Different effects of peat degradation on dissolved organic carbon and nitrogen

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Abstract

The dynamics of dissolved organic matter (DOM) are closely related to organic matter processes such as decomposition, humification, and stabilization of organic matter in soils. Nevertheless, it is unknown whether dissolved organic carbon (DOC) and nitrogen (DON) have an equal response to these processes. We analyzed DOC and DON concentrations of topsoil, groundwater, and surface water of six differently used sites characterized by differences in peat degradation. Carbon (^{13}C, ^{14}C) and nitrogen (^{15}N) isotopes of DOM were related to a humification index in order to detect differences between DOC and DON at different stages of humification. The results show that intact peatlands had higher DOC concentrations than degraded peatlands. Clearly, DOC concentration depends on soil organic carbon content and therefore on the degree of peat degradation. However, intact and degraded peatlands showed similar DON and inorganic N concentrations suggesting that DON release is dependent on soil inorganic N rather than the organic N pool. Moreover, a high degree of peat degradation resulted in lower DOC/DON ratios than the C/N ratio of the solid phase indicating a preferential release of DON from soil organic matter. Further, δ^{13}C ratios and the radiocarbon age of DOM increase with peat degradation and humification indicating a high C turnover, an increased microbial modification and age of DOC. On the other hand δ^{15}N ratios decrease, probably as a result of N fertilization. The promoting effect of inorganic N on DON release and a high humification of DOM at sites treated with inorganic N fertilizers suggest that N fertilization promotes a release of amino acids depleted in ^{15}N and subsequent condensation with carbohydrates to humic substances. © 2002 Elsevier Science Ltd. All rights reserved.

1. Introduction

Dissolved organic matter (DOM) in soils plays an important role in the biogeochemistry of carbon, nitrogen, and phosphorus, as summarized by Kalbitz et al. (2000a). It affects the pedogenesis, the mobilization, and transport of pollutants in soils (Dawson et al., 1978; Kalbitz et al., 1997; Temminghoff et al., 1997; Raulund-Rasmussen et al., 1998; Marschner, 1999). DOM is generally regarded as a continuum of organic molecules of various sizes and structure. Only small proportions of DOM have been identified (Thurman, 1985; Herbert and Bertsch, 1995).

Although the pool of DOM in soils is only 0.04–0.22% of soil organic carbon (Zsolnay, 1996), it represents the most mobile part of it. Therefore, DOM might be a useful indicator for processes affecting the C and N cycles. Numerous studies have also shown effects of environmental factors and management activities on DOM (Kalbitz et al., 2000a).

Usually, DOM is quantified by the analysis of dissolved organic carbon (DOC) but little is known about the N component of DOM, the dissolved organic nitrogen (DON). It is often assumed that DOC and DON have similar behavior in soils and aquatic ecosystems. However, in field and laboratory studies of the dynamics

of DOM, there are indications of the decoupling of the C and N cycle by environmental changes (Kalbitz et al., 2000a). For instance, Prechtel et al. (2000) showed a distinct increase of DON release after drought, whereas DOC was hardly affected. Furthermore, Williams and Silcock (2000) reported a higher seasonal variability of DON compared with DOC. Causes and mechanisms for these different behaviors of DOC and DON are largely unknown.

Decomposition, humification, and stabilization of SOM are closely related to DOM dynamics (Zech et al., 1997). The main transformations occurring during litter decomposition and humification are loss of polysaccharides and phenolic moieties, modification of lignin structures, and enrichment in recalcitrant, non-lignin aromatic structures (Zech et al., 1997). However, effects of an increased decomposition and humification of litter and SOM on DOM have been examined little so far. There was no clear effect of decomposition of different deciduous foliage on the dynamic of DOC and DON release over a period of 15 weeks (Magill and Aber, 2000).

In previous studies we observed that long-term intensive land use (50 to $\geqslant$ 200 years) of peat soils resulted in enhanced peat decomposition and in increasing amounts of aromatic structures and polycondensation of dissolved organic molecules (Kalbitz et al., 1999). Isotopic ratios of water-soluble fulvic acids (FA) were also affected by differences in peat decomposition due to land use (Kalbitz et al., 2000b). Therefore, land use impacts altered the soil conditions for DOM release, i.e. a changed amount and composition of soil organic matter. These studies supported the concept that DOM can serve as an indicator for processes in soils affecting the C and N cycle like decomposition and humification. However, it is unknown whether DOC and DON have an equal response to these processes. The known degrees of humification of DOM within an area characterized by similar environmental conditions offers the unique possibility to study the different effects of humification on DOC and DON.

