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SOILS ISSUES

CONTROLS ON THE DYNAMICS OF DISSOLVED ORGANIC MATTER IN SOILS: A REVIEW

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Dissolved organic matter (DOM) in soils plays an important role in the biogeochemistry of carbon, nitrogen, and phosphorus, in pedogenesis, and in the transport of pollutants in soils. The aim of this review is to summarize the recent literature about controls on DOM concentrations and fluxes in soils. We focus on comparing results between laboratory and field investigations and on the differences between the dynamics of dissolved organic carbon (DOC), nitrogen (DON), and phosphorus (DOP).

Both laboratory and field studies show that litter and humus are the most important DOM sources in soils. However, it is impossible to quantify the individual contributions of each of these sources to DOM release. In addition, it is not clear how changes in the pool sizes of litter or humus may affect DOM release. High microbial activity, high fungal abundance, and any conditions that enhance mineralization all promote high DOM concentrations. However, under field conditions, hydrologic variability in soil horizons with high carbon contents may be more important than biotic controls. In subsoil horizons with low carbon contents, DOM may be adsorbed strongly to mineral surfaces, resulting in low DOM concentrations in the soil solution. There are strong indications that microbial degradation of DOM also controls the fate of DOM in the soil.

Laboratory experiments on controls of DOM dynamics have often contradicted field observations, primarily because hydrology has not been taken into account. For example, laboratory findings on the effects of plant species (conifer vs. deciduous) on DOM release from forest floors and on the effects of substrate quality (e.g.: C/N ratio) or pH on DOC concentrations were often not confirmed in field studies. The high adsorption capacity of soil clay minerals and oxides for DOM shown in laboratory studies may not control the transport of DOM in soils in the field if macropore fluxes dominate under field conditions. Laboratory findings about the biodegradability of DOM also await verification under field conditions.

Studies that include DON and DOP dynamics in addition to DOC are few. The rate of release and the fate of DOC, DON, and DOP in soils may differ to a far greater extent than previously assumed. Controls established for DOC might thus be not valid for DON and DOP.

Despite intensive research in the last decade, our knowledge of the formation and fate of DOM in soils and its response to changing environmental conditions is still fragmented and often inconsistent. Predictions at the field scale are still very uncertain, and most of the information available today is the result of studies on temperate soils and forest ecosystems. Thus, future research on controls of DOM dynamics should be extended to soils under different land uses and in other climate zones. Emphasis should also be given to: (i) the effects of soil organic matter

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properties on the release of DOM (ii) environmental factors controlling DOM quantity and quality (iii) the assessment of biological versus physico-chemical controls on the release and retention of DOM in soils, and (iv) the differences between DOC, DON, and DOP. Finally, if our goal is to predict DOM concentrations and fluxes in soils, future research on the controls of DOM dynamics should have a strong focus on field studies. (Soil Science 2000;165:277–304)

Key words: Dissolved organic matter (DOM), dissolved organic carbon (DOC), dissolved organic nitrogen (DON), dissolved organic phosphorus (DOP), soils, controls.

SOIL solutions contain varying amounts of dissolved organic matter (DOM), which originate from plant litter, soil humus, microbial biomass or from root exudates. Only small proportions of DOM, mostly low molecular substances such as organic acids, sugars, amino acids, can be identified chemically (Herbert and Bertsch, 1995). Most of what is collectively termed 'dissolved organic matter' in soils is complex molecules of high molecular weight, namely, humic substances. Similar to soil organic matter, a general chemical definition of DOM is impossible. Dissolved organic matter is often defined operationally as a continuum of organic molecules of different sizes and structures that pass through a filter of 0.45 μm pore size. However, many studies in the 1980s and early 1990s used other filter sizes, making it difficult to compare results quantitatively. Fractionation and characterization methods of DOM are reviewed by Herbert and Bertsch (1995).

Recent findings emphasize the turnover of dissolved organic carbon (DOC), -nitrogen (DON) and -phosphorous (DOP) in soils as major pathways of element cycling. Throughout this review we use the term DOM for general aspects, whereas the terms DOC, DON, and DOP are used to discuss specific results concerning the various components. Typical DOC concentrations in soil solutions are listed by Herbert and Bertsch (1995) and Zsolnay (1996). In forest soils the flux of total N from the forest floor into the mineral soil has sometimes been shown to be dominated by DON (Lajtha et al., 1995; Lepistö et al., 1995; Currie et al., 1996). Chardon et al. (1997) emphasize the importance of DOP as the predominant form of P in subsoil solutions of agricultural soils (more than 70% of total P in 70 cm depth), whereas Donald et al. (1993) stress the importance of DOM for redistribution and loss of P in forest soils.

Dissolved organic matter is a major controlling factor in soil formation (Dawson et al., 1978; Petersen 1976), mineral weathering (Raulund-Rasmussen et al., 1998), and pollutant transport (Kalbitz et al., 1997; Temminghoff et al., 1997; Marschner, 1999).

The observed DOM concentration or flux in soils is the net result of processes that *release* DOM—such as leaching from litter or desorption from the solid phase—and processes that *remove* DOM, such as adsorption or decomposition (Fig. 1). These, in turn, depend on external environmental factors such as temperature and precipitation and the physical/chemical characteristics of the soil. In addition, different methods of extraction and collection may yield different estimates for DOM concentration or fluxes (Herbert and Bertsch, 1995).

Tipping (1998) postulated that a pool of potential DOM exists as a part of soil organic matter (SOM) "that is not in solution but is part of the soil solids and able to pass into solution under realistic soil conditions." Potential DOM is controlled and replenished by C inputs from plant litter, root exudates, SOM, and from microbial biomass (Fig. 1, A). The processes involved in (A) are physical and chemical alteration of litter during decomposition, the leaching of substances from litter and the formation of soluble humic substances. It is generally assumed that these processes are dominated largely by biota. The actual DOM concentration of the soil solution may be controlled mainly by abiotic processes (B) such as desorption and dissolution from the pool of potential DOM, but this is still under debate (Dai et al., 1996, Guggenberger et al., 1998). Thus, according to current knowledge, both biotic and abiotic controls are involved in the formation of potential and actual DOM. The pool of DOM can be partitioned further into a mobile and an immobile fraction according to the pore sizes of the soil matrix (Zsolnay, 1996; Tipping, 1998). Only the mobile fraction of DOM present in macro- and mesopores is subjected to convective transport by seepage. DOM in micropores is immobile and interacts with the mobile fraction by diffusion. Additionally, smear-

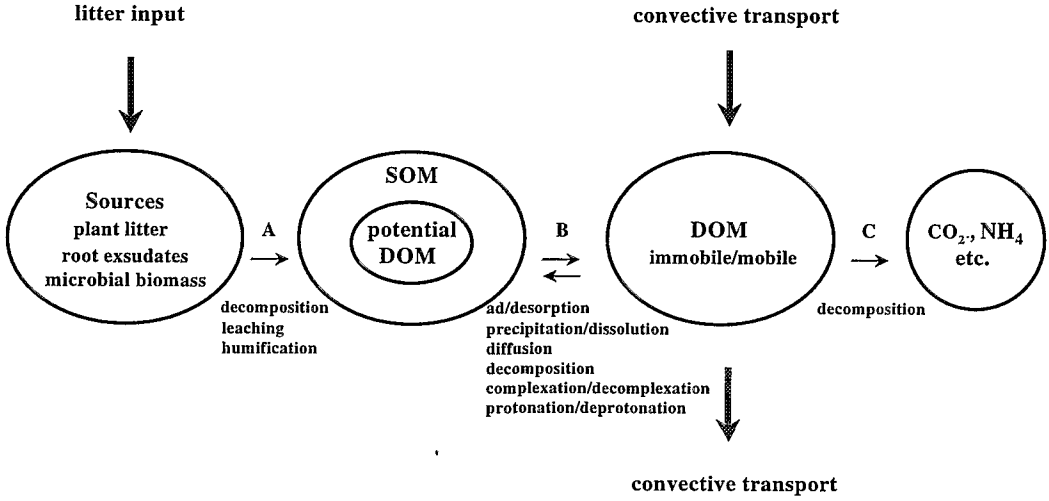

Fig. 1. Conceptual model of the processes involved in the formation of dissolved organic matter (DOM); (SOM: soil organic matter).

ing of the solute front caused by dispersion will occur. Actual DOM can be decomposed (C) or removed from solution by various processes and again become potential DOM (B).

Although DOM is clearly important for a large number of soil processes, the variability of information on its formation, fate, and potential response to changes in environmental conditions and land use makes a comprehensive summary necessary. Controls on DOM have been studied in many laboratory experiments using disturbed soil samples, but fewer studies have focused on the dynamics of DOM under field conditions. Additionally, most of the studies dealing with controls of DOM in soils focus on DOC; much less information is available on DON and DOP dynamics in soils and ecosystems.

The aim of this review is to summarize the recent literature regarding controls on DOM concentrations and fluxes in soils. We focus on comparing results from laboratory experiments with those from field studies. Field observations at the plot scale and on temporal scales from weeks to years are considered. We also concentrate on differences between DOC, DON, and DOP dynamics and their controls. Studies on DOM in surface water and runoff from watersheds will not be considered in detail. We conclude the review by reviewing areas of major uncertainty and proposing areas of future research.

ORIGIN OF DOM

Although the release of DOM has been researched extensively (mainly from forest soil studies), it is still not clear whether DOM originates primarily from recent litter or from relatively stable organic matter in the lower part of organic horizons. McDowell and Likens (1988) hypothesized that leaching and microbial decay of humus rather than of recent litter are largely responsible for the DOC production in the forest floor. Zsolnay (1996) also suggested that humified organic matter is the major source of DOC because of the relatively high proportion of humus in relation to litter in soils.

In contrast, Qualls et al. (1991) reported that, within a deciduous forest ecosystem, the greatest net increases in the fluxes of DOM occurred in the upper part of the forest floor (the litter layer [Oi]). Furthermore, Qualls and Haines (1991) observed unusually high levels of the hydrophilic neutral fraction of DOM in samples collected in November, just after maximum litterfall. Since this fraction is assumed to consist mainly of simple sugars and nonhumic-bound polysaccharides, this result could be interpreted as a significant contribution of recent litterfall to the temporal variation in the DOM production rate.

