationship between temperature and DOC, but with a lag phase of about 2 months. One possible explanation for this time lag is a seasonal variation in amounts of substrate available for DOC production. Fresh litter is known to produce significant amounts of DOC (Hongve 1999). However, long-term monthly measurements (1993-2001) of litter fall at 10 Norway spruce forest plots at Asa Experimental Forest (not LUSTRA plots) showed that there is generally only a moderate seasonal variation in litter fall rate, occasionally with peaks in both spring and autumn (unpublished data from Asa Experimental Forest). Furthermore, ¹⁴C measurements of DOC in Oa horizon leachates and water-extractable organic carbon of Oi, Oe and Oa horizon materials from Asa suggest that recent litter is not the main source of DOC leached from the O horizon (Fröberg et al. 2003). Therefore we suggest that the lag between soil temperature and DOC concentration (Figure 2) is caused by a buffering of DOC in the O horizon by sorption-desorption processes. Like respiration, production of DOC is thought to be a mainly microbially controlled process. Laboratory incubations indicate that DOC production shows similar temperature dependence as carbon mineralization (Christ and David 1996; Gödde et al. 1996). It is therefore reasonable to assume that production of DOC follows the same seasonal pattern as heterotrophic respiration, which seems to be directly related to temperature (Högberg et al. 2001). What is measured in the lysimeters is not the production of DOC, but what is being leached out of the O horizon, which is the net effect of production, mineralization and sorption-desorption processes. It is known that sorption of DOC to organic horizons occurs (Qualls 2000) and solubility of organic carbon has been modelled as a partitioning between solid and solution organic phases by Tipping and Woof (1991). The delay in DOC concentrations could accordingly be explained by sorption or partitioning mechanisms in the O horizon, since the pool of 'sorbed DOC' needs to be built up before any major increase of DOC in the soil solution occurs (Michelzik et al. 2003). In a similar way, when the net production of DOC decreases with deceasing temperature, the large pool of 'sorbed DOC' delays the decrease of DOC in the soil solution.

DOC in relation to soil moisture

In contrast to temperature, variations in soil moisture were not found to have any major influence on DOC leaching. There were no significant differences in DOC concentrations between the moisture classes in O horizon leachates (Figure 3, Table 4). TDR measurements made at Knottåsen showed that there were marked differences in soil moisture content between the moisture classes throughout the year (Figure 4). Measurements of the groundwater level at Asa

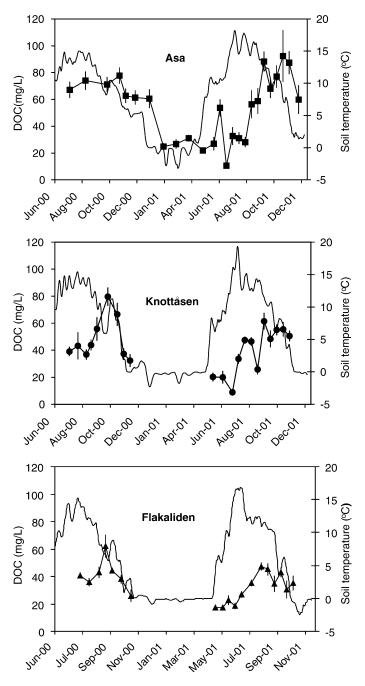


Figure 2. DOC concentrations in soil leachates from the O horizon and modelled soil temperature at 5 cm depth. Points represent average values for all moisture classes (n=8 at Asa, n=9 at Knottåsen and Flakaliden) and bars represent standard error of the mean.

Table 5. Correlation (r-values) between DOC concentrations in the O horizon on different occasions and modelled average temperature and water flux during the measuring period.

		Temperature	Water flux
Asa	May–July	0.07	0.92*
	August-November	-0.69*	0.55
	December-April	0.65	0.80
Knottåsen	May–July	0.46	0.86*
	August-November	-0.14	0.37
Flakaliden	May–July	0.61	0.05
	August-November	0.27	-0.27

^{*}Indicates significant correlation (p < 0.05).

also indicated a clear difference between the moist plots on one hand and the mesic and dry plots on the other. During 2001, the position of the groundwater level was at -70 cm, or deeper at the dry and mesic sites throughout the year. At the moist plots it fluctuated between -40 and -15 cm, except during the summer period (June–August) where it occasionally dropped to > -200 cm. Unfortunately, no measurements of groundwater levels or soil moisture were available for Flakaliden, but the differences in slope position (toeslope, mid slope, or upper slope), vegetation composition and soil organic carbon pools (Table 3) strongly suggest that differences in soil moisture/hydrology between moisture classes also exist at this site. The lacking relationship between moisture and DOC at Knottåsen indicates that soil moisture *per se* plays a minor role in controlling the DOC concentration in O horizon leachates. For example soil moisture was high during both spring and autumn, whereas DOC concentration was high only during autumn (Figures 4a, b).