The main objective of this study was to test the hypothesis that a change in the degree of humification of DOM caused by a different peat degradation has different effects on DOC and DON. We tested this hypothesis by (1) analyzing DOC and DON concentrations in soil and water and by (2) relating the degree of humification of DOM to the natural variation of carbon (^{13}C, ^{14}C) and nitrogen (^{15}N) isotopes of DOM.

2. Materials and methods

2.1. Study area and sites

Samples were taken at the "Droemling" area located in Northeast Germany (28,000 ha, 55–58 m above sea level, 570 mm mean annual precipitation, 8.4 °C mean annual temperature). The sampling sites have been described in detail by Kalbitz et al. (1999, 2000b). Six sampling sites with different land use were selected (Table 1). The soils of the degraded peatlands (sites 1–3) contain only relict amounts of peat as a result of agricultural use for the last 50–200 years. These sites have got considerable amounts of mineral fertilizers, up to 150–200 kg mineral N ha^{-1} year^{-1} (Kalbitz et al., 2000b). The land use pattern of site 3 was changed into unimproved grassland at the beginning of this study in 1996. At the relatively intact peatlands (sites 4–6) peat degradation was much lower than at the degraded peatlands as seen by higher content of organic matter (Table 1).

2.2. Sampling, analyzing, and statistics

Details of sampling and analysis are described by Kalbitz et al. (1999). Briefly, topsoils (0–25 cm depth), groundwater and surface water were sampled from spring 1996 to fall 1998 every 3 weeks. Aqueous saturation extracts of the topsoil were prepared with a relatively narrow soil-to-water ratio (soil/water: 0.3–1.1; w/v) similar to in situ soil solution. Groundwater samples were collected from wells (1–2 m depth) and surface water was directly pumped from the adjacent draining ditch into a container.

All aqueous samples were 0.45 μm filtrated (polysulfon, Fa. Gelman), then analyzed for DOC on a Shimadzu TOC 5050, for total-N by a modified Kjeldahl procedure (VDLUFA, 1991), for nitrate and nitrite by ion chromatography (Dionex DX 100), and for ammonium by colorimetrical analysis using sodium dichloroisocyanurate and sodium salicylate by Spectroquant 118. DON concentration was calculated as the difference between total-N and inorganic-N. Before running the ANOVA procedure (SPSS for Windows 8.0) and multiple comparison tests (Tukey test, $\alpha < 0.05$), data were pooled in two distinct groups, degraded peatlands and intact peatlands, in order to examine the effects of a different degree of peat degradation on DOC and DON. The homogeneity of variance was tested by the Bartlett test.

The degree of humification of DOM was related to the natural variation of carbon (^{13}C, ^{14}C) and nitrogen (^{15}N) isotopes of DOM as follows: for isotopic analysis we extracted fulvic acids (FA) from aqueous extracts of the topsoil, from groundwater and surface water by adsorption chromatography on XAD-8 resin at three sampling dates. Details on the isotopic measurements are given by Kalbitz et al. (2000b) who presented these data in detail. Briefly, $\delta^{13}C$ and $\delta^{15}N$ of the freeze-dried FA were determined using a C/N analyser (Carlo Erba 1108) coupled to a continuous flow isotope ratio mass spectrometry (delta C Finnigan MAT). ^{14}C measurements of the freeze-dried FA were made by accelerator

mass spectrometry (IsoTrace laboratory, University of Toronto) (Beukens, 1992). The ^{14}C concentration is given in pmC (% modern C) and referred to 94.9% of the activity concentration of the NBS oxalic acid standard in 1950. The results are corrected for natural and sputtering fractionation to a base of $\delta^{13}C = -25‰$ (Stuiver and Polach, 1977). In addition to the FA isotopes we recorded synchronous fluorescence spectra of the corresponding water sample to compute a humification index by calculating the ratio of fluorescence intensities at a longer (390 nm) and a shorter (355 nm) wavelength (Kalbitz et al., 2000c). The calculation of this humification index is based on the fact that a shift in the fluorescence intensity from shorter to longer wavelengths is associated with an increasing number of highly substituted aromatic nuclei (Senesi et al., 1989; Miano and Senesi, 1992). After the analysis linear regressions of isotopic data and humification indices

were calculated. Total organic carbon (TOC) and nitrogen (TON) content of the soil were analyzed on an elemental analyzer (LECO CHN-1000).