Casals et al. (1995) observed that most of the organic nitrogen in the leachate in pine forests originated from the freshly fallen litter (Oi) and the partially decomposed litter layers (Oe) of the forest floor (O horizon). According to Huang and Schoenau (1996, 1998) and Michalzik and Matzner (1999), the greatest amount of DOC and DON and water-soluble organic N and P (Huang and Schoenau, 1998) occurred in the Oi layer. The lower organic layers functioned rather as a sink for the organic solutes, leading to a substantial decrease of DOM in the downward flux. Based on

results demonstrating the importance of the labile fraction of forest floor material in the DOM leaching flux, Currie and Aber (1997) assumed in their modeling study of DOM leaching that humus does not contribute to the leaching flux.

The rhizosphere is often associated with a large carbon flux attributable to root turnover and exudation (Edwards and Harris, 1977; Vogt et al., 1983). High microbial activity can result from the input of readily available organic material (Johansson, 1992; Cheng et al., 1996). Substantial amounts of DOM may well derive from this important below-ground C source. However, there has been no attempt to measure directly the contribution of root-derived DOM to the DOM released from the soil.

The soil microbial biomass is also considered a potentially important source of DOM because it is highly labile (Williams and Edwards 1993). Recent studies of fractionation and structural analysis of DOM in soil solutions have shown that microbial metabolites constitute a significant proportion of DOM. According to these studies, the carbohydrate fraction of DOM is chemically different from that of plant residues or bulk humus in that DOM carbohydrates have a higher proportion of hexose- and deoxysugars than pentose sugars. Because pentose sugars are rarely found in microbial cells (Guggenberger et al., 1994a; Huang et al., 1998), DOM may be predominantly of microbial origin. More detailed descriptions are given in a following section (role of decomposer community).

In summary, recent litter and humus are thought to constitute the two most important sources of DOM in soils because of their abundance in the soil. However, the contribution of each of two sources to the DOM production in the forest floor and organic soil horizons is not fully understood. This question of the origin of DOM is important because it can help explain the mechanism of DOM production in organic horizons and the major characteristics of leachate DOM. The importance of other potential sources, such as root exudates and microbial biomass, has also not been evaluated.

SOLID PHASE PROPERTIES AS CONTROLS ON DOM DYNAMICS

Controls on DOM Release from Soil Horizons of High Organic Carbon Content

Amount of Litterfall and Soil Organic Matter

Litterfall represents the most important source of C inputs to the forest floor (Gosz et al., 1976; Nordén, 1994). Yearly terrestrial total litter production is almost equivalent to net terrestrial primary production of about 60 Pg C yr^{-1} (Post, 1993). However, there has been little research on the quantitative effect of litterfall on DOM release. Considering that the largest ecosystem internal flux of DOC occurs as percolate from the forest floor (Zech et al., 1996; Michalzik and Matzner, 1999), the rate of litter incorporation in the forest floor and its rate of degradation into products with varying degrees of humification may ultimately determine the rate of DOM production.

A few studies have found that seasonal changes in the concentration of DOC and DON in forest floor leachate are related to litterfall inputs (Lundström, 1993; Casals et al., 1995; Currie et al., 1996). However, Casals et al. (1995) did not rule out the possibility that this was related to cycles of drying and rewetting and/or to the high temperature that was observed in the same period as high leaching. Although the periods of maximum litterfall and high DOM leaching do not generally coincide (Cortina et al., 1995), it is probable that DOM dynamics in soils are related significantly to the amount of recent litter and organic matter in the soils. Gundersen et al. (1998) observed that DOC flux was correlated to litterfall amount, not to the N status of the sites. They suggested that in forest floor material of several forest sites across Europe, C supply and turnover rate determine DOC leaching from the forest floor. In a modeling study, Currie and Aber (1997) found that relatively high fluxes of DOC and CO_2 were caused by either high relative rates of decay or by high litterfall fluxes. In field studies in experimental forests, the same researchers found that DOC leaching and CO_2 mineralization were correlated positively with the organic matter mass of the forest floors. This is consistent with the results of Tipping et al. (1999), who observed in a field manipulation experiment on three different soils that the soil with the highest organic matter content exported more DOC than the other two soils.

In conclusion, increasing litter production and humus content presumably results in increasing DOM concentration and fluxes. However, this effect cannot be quantified.

Substrate Quality

The quality of litter is determined largely by the dominant vegetation, which thus plays an essential part in controlling DOM concentrations in soil solutions (Kuiters, 1993). Soil solutions from mixed and coniferous stands often contain significantly more DOC and DON than those from hardwood stands (David and Driscoll, 1984;

Cronan and Aiken, 1985; Currie et al., 1996). Cronan (1990) pointed out that DOC export from coniferous forests was 50% higher than from hardwood stands. Modeling work by Currie and Aber (1997) suggest that DOC and DON fluxes are higher in coniferous forests despite slower rates of litter decomposition. Until now, the reason for this difference has been poorly understood. Moreover, Kuiters (1993) showed in a laboratory leaching experiment that much more DOC was released from deciduous leaves (10–25 mg C g^{-1} dry matter) than from coniferous needles (<5 mg C g^{-1} dry matter), possibly as a result of differences in the water permeability of the leaves.

Studies examining the interactions between substrate quality and decomposition rates have focused on the concentrations of nitrogen, lignin, polyphenols, or some combination of these chemical constituents (e.g., Melillo et al., 1982; Melillo et al., 1989). However, few attempts have been made to relate DOM production rates in the soil to the substrate quality of litter and humus. Northup et al. (1995) showed that in a *Pinus muricata* forest ecosystem characterized by extremely acidic and infertile soil, the polyphenol concentration of needle litter controls the release rate of DON. The DON released is assumed to be directly utilized by mycorrhizae minimizing nitrogen loss to competing organisms.

The potential decomposition rate of organic soil is conventionally characterized by its C/N ratio. In the laboratory, however, Michel and Matzner (1999) observed no correlation between DOC and DON release rates and the C/N ratios of forest floor materials sampled from 12 different Norway spruce stands. In contrast to these results, Kalbitz and Knappe (1997) showed in a column leaching experiment that a wide C/N ratio of soil organic matter, a high content of hot-water soluble organic C, and a high proportion of hot-water soluble organic C to the total organic C content determine the amounts of DOC released from the topsoil if the soils have a low capacity to adsorb and precipitate DOC. This is supported by Gödde et al. (1996), who found in a column leaching study that soils with higher C/N ratios showed higher rates of respiration and DOC mobilization. They attributed this result to higher fractions of the more labile Oe (partially decomposed litter) versus Oa (well decomposed organic matter) material in the high C/N soils, making them easier to degrade.

Little information exists about the effect of substrate quality on *in situ* DOM dynamics in soils. Parameters such as the content of carbon or nutrients (N, P) in soil, or ratios between carbon and nutrients (e.g. C/N, C/P) are often not significantly related to DOM release rates (Cortina et al., 1995). Williams and Edwards (1993) also suggested that leaching of DOM is not necessarily constrained by the ratios of C to Na or C to P because during the early stages of decomposition (up to two years), decaying litter can release organic leachates independent of microbial demand for inorganic nutrients (as indicated by the C/N and C/P ratios of the litters). Other possibilities for controls on DOM dynamics in the field are the substrate accessibility for microbes (Bosatta and Ågren, 1991) and the content of soluble organic C in the substrate (Hu et al., 1972; Reinertsen et al., 1984; Cogle et al., 1989).

Decomposer Community

The decomposer community in soil consists of a wide range of bacteria, fungi, protista, and invertebrates (Swift et al., 1979). Microorganisms have received considerable attention because of their "dual roles as a pool of labile nutrients and the agent of decomposition of organic materials" (Lundquist et al., 1999a). Guggenberger et al. (1994a) and Møller et al. (1999) suggested that fungi are the most important agents in the process of DOM production, probably because of incomplete degradation of organic matter by fungi. As described in the previous section, microbial metabolites constitute a significant portion of DOM released from the forest floor. In addition, microbial biomass itself provides an important pool of potential DOM. Yavitt and Fahey (1984), for example, observed a 10-fold increase in DON concentrations in leachate from biocide-treated microcosms, which was considered to be caused mainly by cell death and lysis.

Some evidence indicates that the soil fauna also influences the rate of DOM release by enhancing the rate of turnover of microbial biomass (Williams and Edwards, 1993) and through the release of organic compounds at death (Whalen et al., 1999). In leaching experiments using litter and humus with and without soil fauna, addition of enchytraeid worms (Williams and Griffiths, 1989) and a diverse soil fauna (Huhta et al., 1988; Setälä et al., 1990) increased the rates of inorganic and organic N leaching, presumably as a consequence of reduced microbial immobilization caused by faunal grazing. According to Whalen et al. (1999), 13 to 18% of the N from ^{15}N-labeled decomposing earthworms was transformed into DON in the incubation period of 2 to 16 days, indicating the significance of direct N flux from faunal tissues through mortality.

Controls on DOM Adsorption in Mineral Soil Horizons of Low Organic Carbon Content

Several field studies have shown that concentrations and fluxes of DOM in soil solutions decrease significantly with soil depth (Fig. 2). It is generally assumed that adsorption of DOM to mineral surfaces is far more important than decomposition of DOM in reducing DOM concentrations in mineral soil. A variety of mechanisms has been postulated, including anion exchange, ligand exchange-surface complexation, cation bridging, hydrogen bonding, van der Waals forces, and physical adsorption. Jardine et al. (1989) concluded that physical adsorption (hydrophobic interactions) driven by favorable entropy changes is the dominating process. Kaiser (1996), however, argues that adsorption of hydrophobic DOC components is diminished with increasing soil organic matter content and that ligand exchange should be the most important process (Gu et al., 1994; Edwards et al., 1996).