There were no differences in either concentration or seasonal pattern of DOC in O horizon leachates between the moist plots and the dry and mesic plots (Figures 2 and 4), despite larger carbon pools in the O horizon at the former plots (Table 3). Thus, the amount of carbon in the O horizon is not the main determinant of the DOC concentration in the hydrological gradient. The litter input appears to be the same for all moisture classes, since measurements of the tree biomass and other measurements of the site quality are the same (Berggren et al. 2004). This suggests that the larger pool size of organic carbon at the moist plots is probably the result of a lower decomposition rate of soil organic matter at these plots. At Asa, for example, as indicated above, the position of the groundwater at the moist plots was periodically close to the bottom of the O horizon during 2001, suggesting that soil organic matter decomposition is hampered in the mineral soil as well as in the lower part of the O horizon during part of the year. However, there was no relationship between the position of the groundwater and the DOC concentration in the O horizon leachates. Instead DOC concentrations were the same as those measured at the well-drained plots (Figure 2), indicating that other factors – probably soil temperature – control the seasonal dynamics of DOC in

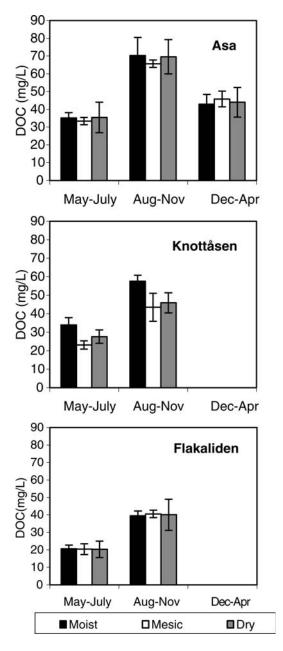


Figure 3. Average DOC concentrations in leachates from the O horizon at moist, mesic and dry plots at Asa, Knottåsen and Flakaliden (n=2 for Asa mesic, n=3 for other moisture classes and sites). Error bars represent standard error of the mean.

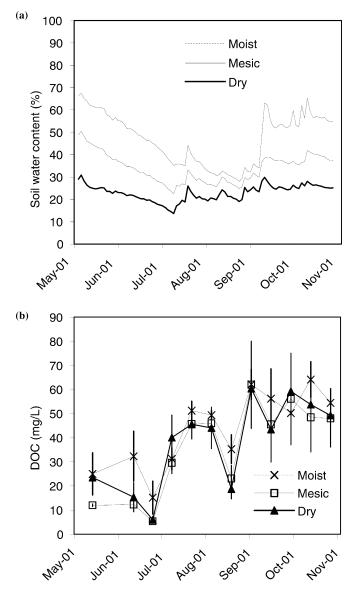


Figure 4. (a) Soil water content measured with TDR at moist, mesic and dry sites at Knottåsen (0-30 cm). Average values for 2 probes per moisture class. (b) Average DOC concentration $(\pm \text{SE})$ in O horizon leachates at moist, mesic and dry plots at the same site (n=3).

O horizon leachates. We propose that the DOC production and transport are related to the turnover of SOM in the upper part of the O horizon, which is not severely influenced by a high groundwater level. The lower part of the

O horizon – periodically subjected to oxygen-deficient conditions – probably does not contribute substantially to the DOC leaving the O horizon.

Under the B horizon, the concentration of DOC was higher at the moist plot than at the mesic and dry plots (Table 4), which can be explained by the higher carbon content of the mineral soil at the moist plots (Table 3). Previous studies have shown that the ability of mineral soil to sorb DOC decreases with increasing carbon content (Vance and David 1992; Kaiser and Guggenberger 2003). In addition, reducing conditions may decrease DOC sorption (Hagedorn et al. 2000), which could contribute to the greater DOC concentrations under the B horizon at the moist plots.