3. Results and discussion

3.1. DOC concentrations

Dissolved organic carbon (DOC) concentrations in topsoil, groundwater and surface water were significantly higher at the intact peatlands (46–87 mg C l^{-1}) than at the degraded peatlands (25–35 mg C l^{-1}, Table 2). These findings emphasize the importance of soil organic matter (SOM) as the main source of DOC (McDowell and Likens, 1988; Zsolnay, 1996). Clearly, DOC release depends on soil organic matter content and therefore on peat degradation.

Table 1
Land use, soil type and some properties of topsoil horizons of the study area ("Droemling", Germany)

Site	Land use	Soil depth (cm)	TOC[a] (g kg^{-1})	TON[b] (g kg^{-1})	TOC/TON	DON[c] (mg N l^{-1})
Degraded peatlands—Mollic Gleysols						
Site 1	Intensive arable land	0–18	41.8	3.60	11.6	6.8 (0.9)
Site 2	Intensive grassland	0–32	47.2	3.65	12.9	8.1 (1.6)
Site 3	Changing land use[d]	0–26	46.2	2.58	17.9	8.5 (1.1)
Relatively intact peatlands—Terric Histosols						
Site 4	Unimproved grassland	0–37	205.6	11.80	17.4	12.0 (1.4)
Site 5	Grassland in natural succession	0–28	86.8	8.76	9.9	6.4 (1.4)
Site 6	Almost natural woodland	0–30	452.7	30.70	14.7	6.8 (0.6)

[a] Total organic carbon content of the soil.
[b] Total organic nitrogen content of the soil.
[c] DON concentration in aqueous extracts of the topsoil (mean and standard error of 41 sampling dates from May 1996 to December 1998).
[d] Changing land use from intensive crop production to unimproved grassland in Spring 1996.

Table 2
DOC concentrations (mg C l^{-1}) and DOC/DON ratios of topsoil (aqueous extracts), ground and surface water (mean of 41 sampling dates)

Site	DOC (mg C l^{-1})			DOC/DON		
	Topsoil	Groundwater	Surface water	Topsoil	Groundwater	Surface water
Degraded peatlands[a]	34.7±0.8	24.9±0.6	29.4±2.8	9.5±1.2	9.4±1.1	12.3±2.1
Site 1	33.1	28.8	10.5	7.7	8.1	7.1
Site 2	38.6	19.8	61.8	12.5	12.7	23.2
Site 3	32.4	26.1	26.5	7.9	8.0	6.5
Relatively intact peatlands[a]	87.1±4.5	46.0±3.7	52.6±3.2	16.2±1.5	17.5±2.4	14.6±1.6
Site 4	57.8	24.9	21.7	8.8	5.6	5.3
Site 5	59.5	37.1	75.1	15.5	23.4	21.1
Site 6	144.1	76.0	72.8	25.9	29.7	23.6

[a] Mean and standard error of the three sites.

3.2. DON concentrations and DOC/DON ratios

While total organic nitrogen (TON) concentrations of topsoils were much higher in intact peatlands (9–31 g kg^{-1} soil) relative to the degraded peatlands (3–4 g kg^{-1} soil) dissolved organic nitrogen (DON) concentrations did not show any significant difference (6–12 mg l^{-1}, Table 1). Furthermore, organic and inorganic N of aqueous extracts from topsoils were in the same concentration range (5–12 mg l^{-1}) as reported by Murphy et al. (2000) for agricultural soils. Moreover, we found similar concentrations of inorganic N in the same samples for degraded ($\sim$5.6 mg l^{-1}, $n = 109$) versus intact peatlands ($\sim$5.1 mg l^{-1}, $n = 108$). Therefore, DON release in the topsoil seems to be affected by the smaller pool of inorganic N rather than by the large pool of soil organic N.