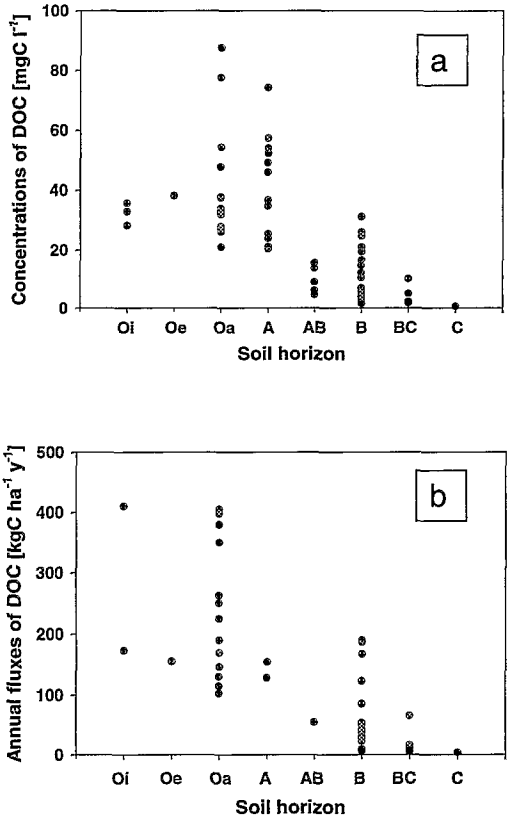

Fig. 2. DOC concentrations (a) and DOC fluxes (b) in different soil horizons (Michalzik et al., 1999; data from 23 references).

The steric position of bound ligands would play an important role in the strength of the bond (Kaiser, 1996), and the formation of strong covalent bindings and multiple site bindings causes a pronounced hysteresis of the adsorption. Thus, DOC sorption is largely irreversible under natural soil conditions. Gu et al. (1994) report that 72 to 92% of DOC sorbed to Fe oxides was irreversibly bound. Inasmuch as Fe and Al oxides and hydroxides are the most important sources of variable charge in soils (Jardine et al., 1989; Moore et al., 1992; Kaiser and Zech, 1998a), DOC adsorption can be related quantitatively to the Fe and Al oxide/hydroxide content of soil samples (Moore et al., 1992). In addition to oxides and hydroxides, clay minerals are also important adsorbents for DOC in soils. DOC adsorption to kaolinite is more effective than its adsorption to illite (Jardine et al., 1989), and DOC concentrations in catchment runoff are negatively correlated to the clay content of soils in the catchment (Nelson et al., 1993). The surface area of minerals is a key factor influencing the adsorption capacity (Gu et al., 1994; Mayer 1994a and b). The adsorption process is relatively rapid: in batch experiments ad- and desorption was completed almost within 2 to 12 hours (Dahlgren and Marrett, 1991; Kaiser and Zech, 1998b).

Organic matter already adsorbed to mineral surfaces reduces the potential adsorption capacity of the adsorbent (Vance and David, 1992; Moore et al., 1992). The maximum adsorption of unfractionated DOC onto Al and Fe oxides/hydroxides ranges from 0.2 to 2.2 mg C m^{-2} surface area (Tipping, 1981; Day et al., 1994; Gu et al., 1994; Kaiser and Zech 1997a). That DOC sorption reaches a maximum suggests that only a limited number of adsorption sites is available and a monolayer of adsorbed DOC is formed on mineral surfaces (Mayer 1994a and b).

The adsorption of DOM leads to fractionation of DOM: hydrophobic compounds are removed selectively from the soil (Jardine et al., 1989; Kaiser and Zech, 1998a), and hydrophilic substances are released into the soil solution. The observation that hydrophilic DOM compounds have higher N contents than hydrophobic DOM compounds (Qualls and Haines, 1991; Gu et al., 1995; Kaiser et al., 1997) suggests that DON would be less strongly adsorbed than DOC, but this has not been investigated to date. One field observation showing that the DOC/DON ratio of soil solutions decreases with soil depth (Qualls and Haines, 1991) supports this hypothesis, although the changing ratio could be attributable

to other factors (e.g., preferential release of hydrophilics) (Kaiser and Zech, 1998b). High molecular weight fractions are preferentially adsorbed compared with low molecular weight components (Gu et al., 1995). The presence of aromatic rings, carboxylic acids, N- and S-containing groups, and amino acid residues in organic molecules increases the adsorption capacity (McKnight et al., 1992).

Anions in soil solutions such as sulfate and phosphate compete with DOC for adsorption sites in forest soils (Tipping, 1981; Jardine et al., 1989; Vance and David, 1992; Gu et al., 1994), but DOC shows a greater affinity for soil than sulfate (Kaiser and Zech, 1998a). Furthermore, DOC adsorption is decreased under anaerobic conditions (Kaiser and Zech, 1997b).

Studies on the effects of pH on DOC sorption have shown widely varying results. The adsorption of organic matter on iron oxides (Tipping 1981; Gu et al., 1994), aluminum hydroxides (Parfitt et al., 1977; Davis 1982), and bulk soil (Jardine et al., 1989; Kennedy et al., 1996) has been shown to increase with decreasing pH as a result of increasing positive charge on the hydroxides. Tipping (1981) showed that maximal adsorption of humic substances onto geothite occurs at pH 5. However, David and Zech (1990) found decreasing DOC adsorption in the B horizon of an acid forest soil with decreasing pH, and, in a batch adsorption experiment, Vance and David (1992) saw no effect of pH (between pH 3 and 6) on DOC adsorption in mineral soil samples. The main reason for these observed differences in the laboratory is that soil minerals have their maximum adsorption capacities at different pH values. Kaiser (1996) concluded that the pH of most natural soils (normally ranging between 3.5 and 6) does not significantly affect DOM adsorption because the adsorption capacity of sesquioxides is diminished substantially only at a pH greater than 6.0 or less than 4.5 to 3.5.

Laboratory studies show that disturbed soil samples can adsorb DOC rapidly and have large adsorption capacities, suggesting low rates of DOC transport to deeper horizons. However, under field conditions, the transport of DOC in soil profiles is often dominated by the flow regime and macropore transport (Fig. 3). Jardine et al. (1990) found significant transport of DOC during storm events and postulated reduced contact time between the solid and solution phases as the cause for this unexpected finding (see Precipitation and Water Fluxes). In contrast, Li and Shuman (1997) concluded that adsorption properties derived from batch experiments could be used to predict the transport of DOC in sandy soils. The limited value of laboratory findings for the prediction of DOC transport in a sandy aquifer under field conditions was pointed out by McCarthy et al. (1996) in an experimental field study. They report the unexpected transport of large, strongly-binding DOC compounds

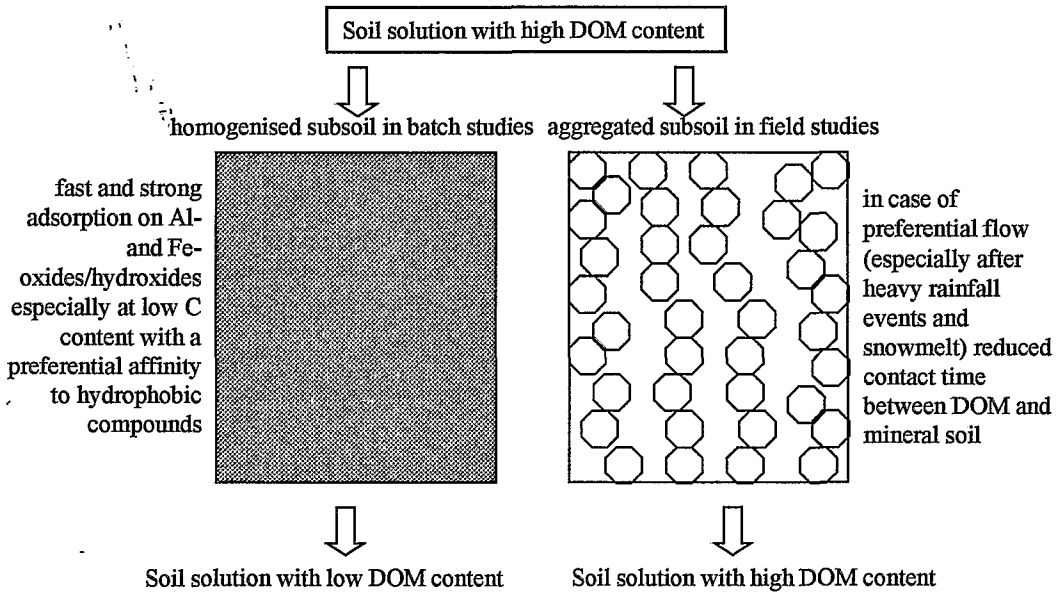

Fig. 3. Effects of soil aggregation on DOM sorption.

from sandy soils under field conditions due to saturation of sorption sites.

In summary, DOM is strongly adsorbed in the mineral soil to Al and Fe oxides/hydroxides and clay minerals, especially if the surface of the adsorbants has low pre-existing levels of adsorbed C. This results in low DOM output from these types of soils. The individual behavior of DON in relation to DOC has not been investigated. Predictions of DOM mobility in soils based on laboratory studies are potentially misleading if macropore fluxes dominate the field situation.

SOIL SOLUTION CHEMISTRY AS A CONTROLLING FACTOR FOR DOM DYNAMICS

pH

pH Effects on DOM Dynamics via Physico-chemical Processes

The effect of pH on the release of DOM is sometimes contradictory insofar as pH can affect different controls on DOM release. Since DOM probably consists mainly of high molecular, polyfunctional electrolytes, its solubility should depend on its charge density (Tipping and Hurley, 1988; Tipping and Woof, 1990), which in turn depends on the pKa value and the pH of the soil solution. Tipping and Woof (1990) calculated that an increase in soil pH of 0.5 units would lead to increases of about 50% in the amount of mobilized organic matter. Therefore, the solubility of DOM is diminished by the high degree of protonation resulting from low pH (Tipping and Hurley, 1988).

Protonation of functional groups might, therefore, reduce the solubility of DOM by altering the steric conformation when intramolecular bonds are cleaved and van der Waals forces and proton bridging become more effective. On the other hand, the dissolution of organic complexes containing polyvalent cations, and the advancing saturation of exchange sites, should result in enhanced mobility of DOM. The interaction of these and other mechanisms make the net effect of pH variation in soil layers difficult to estimate.

Almost all laboratory studies show that DOC release from organic soil horizons is positively correlated to pH (Whitehead et al. 1981; Hay et al. 1985; Tipping and Hurley 1988, Gödde et al. 1996; Jozefaciuk et al. 1996; Kennedy et al. 1996; Hajnos et al.1999; You et al. 1999).