DOC in relation to water fluxes

The water flux (mm per day during the sampling period) at 5 cm depth was found to have no or only a small effect on DOC concentrations in the O horizon leachates (Table 5). For the summer period, positive correlations between DOC concentration and water fluxes were found at Asa and Knottåsen, whereas no such correlations were found at Flakaliden and for the autumn (winter) period at Asa and Knottåsen. This can be compared with other studies where negative (McDowell and Wood 1984) or no (Michalzik and Matzner 1999) correlations have been found between water flux and DOC concentration in the forest floor. The positive correlation during summer at Asa and Knottåsen, may have been caused by rewetting of dried organic matter, resulting in high DOC concentrations (Lindquist et al. 1999; Prechtel et al. 2000; Schaumann et al. 2000). Overall, the correlation between water flux and DOC concentration was poor though with no dilution effect at any of the sites. This is in agreement with a controlled water flux manipulation experiment by Park and Matzner (2003), who noticed that doubled water fluxes did not change DOC concentrations under the O horizon. It is also consistent with the idea of sorption-desorption processes and a relatively large pool of 'sorbed DOC', which controls the short-term dynamics of DOC and results in a significant buffering of the DOC concentration.

In the B horizons the concentration of DOC was relatively constant throughout the year, showing no tendency for increased concentrations at high water fluxes, indicating that sorption controls DOC concentrations in this horizon.

DOC fluxes and carbon pools

Annual fluxes of DOC to and within the ecosystems at the dry and mesic plots at Asa, Knottåsen and Flakaliden are shown in Table 6. At all sites, annual DOC fluxes followed the general pattern known for temperate and boreal forest ecosystems, with low to moderate fluxes in bulk deposition and

Table 6. Annual DOC fluxes (g m⁻²) in precipitation, throughfall, O horizon and B horizon leachates at dry and mesic plots during 2001.

	Asa	Knottåsen	Flakaliden
Precipitation	1.6	2.2	3.2
Throughfall	7.1	3.3	2.8
O horizon	26.3	10.7	13.4
B horizon	1.3	0.4	0.9

DOC fluxes with precipitation and throughfall are calculated from measured volumes and concentrations. Values for O and B horizons are based on modelled water flux and measured concentrations.

throughfall, high fluxes in O horizon leachates and, again, low fluxes in leachates collected from the B horizon (summarized by Michalzik et al. 2001). The same pattern is also shown for the concentrations reported in Table 4. Fluxes at the three sites were within the range reported by Michalzik et al. (2001), although in the lower part of the spectrum. Like DOC concentrations, DOC fluxes were in general highest at Asa, whereas the fluxes of DOC at Flakaliden and Knottåsen were quite similar. The DOC flux below the O and B horizon at Flakaliden was probably relatively high in 2001 due to excessive precipitation, 60% higher than normal (long-term average 1961-1990). Precipitation was close to normal at the other two sites. The DOC flux out of the O horizon was in general highest during the autumn (Figure 5), i.e. the period with both the highest DOC concentrations (Figure 2) and with relatively high water flux. The sorption efficiency of DOC between the O and the bottom of the B horizon was high at all sites, retaining 94–96% of the amount leaving the O horizon (Table 6). This sorption is high compared to soils in many other temperate forests (Michalzik et al. 2001). Soils at the LUSTRA sites are poorly aggregated, which suggests that preferential flow is of minor importance and that there is a good contact between soil particle surfaces and the percolating solution. Furthermore, the B horizons of Swedish Podzols are generally rich in inorganic iron (goethite and ferrihydrite) and aluminium (imogolite-type minerals) precipitates (Karltun et al. 2000), which have reactive surfaces. The total amount of carbon sorbed in the B horizon was between 11 and $25 \text{ g m}^{-2} \text{ yr}^{-1}$, which corresponds to 0.2-0.3% of the carbon stocks in the mineral soil down to 50 cm depth for dry and mesic plots (Table 3).