The promoting effect of soil inorganic N on the release of DON becomes also evident looking at C/N ratios of the solid and the liquid phase. These ratios showed that degraded peatlands topsoil are depleted in C versus N in the water phase (Fig. 1). Thus, our results are explained by a preferential release of DON from the solid phase of soil organic matter in degraded peatlands possibly due to the input of 150–200 kg mineral N ha^{-1} year^{-1}. Neff et al. (2000) reported that N fertilization increased DON fluxes by 50% and decreased the DOC/DON ratio in a soil sample with a low nitrogen content. Also Williams et al. (1999) found that the amount of DON in water extracted from moss increased linearly with the amount of inorganic N added. However, this added inorganic nitrogen was not directly incorporated in DON as indicated by an applied ^{15}N tracer (Williams et al., 1999). Furthermore, in forest soils, DON concentrations and fluxes increased after nitrogen addition, whereas DOC concentrations and fluxes remained unchanged (Currie et al., 1996; McDowell et al., 1998). It is, therefore, suggested that DON release is promoted when inorganic nitrogen is added to the soil.

3.3. Carbon and nitrogen isotopes in relation to DOM humification

3.3.1. δ^{13}C ratios in relation to DOM humification

An enhanced humification of DOM could be related to an enrichment of ^{13}C in DOM as shown in Fig. 2. Melillo et al. (1989) and Bol et al. (1999) have shown a ^{13}C enrichment with increased decomposition of organic matter which could be attributed to enhanced microbial contribution of ^{13}C enriched (re)synthesized compounds (Huang et al., 1996, 1998). In a previous study, Kalbitz et al. (2000b) showed that strong peat degradation resulted in a ^{13}C enrichment as indicated by higher values at the degraded peatlands compared with the intact peatlands. Haider (1999) emphasizes the outstanding role of microbes for decomposition and humification of plant residues. Therefore, we conclude that

an increasing degree of humification of DOM is related to an increasing microbial modification of water-soluble organic carbon compounds.

The positive relationship between humification of DOM and its δ^{13}C ratios supports the findings that the relative concentration of ^{13}C depleted lignin-derived aromatics are decreasing whereas the portion of ^{13}C enriched O-alkyl compounds remains more or less constant during humification (Haider, 1999; Schulten and Gleixner, 1999; Kracht and Gleixner, 2000). The positive relationship between humification of DOM and its δ^{13}C ratio also indicates an increasing formation of microbial biomass during humification, which is enriched in ^{13}C (Kracht and Gleixner, 2000).

3.3.2. ^{14}C contents in relation to DOM humification

The significant correlation between the ^{14}C content and the humification index of DOM in Fig. 3 shows that the humification degree of DOM increases with its age. As reported for δ^{13}C ratios strong peat degradation resulted in an increasing radiocarbon age (decreasing ^{14}C values) of DOC. This is indicated by higher values at the degraded peatlands compared with the intact peatlands (Kalbitz et al., 2000b). Bol et al. (1999) concluded that decomposition of SOM is linked to an increased radiocarbon age. This conclusion can be extended to humification of DOM.

The significant increase of the ^{14}C content after a short-term land use change (Fig. 3) is not linked with decreasing humification indices as expected from the relationship presented above (Fig. 3). The input of newly synthesized organic carbon with modern ^{14}C values after this land use change reflects a changed C turnover probably by an increased microbial activity. On the other hand, these newly synthesized organic compounds should be characterized by a similar humification. The positive relationship between humification and age of DOM seems to exists after this land use change with a constant slope but shifted to a different level.