In contrast to organic soil horizons, both batch and column experiments in the laboratory have shown enhanced mobilization of DOC from the solid phase in spodosol soil mineral horizons with decreasing pH (David et al. 1989; Jardine et al. 1989; Vance and David 1989; Guggenberger et al. 1994b). These findings have been attributed to enhanced solubilization of metal-organic complexes by proton competition. In opposite to these mobilizing studies (use of DOC-free extracting solutions), most adsorption experiments (use of DOC solutions) showed the opposite effect (increasing adsorption with decreasing pH; see section Controls on DOM Adsorption).

In addition to the quantity of DOM mobilized, changes in solution acidity can affect its quality. Cronan (1985), David et al. (1989), Vance and David (1989), and Guggenberger et al. (1994b) reported a decrease in the relative amount of hydrophobic acids and a corresponding increase in hydrophilic acids under acid irrigation of spodosol soil columns.

pH Effects on DOM Dynamics via Biological Processes

Guggenberger and Zech (1993), Guggenberger et al. (1994a), and Dai et al. (1996) all characterized DOM as plant-derived organic substances, transformed and modified by microbial metabolism. Therefore, the effect of acidity on microbial activity should be regarded as a crucial factor in controlling DOM in natural ecosystems. Unfortunately, information about the effects of pH on microbial DOM production is only fragmentary.

Chang and Alexander (1984) and Guggenberger et al. (1994b) showed that decreasing DOC mobilization with decreasing pH from incubated spodosol soil columns was, in most cases, accompanied by reduced carbon mineralization. However, Cronan (1985) observed neither reduced DOC mobilization nor lowered carbon mineralization with decreasing irrigation water pH (at pH $\geq$ 3.5) for intact soil cores from coniferous, hardwood, and mixed stands. Curtin et al. (1998) observed no significant correlation between soil pH (ranging from 5.1 to 7.9) and the N mineralization rates of 61 agricultural soil samples from throughout Canada. Experimentally raising the pH from 5.7 to 7.3 (by addition of $Ca(OH)_2$) in these soils, however, increased DOC and DON concentrations of aqueous soil extracts and resulted in higher mineralization rates for C and N (see liming effects).

Cronan (1985) also reviewed the effects of acid deposition on leaching and organic matter decomposition. He postulated that increased H^+ deposition increases the short-term leaching of cations and soluble organic constituents from

forest litter. However, the studies he reviewed generally concur that the biologically mediated mineralization of organic matter is relatively unaffected by acid precipitation at pH > 3.0. Vance and David (1991a) showed that soil respiration is affected little by acid inputs. They also showed that DOC release from columns filled with either forest floor or a reconstructed profile (forest floor and B horizon) was not affected by pH 3.7 solution compared with pH 4.8. Even lowering the extractant pH to 3.0 resulted in only a minimal DOC decrease.

Blagodatskya and Anderson (1998) showed that the fungal-to-bacterial respiratory ratio was significantly higher at pH 3.0 compared with pH 6.0. Taking into account the importance of fungi for DOM release (Guggenberger et al., 1994a; Møller et al., 1999), these results suggest that decreasing pH could cause elevated DOM release.

pH Effects as a Combination of Physico-chemical and Biological Processes as Revealed in Field Studies

The controls on DOM mobilization identified in the laboratory are often not corroborated in the field because of the spatial variability of soil properties and the much longer timescale (months to years vs. hours to days for lab studies). Information about pH-DOM relationships is seldom available at the field scale. Studies investigating the effects of acid deposition on hardwood forests (Liechty et al., 1995), Scots pine forests (Schaaf et al., 1995), or soil solution pH in peaty podzols (Chapman et al., 1995) or Norway spruce (Michalzik and Matzner, 1999) found no effect of acidity on DOC concentrations or fluxes. Michalzik and Matzner (1999) also showed this for DON. Nitrogen fertilization experiments also show no effect of elevated proton fluxes on DOC and DON fluxes in both forest floor leachate and soil solution (Fernandez and Rustad, 1990; Emmett et al., 1998; Stuanes and Kjønass, 1998). Five years of artificial catchment acidification also failed to induce any significant changes in DOC concentrations in a lake in western Norway (Hessen et al., 1997).

One reason for the lack of sufficient correlation between DOC and pH in soil solutions from the field might be that the soil solution in a particular soil horizon has already equilibrated with the bulk soil. This may be especially true with tension lysimeters, which collect water that is held against the gravitational gradient and thus, probably, have a high residence time. Michalzik et al. (1998) accounted for this by correlating the DOC concentrations of leachates from different O layers of spruce and hardwood forest floors with the solution pH of the overlying horizon. Remarkably, these correlations were also not statistically significant.

In contrast, Schindler et al. (1992, 1997) demonstrated a rapid decline in DOC concentration in bog pools of an experimentally acidified wetland, and Cronan and Aiken (1985) found a strong negative correlation between DOC concentrations and the pH between 4.8 and 3.5 of O/A horizon leachates for three forested watersheds. The same was shown by Ross and Bartlett (1996) for spodosol soil solutions from another site. However, Guggenberger and Zech (1993) and Zech et al. (1994) detected significantly elevated DOC concentrations and fluxes at coniferous sites with high acidic inputs and lower soil pH in relation to other sites. They suggested that higher acidic inputs intensified the leaching of bridging polyvalent cations such as Al^{3+}, Ca^{2+}, and Mg^{2+} and, therefore, enhanced the solubilization of organic matter.

Whether the mobilization of DON and DOP follows the same patterns as DOC is still unknown as information about these components is scarce. Raubuch and Beese (1997) found that increasing soil acidification led to reduced N mineralization and higher carbon respiration. The decline of DOC concentrations induced by artificial acidification in a northwestern Ontario lake was accompanied by rising DON concentrations, suggesting that DOC and DON may respond differently to acid inputs, at least in aquatic systems (Schindler et al. 1992).

In conclusion, mobilization of DOM from forest floors and soil organic matter seems to be favored by increasing pH values due to physicochemical properties (Fig. 4). The influence of pH on the microbial production of DOM is not clear. DOM release from subsurface horizons is caused primarily by desorption of formerly adsorbed organic matter. At high pH values, adsorption capacity is diminished and, therefore, DOM mobilization is enhanced. At low pH values, dissolution of organo-metal complexes can contribute to leaching of DOM. The most important conclusion is that despite the reported pH effects on the dynamics of DOM in the laboratory, in the field these effects seem to be small, i.e., within the normal pH range of the soils (Fig. 4).

Ionic Strength

The effect of ionic strength on DOM mobilization is ambiguous and may involve several processes that differ in significance for each soil compartment. Solubilization of organic matter

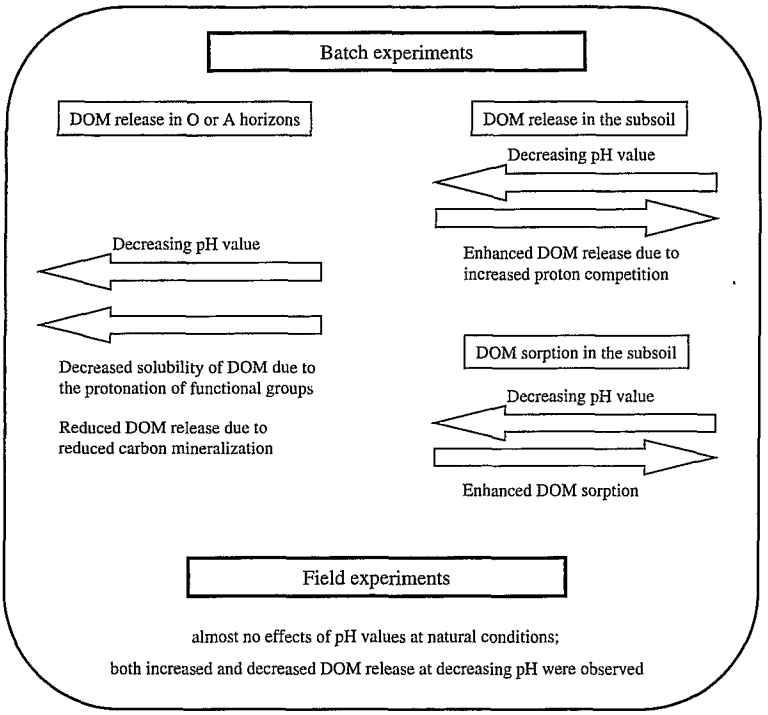

Fig. 4. pH effects on the release and adsorption of DOM in batch and field studies.

might be impeded by high ionic strength solution reducing the charge density of organic substances (Tipping and Hurley, 1988) leading to coagulation. On the other hand, competition between DOM and other anions could displace DOM from adsorption sites, thus enhancing leaching.

Evans et al. (1988) found DOC mobilization from soil columns of A, E (eluvial) and Bs (illuvial accumulation of sesquioxides) horizon material leached with acid solutions to be negatively correlated to ionic strength, a result supported by Kaiser (1992) with A and B horizon samples from acid forest soils, by Tipping and Hurley (1988) with organic soils, by Vance and David (1989) with mineral B horizons, and by Davis (1982) with Al-oxides. Skyllberg and Magnusson (1995) reported the same relationships for Oa and A horizon material of Humic Gleysols but found DOC mobilization from Oe and Oa horizon material of Haplic Podzols from the same site to be insensitive to changes in ionic strength. They hypothesized that the differences were caused by different stages of humification of organic matter in both soils. In a column-leaching experiment with soil from a mixed forest stand, Berry et al. (1990) observed a decrease in DOC mobilization from Oa horizon material with increasing ionic strength of percolation solutions but increased DOC leaching from mineral horizon material and from a reconstructed profile (Oa/EB). The authors gave no explanation for this different behavior of forest floor and mineral soil horizons. Gu et al. (1994) found no difference in the amount of DOC adsorbed onto hematite between NaCl solutions of 0.01 and 0.1*M*. Jardine et al. (1989) also found no significant correlation between DOC adsorption to B-horizon soil samples and the ionic strength of the solution used.

