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Fine-scale temporal characterization of trends in soil water dissolved organic carbon and potential drivers

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ABSTRACT

Long-term monitoring of surface water quality has shown increasing concentrations of dissolved organic carbon (DOC) across a large part of the Northern Hemisphere. Several drivers have been implicated including climate change, land management change, nitrogen and sulphur deposition and CO_2 enrichment. Analysis of stream water data, supported by evidence from laboratory studies, indicates that an effect of declining sulphur deposition on catchment soil chemistry is likely to be the primary mechanism, but there are relatively few long term soil water chemistry records in the UK with which to investigate this, and other, hypotheses directly. In this paper, we assess temporal relationships between soil solution chemistry and parameters that have been argued to regulate DOC production and, using a unique set of co-located measurements of weather and bulk deposition and soil solution chemistry provided by the UK Environmental Change Network and the Intensive Forest Monitoring Level II Network. We used statistical non-linear trend analysis to investigate these relationships at 5 forested and 4 non-forested sites from 1993 to 2011. Most trends in soil solution DOC concentration were found to be non-linear. Significant increases in DOC occurred mostly prior to 2005. The magnitude and sign of the trends was associated qualitatively with changes in acid deposition, the presence/absence of a forest canopy, soil depth and soil properties. The strongest increases in DOC were seen in acidic forest soils and were most clearly linked to declining anthropogenic acid deposition, while DOC trends at some sites with westerly locations appeared to have been influenced by shorter-term hydrological variation. The results indicate that widespread DOC increases in surface waters observed elsewhere, are most likely dominated by enhanced mobilization of DOC in surficial organic horizons, rather than changes in the soil water chemistry of deeper horizons. While trends in DOC concentrations in surface horizons have flattened out in recent years, further increases may be expected as soil chemistry continues to adjust to declining inputs of acidity.

1. Introduction

Reports of increasing concentrations of dissolved organic matter (DOM), as reflected by changes in dissolved organic carbon (DOC), in surface waters over the past three decades (Freeman et al., 2001; Skjelkvale et al., 2001; Evans et al., 2005; Hruska et al., 2009; Couture et al., 2012; Futter et al., 2014; Monteith et al., 2014) have led to concerns both for public health (Ritson et al., 2014) and the fate of terrestrial carbon stocks (Freeman et al., 2001). The DOM trend in upland drinking water sources has exacerbated treatment costs, since most organic carbon has to be removed using expensive coagulation procedures prior to the addition of disinfection agents such as chlorine. Failure to do so can result in excessive production of potentially toxic disinfection bi-products (DPBs such as Trihalomethanes) (Ritson et al., 2014). Separately, the increase in DOC raises various uncertainties with respect to carbon accounting and the extent to which the carbon source/sink status of soils may be changing.

Climate and land use change have both been offered as explanations for rising DOM concentrations and both might imply a long-term destabilisation of terrestrial carbon stocks (Freeman

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et al., 2001). An alternative hypothesis, rapidly gaining acceptance, is that regional scale trends are linked to biogeochemical recovery of soils from anthropogenic acidification (Evans et al., 2006; de Wit et al., 2007; Monteith et al., 2007; Oulehle and Hruška, 2009; Erlandsson et al., 2011). The recovery hypothesis is founded on three elements. The first is evidence that DOM in surface waters, that are not subject to substantial point or diffuse sources of nutrients, tends to have a predominantly allochthonous source (Thurman, 1985). The second is that factors associated with the production of organic matter, and the hydrological forces of DOC transport from soils to waters, have not changed consistently across the area in which the DOC concentrations have changed. The third is the biogeochemical theory that organic matter mobility or solubility will increase in response to reductions in soil water acidity or ionic strength coupled with the observation that there have been very large reductions in sulphur deposition in most regions where DOC concentrations have increased substantially. Furthermore, indications of a recent reduction in the rate of DOC increase in surface waters is consistent with the recovery hypothesis since rates of change in acid deposition have also slowed in recent years (Monteith et al., 2014).