3.3.3. δ^{15}N ratios in relation to DOM humification

In contrast to the δ^{13}C ratios and ^{14}C values, the high humification index of the degraded peatlands is linked to low δ^{15}N ratios as shown in Fig. 4. That means, in opposite to DOC, an increased humification of DOM did not result in an increased microbial change of DON. An increased humification of DOM is rather linked to the incorporation of isotopically light N into DON. Furthermore, strong peat degradation resulted in a ^{15}N depletion as indicated by lower values at the degraded peatlands compared with the intact peatlands (Kalbitz et al., 2000b). These low δ^{15}N ratios support the hypothesis of a low microbial modification of DON because microbes prefer light N (Desjardins et al., 1996; Martinelli et al., 1996; Högberg, 1997; Novák et al.,
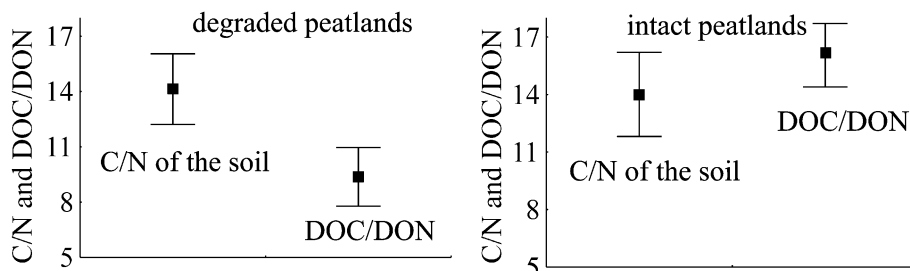
Fig. 1. C/N ratio of the solid and the liquid phase (DOC/DON) of the degraded and the intact peatlands (mean and standard error of 3 sites; solid phase: one sampling date per site of the topsoil horizon; liquid phase: mean and standard error include data of 41 sampling dates for each site).
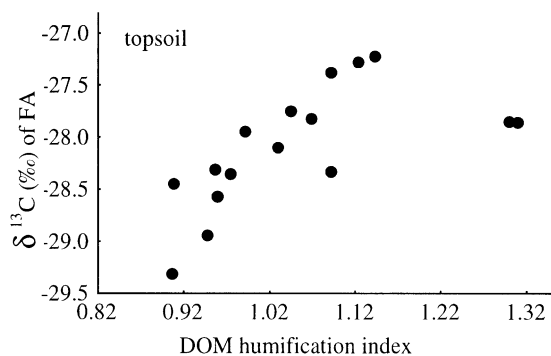

Fig. 2. Dependence of δ^{13}C ratios of water-soluble fulvic acids on DOM humification index (ratio of fluorescence intensities at 390/355 nm; $r = 0.63$).
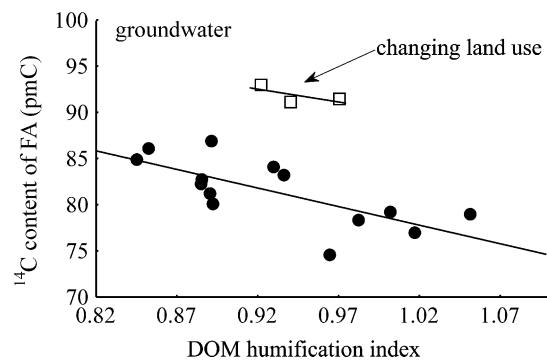

Fig. 4. Dependence of δ^{15}N ratios of water-soluble fulvic acids on DOM humification index (ratio of fluorescence intensities at 390/355 nm; $r = -0.80$). The δ^{15}N ratios of site 3 were not included in these regressions because of changed isotopic values and unchanged humification indices after the short-term land use change at this site.

Fig. 3. Dependence of ^{14}C contents of water-soluble fulvic acids on DOM humification index (ratio of fluorescence intensities at 390/355 nm; $r = -0.72$). The ^{14}C contents of site 3 were not included in these regressions because of changed isotopic values and unchanged humification indices after the short-term land use change at this site.

Fig. 5. C/N ratio of water-soluble fulvic acids (topsoil, groundwater, surface water) in dependence on DOM humification index (ratio of fluorescence intensities at 390/355 nm; $r = -0.50$).

1999). An enrichment of ^{13}C and a depletion of ^{15}N could be explained by a different response of DOC and DON to peat degradation and DOM humification.