Fotovat and Naidu (1998) investigated DOC adsorption onto acidic and alkaline soil samples in relation to ionic strength. They found DOC adsorption on acid soils diminished with increasing ionic strength up to 0.05 M (up to 0.03 M for alkaline soils) but remained constant at higher electrolyte levels up to 0.2 M.

Although information about ionic strength effects on DOC release from litter and soils is not always consistent, increasing ionic strength seems to reduce mobilization of DOC, at least in organic horizons. In addition it may be difficult to tease effects of ionic strength apart from pH effects as the former strongly influences the latter in acid forest soils (Wiklander, 1975; Richter et al., 1988; Kaiser and Kaupenjohann, 1998).

Specific Cations and Anions

The mineral ions in soil solution may affect the solubility of DOM. This section deals with the effects of cations and anions separately although, in reality, their effects on DOM are closely interrelated and are also affected by factors discussed previously.

Cations

The activities of polyvalent cations such as Al^{3+}, Fe^{3+}, Ca^{2+}, or Mg^{2+}, and trace metals such as Cu^{2+}, Mn^{2+}, Pb^{2+}, Cr^{3+}, or Cd^{2+} affect the solubility of organic matter (Baham and Sposito, 1994; Guggenberger et al., 1994b). Monovalent cations such as Na^+ and K^+ also interact with organic matter. Chemical reactions between anionic functional groups of organic molecules and solution cations can reduce the surface charge density, alter the structural conformation of the adsorbed species, and consequently reduce solubility. Additionally, polyvalent cations can link negatively charged functional groups of organic molecules together and, hence, reduce their solubility by flocculation or binding them to negatively charged exchange sites (cation bridging) (Greenland, 1971; Tipping and Woof, 1990, 1991). Supporting this, McDowell and Wood (1984) found that DOC adsorption onto B horizon material was increased strongly by the addition of Al and Fe salts. Dahlgren and Marrett (1991) suggested that the main mechanism of DOC immobilization involved initial solubilization of metals, which lowered C/metal ratios in solution, followed by adsorption of the organometal complexes. The C/metal ratio was, therefore, an important factor for DOC adsorption. McDowell and Wood (1984) also found that DOC mobilization from the forest floor was correlated negatively to the concentrations of acid-soluble Fe and Al in litter. In a series of batch titrations, Mulder et al. (1992) found DOC release from podzol B horizon samples to be strongly reduced by the addition of Al. Precipitation of DOC occurred in soil solution after Al addition even in the absence of soil (Mulder et al., 1992). A striking example for the significance of valence for DOC mobilization is given by Skyllberg and Magnusson (1995), who equilibrated soil samples of two soils from northern Sweden with salt solutions of Al, Ca, and Na prior to centrifugal extraction. The amounts of DOC extracted were highest for Na and lowest for Al (coagulating effect), with the most pronounced differences in the more humic soil samples. In batch experiments, Römkens and Dolfing (1998) showed that Ca enhances the precipitation of DOC from arable spodosols, with the high-molecular weight fraction of DOC preferentially precipitated. Low-molecular weight organic substances were almost unaffected.

Anions

Anions can affect the adsorption of DOC by displacing DOC from sorption sites. The strength of displacement seems to be related to the valence, with $PO_4^{3-} > SO_4^{2-} > Cl^-$ (Gu et al., 1994; Reemtsma et al., 1999). Many authors confirm the role of phosphate in displacing DOM from sorption sites (Tipping, 1981; Kaiser and Zech, 1997a; Beck et al., 1999), even when PO_4^{3-} is applied at concentrations one order of magnitude lower than those of other anions (Gu et al., 1994).

Nodvin et al. (1986a) and David and Zech (1990) showed that F^- is preferentially adsorbed over DOC onto spodosol soils, but its ecological significance is marginal because concentrations of F^- in soil solutions are usually low. In contrast, SO_4^{2-} is the dominant anion in precipitation throughfall and soil solutions in forest soils of many regions, and competition between SO_4^{2-} and DOC for sorption sites is a well established phenomenon (Singh and Johnson, 1986; Nodvin et al., 1986b; Vance and David, 1991a; Kooner et al., 1995; Kaiser and Zech, 1996). Vance and David (1991a) observed in leaching experiments that more DOC is released in the presence of SO_4^{2-} than NO_3^- at the same pH. In addition, they found that the distribution coefficients (solid phase vs. solution) of DOC were substantially higher than those of SO_4^{2-} (Vance and David, 1992). Kaiser and Zech (1997a, 1998a) pointed out that SO_4^{2-} shows no higher affinity to adsorption sites than DOC in acid forest soil samples, on goethite, or on amorphous $Al(OH)_3$. The authors concluded that SO_4^{2-} displaces DOC from adsorption sites only when present in concentrations > 10 mmol L^{-1}.

In summary, polyvalent cations usually reduce DOC leaching from organic soil horizons and enhance DOC retention as a result of cation bridging and precipitation. Sulfate and phosphate compete with DOC for adsorption sites. Phosphate has repeatedly been shown to displace adsorbed DOC, whereas the effect of sulfate in the field is less straightforward.

ENVIRONMENTAL CONTROLS ON THE DYNAMICS OF DOM

Various environmental factors influence DOM concentrations and fluxes in soils. These

environmental controls may be natural or anthropogenic. We first review the effects of the most important natural environmental factors, temperature, precipitation, soil water fluxes, soil moisture, and snowmelt. We then discuss important anthropogenic controls on DOM dynamics in soil. Because only a few studies concerning environmental controls on DOM dynamics address DOM fluxes, we will focus on DOM concentrations.

Temperature

Mulholland et al. (1990) postulated that temperature will always be a factor regulating microbial production of DOC, and "a better understanding of these factors may help to identify upper limits on DOC formation and explain the range of DOC concentrations observed in terrestrial and aquatic environments." Strong correlations between soil organic C pools and climate (Jenny, 1980; Post et al., 1982, 1985) imply temperature may affect DOM release from soils. Although the amount of soil organic matter is negatively correlated with temperature at both the global and the local scale (Post et al., 1982; Kirschbaum, 1995), the relationship between temperature and DOM release is equivocal. DOM production in catchments can increase in warmer climates, but the effect of climate on DOM is probably small because decomposition rates, which remove DOM, also increase (Kramer et al., 1990). Liechty et al., (1995) estimated that the differences in soil temperatures (2.1 °C) could be responsible for as much as 16% increase in DOC concentrations in forest floor solutions at the warmer compared with the colder site.

Soil drainage conditions play an important role in understanding climate effects on DOM release. For well drained and moderately drained soils, there is often an inverse correlation between average soil temperature and DOC concentration in surface soil leachates (Cronan, 1990). Thus, DOC concentrations generally increase in cooler environments. In poorly drained areas, DOC concentrations in surface horizons often reach very high levels, regardless of the climate, from cool northern peatlands to warm southern blackwater swamps. These inconsistent data indicate no general climatic effect on DOC.

DOM production may be affected by diurnal temperature variation as postulated by Kramer et al. (1990), but there are no field studies available that address this hypothesis. On the other hand, numerous field studies show seasonal variability in DOM concentrations and fluxes. In general, DOC concentrations in soil solution are higher in summer than in winter (Collier et al., 1989; Dalva and Moore, 1991; Chittleborough et al., 1992; Federer and Sticher, 1994; Heikkinen, 1994; Guggenberger and Zech, 1994; Liechty et al., 1995; Tegen and Dörr, 1996; Scott et al., 1998; McDowell et al., 1998; Guggenberger et al.; 1998; Tipping et al., 1999). Cronan and Aiken (1985) found mean DOC concentration increased by 26 to 32% in shallow soil solution in summer, but DOC concentrations in the B horizon remain relatively constant. The stability of DOC concentration in deeper soil horizons in summer was also corroborated by Qualls and Haines (1991) and Chapman et al. (1995). Differences in temperature effects between upper and lower soil horizons support the hypothesis that DOC concentration in the topsoil, with higher microbial activity than subsoil, is controlled mainly by microbial processes (Guggenberger et al., 1998) which are, in turn, partly controlled by temperature. This is also supported by the observation that hydrophilic compounds (derived mainly from microbial sources) (Bourbonniere, 1989; Vance and David, 1991b; Scott, et al., 1998) and carbohydrates (Guggenberger and Zech, 1994) are released preferentially into the soil solution during the growing season rather than in the dormant season.

However, studies on the seasonality of DOC concentration also show variable results. Vance and David (1991b) and Dosskey and Bertsch (1997) found no seasonal effects on DOC concentration in soil solution. The lack of seasonality shown by Dosskey and Bertsch (1997) may be due to relatively short, mild winters (South Carolina) with neither concentrated growth seasons in summer, nor litterfall periods in autumn (Dosskey and Bertsch 1997). In other words, this lack of seasonality was caused by a lack of seasons. Evans et al. (1996) report that DOC peaks can occur throughout the year.

Michalzik and Matzner (1999) found a correlation between temperature and both DOC and DON concentrations in the forest floor of a Norway spruce stand. However, fluxes of DOC and DON were not correlated to temperature. They concluded that abiotic processes supercede biological controls on DOM release under field conditions. In contrast, Liechty et al. (1995) found a significant positive correlation between soil temperature and DOC flux in forest floors of two hardwood stands. Unfortunately, studies on the effects of temperature on DOM fluxes in soil are rare. However, it seems reasonable that high DOC concentrations in summer are linked with low water fluxes because of much higher evapotranspiration in the growing than in the dormant

season and that temperature effects on microbial activity may be a secondary factor.

Compared with DOC, there are fewer data available describing temperature effects on DON or DOP dynamics. According to Currie et al. (1996) and McDowell et al. (1998) DOC and DON show the same seasonal pattern, with higher concentrations in summer than in winter. In a study by Michalzik and Matzner (1999), the DON concentrations in the Oe layer were affected more by temperature than were DOC concentrations. Huang and Schoenau (1998) found decreasing concentrations of water-extractable organic nitrogen and phosphorus in summer because of high mineralization rates.