Fate of DOC leached from the O horizon

The ¹⁴C measurements from the mesic plots at Asa and Knottåsen show that there were large differences (13–18% units) in ¹⁴C activity between the Oe and Oa horizons on one hand and the B horizon on the other. This was true for both the solid soil samples and for DOC (Figure 6). At the same time, the differences in ¹⁴C content between solid soil organic matter in the B horizon

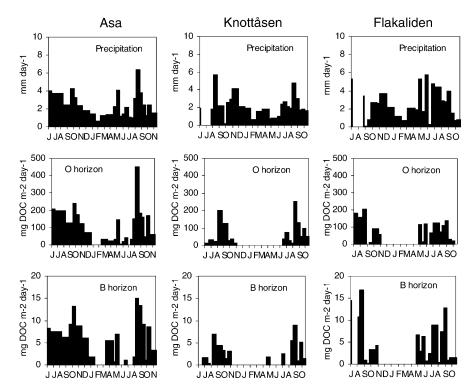


Figure 5. Precipitation amounts and DOC fluxes per sampling period under the O and B horizons at the three different sites. Fluxes of DOC represent the average fluxes at dry and mesic plots.

and DOC obtained from the same horizon were much smaller (<5% units difference). This suggests that the DOC in the B horizon is derived mainly from the soil organic matter in the B horizon itself, i.e. there is a substantial exchange between the pool of solid old carbon and the incoming 'fresh' DOC.

The differences in ¹⁴C between the O and B horizons in this study contrast with a study by Trumbore et al. (1992). They reported DO¹⁴C values in O, Ah, B1, B2 and B3 horizons of a Spodosol in Ontario, Canada, where the radiocarbon values in all horizons were above 100% modern, with no consistent difference between the horizons. This could indicate a substantial preferential flow in that soil. Zero-tension lysimeters were also used for mineral soil horizons in their study, making collection of preferential water possible. The ¹⁴C-signal pattern found in the soils at Asa and Knottåsen was, however, similar to that found in Podzols at Birkenes, Norway (Michalzik et al. 2003). The high degree of sorption and desorption rates of DOC in the mineral soil indicated by the ¹⁴C-data in this study implies that characteristics of the mineral soil (content of inorganic Fe and Al precipitates, texture, amounts of carbon already present in the soil etc.) rather than the incoming DOC

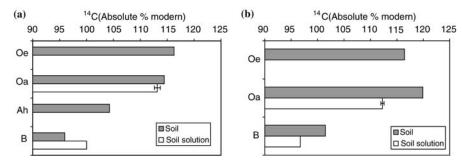


Figure 6. 14 C activity in soil solution and solid soil in different horizons in (a) Asa and (b) Knottåsen. Error bars represent standard error (n=3) based on variations between samples.

concentrations from the organic horizon are controlling the DOC concentration in the mineral soil. Thus DOC properties at the bottom of the B horizon and DOC concentration should be regarded more as a function of the soil horizon in which they are measured, rather than as a function of the horizons above. This also means that external factors such as climate or forest management have only minor effects on concentrations and properties of DOC in the subsoil.

Conclusions

The data from this study indicate that concentrations and fluxes of DOC in O horizon leachates are higher in southern than in northern Sweden. We suggest that this gradient is mainly caused by differences in net primary production, which arise from a combined effect of a gradient in annual mean temperature and nitrogen status, both of which are highest in the south. The higher net primary production in the south results in more litter, i.e. substrate for DOC. It should be noted though, that our sites were low-to-moderately exposed to nitrogen deposition and the positive relationship between net primary production and DOC leaching from the O horizon can probably not be extrapolated to ecosystems subjected to high nitrogen loadings.

Soil temperature probably governs the within-year variation in DOC concentration in O horizon leachates, but there is a time lag of about two months between the peaks in temperature and DOC concentration; the peaks in temperature occur in July and those in DOC concentration in September. We hypothesize that this time lag is due to sorption processes in the O horizon, i.e. the build-up and depletion of a sorbed pool of potential DOC. The general lack of correlation between water flux and DOC concentration in O horizon leachates can also be ascribed to the buffering effect of sorption–desorption processes in the O horizon material. Soil moisture regime – or seasonal variation in soil moisture – does not seem to be important for the concentration of DOC leached from the O horizon.

The ¹⁴C data showed that DOC collected in the B horizon was not leached directly from the O horizon. Instead it must have been derived from organic carbon stored in the B horizon itself. In contrast to the O horizon, climate effects were of minor importance for DOC leaching from the subsoil. Instead, the properties of the mineral soil governed the DOC leaching out of the B horizon.

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