Besides, an increased humification of DOM is also related to a selective preservation of nitrogen as indicated by a linear correlation (Fig. 5) using the published
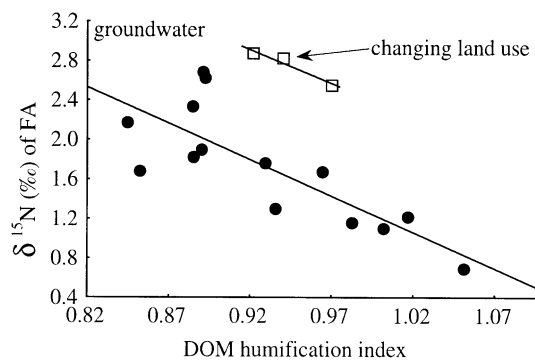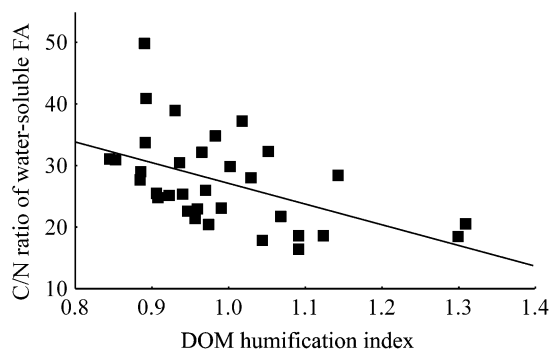
C/N ratios of water-soluble FA (Kalbitz et al., 1999) and the humification indices of DOM (Kalbitz et al., 2000c).

The isotopically light N of FA from the degraded peatlands might derive from the added mineral fertili-

zers. According to Clark and Fritz (1997), the $\delta^{15}N$ ratios of mineral fertilizers are often lower than soil organic N. However, a direct incorporation of this fertilizer N into DON is not likely because in ^{15}N tracer experiments in a bog peat and in a grassland soil less than 0.1% of added inorganic N could be detected in DON (Williams et al., 1999; Shand et al., 2000). A promoting influence of the inorganic N pool on DON release is more likely (see above). At the moment we can only speculate about mechanisms for this promoting effect of the inorganic N pool. Qualls (2000) emphasized that microbial dissolution of organic compounds like proteins is important for the production of DON. This microbial dissolution could be promoted by an addition of inorganic N (see Section 3.2.). Then, the Maillard reaction in which amino acids (released during the hydrolysis of proteins) and carbohydrates condense to form humic substances, could be responsible for increasing DON release (Qualls, 2000). This pathway of humic substances formation is supported by higher humification indices at sites with an addition of inorganic N (degraded peatlands). Higher contents of amino acids in soils treated with inorganic N than in untreated soils also support this hypothesis (Bol et al., 1998a). However, these amino acids vary in the $\delta^{15}N$ ratios in a broad range (-12 to $+26‰$) (Bol et al., 1998b). The $\delta^{15}N$ ratios of bulked amino acids can be higher or lower than the $\delta^{15}N$ of the soil (Bol et al., 1998b; Ostle et al., 1999). A determination of individual amino acids (content, $\delta^{15}N$ ratio) of degraded and intact peatlands would allow to test our hypothesis of a promoting influence of added inorganic N on the release of amino acids.

Similar to the ^{14}C contents increasing $\delta^{15}N$ ratios after a changing land use (Fig. 4) indicated that an increased microbial activity do not result in an increased humification of DOM. However, the increasing $\delta^{15}N$ ratios mean that the released DON after this land use change was strongly microbial altered (Högberg, 1997; Novák et al., 1999). On the other hand, the ^{14}C values show a release of newly synthesized organic carbon. Also in this case DOC and DON show a different pattern.

4. Conclusions

DOC release depends on soil organic matter content and therefore on peat degradation whereas DON release is independent of it. The inorganic N pool affects the release of DON rather than the pool of soil organic nitrogen. Therefore, degraded peatlands characterized by a high degree of peat decomposition and an addition of inorganic N fertilizer release DON from SOM preferentially. High humification of DOM is linked with a high C turnover and high microbial modification and age of DOC but with low $\delta^{15}N$ ratios of DOM. It is

suggested that an addition of mineral N fertilizer increases the microbial mobilization of amino acids followed by a condensation with carbohydrates to humic substances. The different response of DOC and DON to enhanced peat degradation and DOM humification emphasizes the necessity to consider DON besides DOC in further DOM research.

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