Some studies have addressed the effects of temperature on DOC in the laboratory, but we have found no such studies for either DON or DOP. The results of laboratory studies are more consistent than field investigations, with nearly all showing that an increase in temperature results in increased DOC concentrations. Only Seto and Yamagiya (1983) and MacDonald et al. (1999) did not find temperature effects on DOC concentrations in soil solution. Decomposition of DOC was probably enhanced to the same extent as DOC production. Tipping et al. (1999) concluded that warming and drying can accelerate the production of potential DOM in organic soil. Gödde et al. (1996) reported Q_{10} values of 2.0 and 1.5 for DOC from eight different red spruce stands between the ranges of 3 and 10 °C and 10 and 20 °C, respectively. Christ and David (1996b) showed relatively constant Q_{10} values (near 2.0) over a broad temperature range (3–28 °C) for DOC from one red spruce stand. The DOC response to temperature seems to depend on site conditions. In both studies, the Q_{10} values for DOC release are lower than those for respiration. Thus, laboratory studies suggest microbial respiration is more susceptible to increasing temperature than DOC release, implying that DOC release with increasing temperature may be caused partly by physical leaching rather than being entirely dependent on microbial activities (Moore, 1998).

Summarizing the effects of temperature on DOM in soils, there is a trend of increasing DOM concentrations with increasing temperature that is more obvious in laboratory experiments than in field studies. It seems unlikely that the release of DOM (DOM fluxes) under field conditions depends entirely on temperature. Climatic and hydrological conditions, litterfall and litter quality, and soil texture and other soil properties can modify and even mask the temperature response of DOM in the field. It is not clear at present how the release of DON and DOP is affected by temperature.

Soil Moisture

One of the most consistent findings in both field and laboratory studies is that DOC concentrations increase following rewetting after dry periods (McDowell and Wood, 1984; Zabowski and Ugolini, 1990; Haynes and Swift, 1991; Chittleborough et al., 1992; Kalbitz and Knappe, 1997; Lundquist et al., 1999b; Tipping et al., 1999; Zsolnay et al., 1999). It is likely that reduced rates of decomposition in dry soils cause microbial products to accumulate. This, together with cell death and lysis, can contribute to high DOC concentrations in the soil leachate after dry periods (rewetting effect).

Lundquist et al. (1999b) gave three possible explanations for the increase in DOC during rewetting cycles: (i) reduced microbial utilization of DOC in dry periods, (ii) enhanced turnover of microbial biomass and condensation of microbial products by rewetting, and (iii) disrupted soil structure making previously sequestered carbon more available as DOC. According to Christ and David (1994, 1996b), the high proportions of both hydrophilic neutrals and bases suggest a disruption of microbial biomass after soil drying, whereas wetter conditions favor the release of hydrophobic acids. Field studies indicate that the duration of dryness, temperature, and water fluxes are all correlated positively to the amount of DOC released at the onset of soil leaching after a dry period (Chittleborough et al., 1992; Tipping et al., 1999).

In contrast, in another field study, Guggenberger and Zech (1994) found no effects of soil moisture on DOC concentration and composition. Studies on the overall relationship between soil moisture and DOC concentration have shown varying results. Falkengren-Grerup and Tyler (1993) reported higher DOC concentrations at higher water contents in forest floor samples. This positive correlation between the DOC concentration and moisture in well drained soils might be attributable to enhanced biological activity with increasing moisture, subsequently increasing the formation of water-soluble organic compounds. Christ and David (1996b) support these results by observations in the laboratory and conclude that soils would release the most DOC during wet summers.

In most cases, anaerobic conditions caused by saturation increase the release of DOM from soils (Mulholland et al., 1990; Sedell and Dahm, 1990).

Several studies (summarized by Moore 1998) have shown that differences in DOC concentrations among streams are strongly related to the amount of wetland (peatland) drainage contributing to streams (Eckhardt and Moore, 1990; Hemond, 1990; Koprivnjak and Moore, 1992; Hope et al., 1994; Mulholland 1997). Anaerobic decomposition of organic matter is less efficient than decomposition under aerobic conditions, with a higher proportion of water-soluble intermediate metabolites released (Otsuki and Hanya, 1972; Mulholland et al., 1990). Laboratory experiments also show that waterlogged soils release higher levels of water-extractable organic carbon than controls. (Homann and Grigal, 1992; Kalbitz et al., 1997).

In summary, increased concentration of dissolved organic matter (i) after dry periods and (ii) as a result of anaerobic conditions seems to be the most important effect of soil moisture on the dynamics of DOM. This conclusion can be drawn from both field and laboratory studies.

Precipitation and Water Fluxes

In addition to temperature and soil moisture, the dynamics of precipitation and water fluxes are also greatly responsible for seasonal changes in concentrations and fluxes of dissolved organic matter in soils. An inverse relationship between DOC concentration and water fluxes in organic soil horizons suggests that a simple leaching model might explain some of the seasonal changes in DOC (McDowell and Wood, 1984). Following this up, Easthouse et al. (1992) proposed that there would be considerable leaching and dilution of DOC during high precipitation events in O horizons. The contact time between the soil and the soil solution is decisive (McDowell and Wood, 1984; Michalzik and Matzner, 1999). Thus, in the spring, with more water passing through the forest floor and short contact times, DOC concentrations are lower; in summer, however, low soil water content and longer contact times may lead to higher DOC concentrations (McDowell and Wood, 1984; Bourbonniere 1989). Cronan (1990) and Heikkinen (1994) report increasing DOC concentrations from long contact times in soil horizons with high organic matter content. This concept, derived from field studies, is supported in the laboratory where, for instance, DOC adsorption is more pronounced at lower than at higher pore water velocity (Weigand and Totsche, 1998).

Storm events can significantly alter DOC concentrations and fluxes throughout the year by shifting dominant flowpaths toward preferential flow through macropores, runoff, and lateral flow. High pore water velocity leads to low contact times between soil solution and the solid matrix and creates conditions of chemical and physical nonequilibrium. Thus, adsorption of DOC is diminished in mineral soil horizons (Yavitt and Fahey, 1985; McDowell and Likens, 1988; Moore, 1989; Meyer, 1990). Flushing of DOC adsorbed on aggregate surfaces and concentrated in subsoil horizon micropores also contributes to increasing DOC concentrations and fluxes at the beginning of storm events (Jardine et al., 1990; Chittleborough et al., 1992).

For most storm events, the highest DOC concentrations were observed in the initial breakthrough phase of soil solution at any depth, with DOC decreasing rapidly to a baseline level as flow continues (Jardine et al., 1990; Easthouse et al., 1992). Boyer et al. (1996), modeling DOC dynamics as a function of water flow, suggested that DOC in the upper soil might build up during periods of low flow and be transported into streams during periods of high flow. Tipping et al. (1997) interpreted the observation that the highest DOC concentrations and fluxes occur in the fall in terms of soil hydrology and humification. They argued that in summer, the microbial processing of organic matter produces "potential dissolved organic carbon." At this time, percolating water penetrates relatively deeply into the soil, where DOC adsorption maintains low soil solution concentrations. As the soil wets up in fall, lateral flow becomes more significant. The proportion of the drainage water that passes through only surface horizons, and is therefore rich in DOC, increases.

Field data showing that DOC release increases after large rainfall events are confirmed in soil column studies in the laboratory as both high water fluxes (Kalbitz and Knappe, 1997) and high leaching frequency (Christ and David, 1996a; Gödde et al., 1996) increase the amounts of DOM released.

In contrast, the level of rainfall had no effect on the concentration of DOC in the soil solution in sandy soils (Dosskey and Bertsch, 1997). Here, preferential water and solute flow through macropores may be much reduced and, therefore, water flux intensity may be less important to DOC release than for more clayey soils. The lack of correlation between DOC/DON concentrations and water fluxes has been shown for forest floor of a coniferous stand (Michalzik and Matzner, 1999), for both forest floor and mineral soil solution (only DOC) (Guggenberger and

Zech, 1994), and for forest floor leachates of both a hardwood and a coniferous forest (Michalzik et al., 1998). Since the effects of rainfall events on DOM concentrations and fluxes occur on a time scale of hours to days, a relatively high sampling frequency is necessary to detect possible effects (Michalzik et al., 1998).

The most significant effect of precipitation and water fluxes on DOM is the release of DOM at the beginning of large rainfall events. Initial DOM concentrations and fluxes in mineral subsoil horizons are highest under the following conditions: distinct preferential flowpaths, large accumulations of potential water-soluble organic compounds in the soil (caused, for example, by elevated temperature or dry periods without soil leaching), and very intensive rainfall. In addition to flushing out stored or adsorbed DOM at the beginning of storms, simple dilution of DOM during high precipitation events in O horizons is often observed. Temperature-dependent seasonal changes in DOM release from the forest floor and A horizon may be masked by these effects of precipitation and water flux (Mulholland et al., 1990).

Freeze/Thaw Cycles and Snowmelt

Laboratory studies have shown that simulated freeze/thaw cycles also cause DOM release (reviewed by Zsolnay, 1996), with the amount released a function of the water content of the soil before freezing. DeLuca et al. (1992) suggest that freeze/thaw events in soil disrupt microbial tissues (similar to the effects of drying and re-wetting or chloroform fumigation).

During snowmelt, the highest concentrations of DOM in forest floor leachates (Yavitt and Fahey, 1985) and upper soil horizons (Boyer et al., 1997) are observed in the early stages, with rapidly declining concentrations as melting progresses. Most of the DOM leached at snowmelt probably accumulated under the winter snowpack by decomposers continuously releasing soluble organic matter (Fahey, 1983; Fahey et al., 1985; Yavitt and Fahey 1985). In addition to this long-term accumulation of potential water-soluble organic compounds, organics could have become soluble in a rapid flush in spring just before snowmelt, as microbes responded to the initial wetting front reaching the forest floor (Yavitt and Fahey 1985). Another possibility for increased DOM concentration at the beginning of snowmelt is disrupted soil structure (caused by freeze/thaw cycles) making previously-stabilized organic matter more available as DOM.

As usual, there are far fewer studies for DON or DOP than there are for DOC. However, one study did show differences in the behavior of DOC and DON in forest floor leachate during snowmelt (Yavitt and Fahey, 1985). DOC concentrations decreased more than 3-fold during the snowmelt period, whereas DON concentrations decreased by 8-fold, resulting in an increasing DOC/DON ratio from 20:1 to 75:1. The authors gave no explanation for this phenomenon.