While statistical relationships between the rate of change in acid deposition and DOC concentrations in water, modified by acid sensitivity (Monteith et al., 2007), hint strongly at a controlling effect of atmospheric deposition on catchment soils, there is still no clear consensus regarding underlying mechanisms. The organic soils of upper horizons and near-stream zones are considered to be the main source of DOM in headwater catchments (e.g. Brooks et al., 1999; Billett et al., 2006; Evans et al., 2007; Winterdahl et al., 2011), but considerably fewer long term soil chemistry monitoring records exist in comparison with surface waters, and there are, therefore, relatively limited opportunities to test the various DOC trend driver hypotheses at source. Where monitoring of soil solution has been conducted in regions recovering from acidification, DOC concentrations in organic surficial layers have often been found to be increasing at a similar rate to those observed in surface waters (Borken et al., 2011; Stutter et al., 2011), but DOC trends in deeper horizons have both positive and negative gradients, with increases at some sites (Borken et al., 2011; Stutter et al., 2011; Ukonmaanaho et al., 2014) and decreases at others (Wu et al., 2010; Borken et al., 2011; Löfgren and Zetterberg, 2011; Stutter et al., 2011), possibly reflecting the adsorption/desorption properties of more mineral-dominated layers and differences in redox state. Differences in behaviour between surface waters and some soil waters may therefore result from differences in soil properties, in addition to land use type (e.g. Vanguelova et al., 2010; Borken et al., 2011; Stutter et al., 2011). Interactions with climatic factors also need to be taken into account since investigations into the influence of temperature and hydrology on DOC have demonstrated the potential for regional warming and changes in precipitation patterns to affect DOC production and export (Freeman et al., 2001, 2004; Pastor et al., 2003; Clark et al., 2005). As acid deposition declines to very low levels, and significant changes in temperatures and precipitation are being forecast in response to the continuing accumulation of greenhouse gasses in the atmosphere, it is increasingly important that sensitivity to climatic parameters is quantified accurately.

Attempts to link DOC dynamics in both soils or surface waters and potential environmental drivers have been based predominantly on the use of linear statistics. Yet recent analysis of the first two decades of data from the UK Upland Waters Monitoring Network (Monteith et al., 2014) demonstrates that trends in stream water chemistry have rarely been linear over this timescale. For example, sulphate concentration, a surrogate for sulphur deposition that has been closely linked with recent DOC trends, declined mostly in the latter half of the 1990s, while chloride concentration (primarily a surrogate for sea salt deposition) fell from particularly high levels in the late 1980s to early 1990s and was relatively stable in later years. Since the degree of long-term change in DOC can be orders of magnitude smaller than spatial and seasonal variation (Clark et al., 2010), a substantial component of the potentially valuable diagnostic temporal variation can be lost in the process of reducing time series data to monotonic linear trend components (e.g. Sen Slope) when changes in both driving and response variables are visibly non-linear. Neither parametric linear regression nor non-parametric Mann–Kendal procedures capture short-term, local, variation in the mean in this type of time series data. Effective characterisation of patterns of inter-annual scale variation could, therefore, provide new insights into potential cause-effect relationships that would benefit process understanding.

Consequently, in this paper, we apply non-linear techniques to describe inter-annual scale temporal dynamics in meteorological variables, soil water DOC and other chemical determinants. Data are derived from two methodologically compatible environmental monitoring networks: the Intensive Forest Monitoring Level II (FLII) and the Environmental Change Network (ECN). These are the only networks in the UK that have systematically collated co-located soil and deposition chemistry and meteorological measurements since the mid 1990s. They thus provide unique opportunities to explore linkages between external drivers and the soil profile properties from different soils types, vegetation and land management, and thus allow a clearer understanding of key processes governing DOC production.

2. Methods

2.1. Field sites

Data from five UK FLII and four terrestrial ECN sites across upland and lowland locations were used for this study (Fig. 1). FLII sites were established in 1995 (Vanguelova et al., 2007) and form part of the European forest monitoring network (ICP Forests) that aims to improve understanding of the effects of air pollution and other environmental factors on forest ecosystem structure, function and health. Monitoring of ECN sites started officially in 1993 with the objectives of gathering long-term datasets to improve understanding of the causes and consequences of environmental change across a range of semi-natural and agricultural habitats in the UK (Morecroft et al., 2009; Sier and Monteith, in this issue).

The FLII sites were composed of stands of Oak (*Quercus robur*, *Quercus petraea*), Scots pine (*Pinus sylvestris*) and Sitka spruce (*Picea sitchensis*) under standard forest management practices, including thinning and brashing during their growth cycle. The plots varied in planting year between 1920 and 1974 and cover a range of forest production classes. The ECN terrestrial sites represented non-forest environments, i.e. upland grassland or moorland vegetation, subject to seasonal grazing, mainly by sheep. Site characteristics are provided in Table 1. The sites ranged in altitude from 20 m to 690 m above sea level. Mean annual temperature (MAT) (for period 2002–2006) varied from 6.1 °C at Moor House up to 11.6 °C at Thetford; and mean annual precipitation (MAP) (for the period 2002–2006) from 605 mm at Thetford to 3420 mm at Snowdon.

Soils at all FLII and ECN sites were surveyed between 1993 and 1995. In each plot the soil was described according to the FAO soil classification system and classified according to the World Reference base for soil classification (WRB, 2007). FLII were also classified using the Soil Classification for England and Wales (Avery, 1980) and the Scottish Soil Classification system (Soil Survey of Scotland Staff 1981). FLII soil sampling and analyses were carried out according to the UNECE ICP Manual for Soil Sampling and Analysis (2006). ECN sites surveys were conducted according to The UK Environmental Change Network Protocols for Standard Measurements at

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