Most studies show an increase in DOC leaching during the early stages of snowmelt. If the forest floor has good percolation throughout the year, no decomposition products can accumulate and there will be no significant increase in concentrations or fluxes of DOM during snowmelt (Currie et al., 1996).

N-Effects

Although various aspects of the effects of elevated N deposition and N fertilization have been studied, little is known about the effects of N deposition on DOM turnover. Since it is hypothesized that large amounts of labile C are required to drive N immobilization (Aber, 1992), DOC concentrations and fluxes should, in theory, be high under N-limited conditions and low under N-saturated conditions (Gundersen et al., 1998). In a field N-addition experiment (Currie et al., 1996; McDowell et al., 1998), concentrations and fluxes of DOC from the forest floor remained unchanged, whereas DON concentrations showed a 200 to 300% increase with the highest rate of N amendment (150 kg N ha^{-1} yr^{-1}) and lesser but significant DON increases at lower-N treatments. At present, it is speculative to assess the ecological significance of this additional DON production (McDowell et al., 1998).

In a field study, Cronan et al. (1992) showed that the DOC release rate decreased by 20% after N fertilization (NH_4Cl) of a forest soil. This could be due, in part, to decreasing pH and increasing ionic strength (see sections pH, Ionic Strength). Stuanes and Kjønass (1998) and Emmett et al. (1998) found no changes in DOC or DON concentrations and fluxes in either the forest floor or the A horizon after N addition to conifer forests in Sweden and Wales, respectively. In a comparison of five sites (NITREX project), Gundersen et al. (1998) found no consistent long-term response in DOC leaching to N addition and no clear relationship between DOC flux and the N status of control sites. The original hypothesis that DOC should decline under conditions of N saturation was rejected (Aber et al., 1998; Gundersen et al., 1998; McDowell et al., 1998), and there was no evidence that available C decreases with N availability (Gundersen et al., 1998).

In contrast to the previously discussed studies, Zech et al. (1994, 1996) suggested, based on chemical structural studies, that elevated N deposition may increase in DOC mobilization rates in the forest floor by stimulated microbial activity and N-induced suppression of ligninase, which results in an accumulation of moderately degraded and highly water-soluble soft-rot products.

For arable soils, Chantigny et al. (1999) report that decreasing soil mineral N content was consistently associated with an increase in water-soluble organic carbon. In a field study, however, Rochette and Gregorich (1998) found no effect of inorganic N addition on DOC concentrations in an arable soil.

Many laboratory studies have addressed the effects of nitrogen on organic matter decomposition. A few laboratory experiments show that DOC production in soils can be sensitive to N status, but the results are not consistent. Nitrogen addition as urea (Homann and Grigal 1992, who used O and A horizons of forest soils) resulted in increased release of water-soluble organic carbon. Gödde et al. (1996), however, found significantly lower DOC production in samples with high total N and low C/N ratio in soil organic matter, whereas Michel and Matzner (1999) could not find an effect of the C/N ratio in soil organic matter on the release of DOC and DON.

At present, a clear conclusion about the effects of an increased N-input cannot be drawn. In most cases, DOM dynamics appear to be independent of N-inputs. In some cases, however, both increased and decreased DOM concentrations are reported.

Land Use and Management Effects

Land use changes, such as clear-cutting of forest stands, afforestation, liming and fertilization, converting forests into arable sites, and other management activities, influence the dynamics of DOM by (i) changing the input of organic matter, (ii) changing the substrate quality, and (iii) altering the rates, extent, and pathways of microbial degradation and synthesis of organic matter (Cronan et al., 1992).

Clear cutting and afforestation can have diverse effects on DOM concentrations and fluxes (Sollins and McCorison, 1981; Moore, 1989, Hope et al., 1994; Neal and Hill; 1994). Some studies (e.g., Johnson et al., 1995) have found an increase in DOC concentrations and fluxes in the forest floor and mineral soil horizons after clearcut, some (e.g., Meyer and Tate 1983) have found a decrease, and others (e.g., McDowell and Likens, 1988; Moore and Jackson, 1989) have found little change in DOC. Lepistö et al. (1995), finding no relationship between DOC concentration and clear cutting, postulated that the effect of clear cutting on DOC fluxes was caused mainly by increased runoff volume. Quideau and Bockheim (1996, 1997) reported increased DOC concentration in the soil solution after the afforestation of a former prairie site. However, another study has shown that the afforestation of a wetland did not affect DOC concentration in soil solution (Collier et al., 1989).

Peatland drainage also has variable effects on DOC concentrations, although DOC fluxes generally increase because of higher runoff (summarized by Moore, 1998).

Concentrations of C in solutions from soils in northern Saskatchewan (Cryoboralfs) decreased in the order aspen forest > recently cleared forest > wheat/fallow field (Ellert and Gregorich, 1995). In general, forest soils have higher topsoil DOM concentrations than arable soils (Seto and Yui, 1983). However, the cultivation of a forest soil led to a 2- to 5-fold increase in DOC concentration due to intensified mineralization during the first stage of cultivation (Delprat et al., 1997). During the second stage, once the original organic matter was stabilized, DOC concentrations decreased with increasing time of cultivation (Delprat et al., 1997). After 26 years of corn cultivation, the DOC concentration was less than that of the neighboring forest soil (Delprat et al., 1997). Reviewing long-term field experiments, Campbell et al. (1999) found that water-soluble organic C levels in arable soils were highest in the most fertile treatments and in fields under crop rather than fallow. In a field study, flow-weighted mean concentrations of total organic nitrogen in watershed discharge (streams) increased as the proportion of cropland in the watershed increased (Jordan et al., 1997). Studies comparing effects of different crops on DOM concentrations and fluxes in the field are not available, although there are considerable differences between the water-extractable organic carbon contents of various crop plants (Zsolnay, 1996). More information concerning plant species effects on dissolved organic matter is given in the section titled Substrate·Quality.

Much information is available about the effects of liming on the dynamics of DOM in forest soils. In most cases liming results in increased DOC concentrations in upper soil horizons (Grieve, 1990; Göttlein and Pruscha, 1991; Göttlein et al., 1991; Andersson et al., 1994; Gensior, 1995; Kreutzer, 1995; Erich and Trusty, 1997). This may be caused by the higher solubility of DOM at

higher pH (Thurman, 1985), stimulation of microorganism activity (Kreutzer, 1995), and a combination of these two factors (Andersson et al., 1994). Exceptions to these studies include Cronan et al. (1992), who reported no effects of liming on DOC concentration of forest soils, and Simard et al. (1988), who found that DOC flux from the A horizon of an arable soil decreases with increasing lime amendment, possibly because of an enhanced decomposition of DOM and/or a reduced solubility by increased ionic strength and Ca^{2+} concentration (see Ionic Strength and Specific Cations and Anions sections).

Only a few studies are available on the effects of liming on DON. Both Göttlein et al. (1991) and Kreutzer (1995) demonstrated that in addition to observed increases in DOC, DON leaching from forest floors also increased in limed plots. Since both DOC and DON increased, the DOC/DON ratio was unaffected by liming (Göttlein, et al., 1991).

In agricultural soils, fertilization with organic compounds has been shown to increase the content of water-extractable organic carbon by a factor of 2.7 to 3.2, depending on the kind of fertilizer added (Zsolnay and Görlitz, 1994; Rochette and Gregorich, 1998; Gregorich et al., 1998). This effect is probably based on the direct addition of water-soluble organic carbon to the soil (Zsolnay, 1996). In manured soils, soluble C increased sharply after manure application, followed by a gradual decrease (Rochette and Gregorich, 1998). Cronan et al. (1992) reported an increase of DOC concentration in the forest floor after a sawdust amendment. Organic farming results in higher water-extractable organic carbon content than conventional farming, probably because of the higher input of organic matter by animal manure (Lundquist et al., 1999b).

No information is available about the effects of tillage on DOM dynamics in agricultural soils, although one would expect DOM release to increase after soil tillage as the result of enhanced mineralization of organic matter.

Although the effects of forest clear cutting and afforestation on DOM dynamics are still unclear, both liming and organic fertilization consistently result in enhanced release of DOM due to stimulated mineralization and the addition of water-soluble organic matter, respectively. Both field and laboratory studies support this conclusion.

CONTROLS ON THE DECOMPOSITION OF DOM

Although Guggenberger et al. (1998) postulate predominantly abiotic controls on DOM retention in mineral soil, numerous laboratory studies show that a considerable portion of DOM can be decomposed by microbes. These studies ranged in duration from hours (Yano et al., 1998) to days (Servais et al., 1987; Lucena et al., 1990; Block et al., 1992; Boyer and Groffman, 1996) to a few months (Zsolnay and Steindl, 1991; Qualls and Haines, 1992).

According to Yano et al. (1998), between 12 and 44% of DOC released from the forest floor could be decomposed in solutions by the indigenous microbes. DOC decomposition was also related to nitrogen status: microbial degradability of DOC was higher (43–44%) from plots with a higher N amendment than from the control plots (12–15%). Other experiments have estimated that the biodegradable fraction of DOC is between 10 and 40% (Grøn et al., 1993; Boissier and Fontvieille, 1993; Nelson et al., 1994; Boyer and Groffman, 1996). The range may be partly a consequence of different methods of extracting soil solutions (lysimeter, aqueous soil extracts).

These studies highlight that a distinction between a labile and a refractory DOC fraction is ecologically reasonable (Zsolnay and Steindl, 1991). Zsolnay and Steindl (1991) found in aqueous soil extracts that the solution content of refractory organic carbon is relatively constant, whereas the labile part of DOC ranges between 16 and 68%. Zsolnay (1996) concluded that much of the fresh DOC introduced or produced in the soil has a high substrate value for microbes.

The bioavailable portion of DOC (labile fraction) depends on soil depth (declining with depth) and land use (higher in cornfield than in forest soils) (Boyer and Groffman, 1996). Although DOC concentration, microbial biomass and respiration were significantly higher in organic than in conventional crop farming, the portion of labile DOC was lower in the organic than in conventional soil (Lundquist et al., 1999b). The authors gave two explanations for this phenomenon: (i) the organic soil microbial community may deplete labile DOC to a greater degree than the conventional microbial community and (ii) the type of organic inputs to the two soils may influence the biodegradability of DOC.

Only a few studies have been conducted regarding the biodegradability of DON. According to the work of Qualls and Haines (1992) on soil solution sampled from various depths of an oak forest, DON does not decay faster than DOC. This is consistent with the hypothesis that hydrolysis of organic N is linked to mineralization of DOC rather than occurring selectively in response to the biochemical need for N. Zsolnay

(1996), on the other hand, reviewed differences in the biodegradability of water-extractable organic nitrogen and carbon of leaves (Ohta et al., 1986) and agricultural soils (Scherer et al., 1992) and concluded that DON was more biodegradable than DOC. It is not possible at present to resolve these contradicting studies.

Isotope studies support the hypothesis that DOM released in the topsoil is subject to further microbial decomposition. Schiff et al. (1990) and Ludwig and Beese (1997) reported decreasing $\delta^{13}C$ ratios of DOM with increasing soil depth. This can be explained by preferential decomposition of compounds with relatively high $\delta^{13}C$ ratios (carbohydrates, amino acids, pectin, hemicellulose) compared with compounds characterized by relatively low $\delta^{13}C$ ratios (lignin, cellulose). DOM, therefore, seems to be fractionated during soil percolation according to its microbial degradability.

In addition to the high variability of the biodegradable portion of DOM, little is known about the microbial stability of DOM once it is adsorbed onto mineral surfaces. A plausible hypothesis is that the stability of adsorbed organic matter increases because interactions between organic matter and mineral phases decrease the ease of degradation of the organic matter (Sollins et al., 1996; Miltner and Zech, 1998). Another possibility is that the accessibility of sorbed organic matter to microbes and enzymes is diminished (Sollins et al., 1996). Neither of these hypothesis has been tested.

It is reasonable to assume that the chemical properties of DOM affect the rate by which it is decomposed by microbes. Hydrophobic acids, which are preferentially adsorbed in mineral soil horizons, are less accessible to microbial decomposition than hydrophilic components (Qualls and Haines, 1992). Therefore, the more labile DOM components should be preferentially decomposed in soil solution. Mousset et al. (1997), however, did not find any correlation between the proportion of hydrophilic DOM components and the biodegradable part of DOM. Nevertheless, correlations have been found between the biodegradable part of DOM and (i) its elemental composition (Sun et al., 1997) as well as (ii) its specific adsorption in UV light (Zoungrana et al., 1998; Gilbert 1988). Boyer and Groffman (1996) found water-extractable humic acids were more bioavailable than water-extractable fulvic acids. These results challenge the generally accepted opinion that humic acids, because they are high molecular weight organics (Tate, 1987), are more refractory than low molecular weight fulvic acids. Piccolo (1998) suggests that humic substances are associated aggregates of small molecules rather than macromolecules. Therefore, a prediction of the bioavailability of DOM on the basis of its molecular weight seems inappropriate. According to Piccolo (1998), the hydrophobicity of organic matter controls its accumulation and rate of decomposition.

In summary, about 10 to 40% of DOM may be easily decomposed by microbes. At present, it is impossible to quantify the relevance of microbial decomposition in DOM retention in mineral soil horizons. Differences between the microbial stability of DOM in solution and that of DOM adsorbed on mineral soil are not quantified. Microbial decomposition of adsorbed DOM is crucial for the renewal of adsorption sites in mineral soil horizons.

CONCLUSION

Throughout the last 20 years (and especially in the last 5 years) there has been a great deal of research regarding DOM dynamics, primarily in forest soils. The most important controls on DOM concentrations and fluxes are summarized in Table 1. Litter and humus are the most important DOM sources in soils. However, it is impossible to quantify either the contribution of each of these sources to DOM release or the effects of increasing amounts of litter and humus on DOM release. We cannot list the properties of soil organic matter that would definitely lead to high DOM release. High microbial activity and the abundance of fungi are positively related to DOM (DOC, DON) concentrations in soil solutions, as are environmental conditions and management practices supporting enhanced mineralization. Certain key properties of soil solutions, such as pH, ionic strength, and phosphate concentration, can influence DOC concentrations greatly. In mineral subsoil horizons, DOC can be strongly adsorbed to oxides and clay minerals, resulting in low DOC concentrations. Preferential flow through fissures and macropores in structured soils can affect DOC output, especially after storm events.

With respect to the objectives of our review, the following conclusions can be drawn:

1. *Biotic vs. abiotic control on DOM concentrations and fluxes in soil horizons of high carbon content:* In theory, DOC concentrations should be mainly biotically controlled, with temperature-dependent seasonal fluctuations and increased concentrations caused by an enhanced mineralization after changes in environmental condi-

TABLE 1

Summary of factors controlling DOM† concentrations and fluxes in soils established from laboratory and field observations

	Laboratory concentration	Field concentration	Field flux	Laboratory concentration	Field concentration	Field flux
	forest floors and soils horizons (A-horizons) of high C content			subsoil horizons of low C content		
Solid phase properties						
Amount of litter		↑	↑			
Organic matter content (amount of humus)	↑	↑	↑	↑	↑	↑
Portion of coniferous forest vs. deciduous forest	⇓	⇑	↑		↑	↑
C/N ratio	↑	→	→			
Microbial activity	↑	↑	→			
Portion of fungi	↑	↑	→			
Fe-, Al-oxides/hydoxides	⇓	↓	↓	⇓	↓	↓
Clay‡	⇓	↓↑	↓↑	⇓	↓↑	↓↑
Solution phase properties						
pH	⇑	→	→	⇑	→	→
Ionic strength	⇓			↓		
Sulfate	→			↑		
Phosphate	⇑			⇑		
Other anions	→			→		
Polyvalent cations	↓			⇓		
Environmental conditions						
Temperature	⇑	⇑	→		→	→
Wet-dry cycles	⇑	⇑	↑			
Water saturation	⇑	⇑	↑			
Water fluxes	→	→	↑		↑↓	↑
Freeze/thaw (snowmelt)	⇑	⇑	⇑		⇑	⇑
N-deposition	→	→	→	→	→	→
Clear cutting		↑	↑		↑	↑
Liming	⇑	⇑	⇑	→	→	→
Organic fertilization	⇑	⇑	⇑	→	→	→

⇑ Strong positive influence in the most studies, ↑ positive influence in many studies, → no clear trend in the reviewed studies (mainly no effect), ⇓ strong negative influence in the most studies, ↓ negative influence in many studies.

†*Mainly DOC (possibly existing differences between DOC, DON and DOP are not included in the table).*

‡Increase in clay content promote preferential flow with high DOM concentrations and fluxes but under conditions without preferential flow—high DOM adsorption.

tions. However, variations in water fluxes through organic soil horizons can mask biotic controls on DOM concentrations. Therefore, hydrological conditions are generally more important than biotic factors for DOM fluxes. Hydrologic controls become more important with increasing time scales, and, on the time scale of several years, water fluxes through the soil are the dominant factor controlling DOM fluxes in the soil. At this time scale, biotic controls are negligible.

2. *Adsorption vs. decomposition of DOM in mineral subsoil horizons:* Adsorption of DOM is the main process determining the retention of DOM in soils. However, there are indications that the microbial degradation of DOM (both in soil solution and that adsorbed on soil minerals) is underestimated in subsoil horizons. This conclusion is based on the following findings:

- up to 40% of released DOM is potentially biodegradable in solutions within a period of days to a few months;
- hydrophilic DOM components with a high bioavailability are preferentially released from the mineral subsoil;
- isotopic fractionation of DOM occurs during soil percolation

It is generally assumed that adsorption diminished the biodegradability of DOM. However, this has not yet been quantified.

3. *Laboratory vs. field studies:* Conclusions about controls on DOM dynamics from laboratory experiments are often not supported in field studies on the scale of experimental plots, catchments, or watersheds or on temporal scales of month to years. The main reason for this discrepancy is that soil hydrological conditions are often not considered in laboratory experiments. Therefore, controls on DOM dynamics identified in the laboratory need to be further studied and quantified in the field. As an extreme example, the effect of species composition (coniferous vs. deciduous) on DOM as reported in the laboratory was opposite that reported for field experiments. Effects of substrate quality (C/N ratio) and pH on DOC concentrations found in the laboratory have not been found in field studies or have not yet been tested in the field. High clay content, which laboratory experiments show should enhance the retention of DOM, may have no effect if preferential flow determines the hydrologic regime in the soil. Many controls tested in the laboratory have also not yet been investigated in the field (Table 1). The biodegradability of DOM, which has been studied only in the laboratory, must also be quantified under field conditions.

DOC versus DON and DOP

Studies on DOM dynamics, which include DON and DOP in addition to DOC, are few. There are strong indications in the literature that the rate of release and fates of DOC, DON, and DOP in the soil may differ to a greater extent than previously assumed. Controls established for DOC might, therefore, not be valid for DON and DOP.

FUTURE RESEARCH NEEDS

Despite intensive research in the last decade and many qualitative results on controls of DOM dynamics (Table 1), quantitative prediction of DOM fluxes at the field scale is still impossible. Because, in practice, DOM dynamics must be predicted under field conditions, the bias between field and laboratory findings concerning controls of DOM dynamics is most critical. Therefore, future research should be focused on comprehensive studies dealing with the relationship between DOM dynamics and environmental factors in the field, with supporting laboratory experiments. The main objectives of such studies should be (i) soil properties and environmental factors controlling DOM quantity and quality as well as sources and sinks of DOM; (ii) the assessment of biological processes vs. physico-chemical controls on the release and retention of DOM in soils; and (iii) differences between DOC, DON, and DOP. In particular, future research should concentrate on:

- Quantification of the various DOM sources under field conditions (recent litter, root-derived DOM [root exudates], microbial biomass, and role of soil fauna, humus)
- Quantification of the effects of substrate quality on *in situ* DOM dynamics in soils, including species effects (e.g., investigating why higher DOC and DON fluxes are observed in coniferous than in deciduous forests despite slower rates of litter decomposition)
- The importance of hydrological conditions for the release and fate of DOM
- Differences in the dynamics of DON and DOP compared with DOC
- Biodegradability of DOM in solution and adsorbed on minerals, including the effects of DOM properties and changing environmental conditions
- Controls on DOM dynamics in soils of different use and in climate zones other than the temperate area
- Developing indicators describing DOM dynamics under field conditions in a simple, holistic approach

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