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Interactive comment on "Trends in soil solution dissolved organic carbon (DOC) concentrations across European forests" *by* M. Camino-Serrano et al.

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Referee 2: However, saying that this article is not very clear is litotes. The abstract and introduction are rather well written. However, the result and discussion sections are extremely hard to read, the number of figures and tables is not tenable (I counted 27 items, figures+tables) and the usefulness of numerous statistical analyses is not convincing since they provide similar results and conclusions. I think that an effort of synthesis is necessary to simplify messages and prevent a dilution of important results with accessory observations. For example, at the end of the reading of your manuscript, I was not able to say whether sulphate depositions increase or decrease soil solution DOC content. In your abstract, it is suggested that DOC concentrations

and sulphate depositions are positively linked, a statement which is then contradicted in the manuscript (e.g. L412-413 but L348-349).

We agree that the manuscript includes many statistical approaches and we understand that this might confuse the reader. This approach diversity comes from the large community of scientists involved in the study by providing data and scientific input. Concerning the temporal analyses of DOC concentrations, we decided to show results from the different statistical methods (LMM, seasonal and partial Mann Kendall) because the approaches are complementary. Each method has pros and cons. This allowed us to show that DOC concentrations have increased during the observation period overall in coniferous forests in the organic layers. However, at individual plots and depths, DOC concentrations did not show any consistent temporal trend (increase, decrease or no change). We could also show that there was no geographical pattern either.

Concerning the multivariate analyses, we propose to shorten the manuscript by removing three figures (Fig 8, Fig. 9, Fig. 10), parts of Figure 7 (7A, 7B), and one table (Table 4). Fig. 9 will be moved to Supplementary Material. This would not change the conclusions of the manuscript. In Figure 7, we would focus on the relationships between the temporal trends of DOC concentrations, forest types and classes of stem volume increment (proxy for forest productivity). The relationships between trends in DOC concentrations and SO4 and NO3 throughfall deposition would be explored in Figure 11.

Referee 2: Moreover, it would be useful to create a figure summarizing the main chemical reactions and biological processes controlling soil solution DOC content

We could include such a figure in the introduction based on the numerous mechanistic models proposed in the literature. Our results come from an exploratory statistical approach (and not deterministic) of a large European dataset and it would be preposterous at this stage to build a model based on such a variety of local (e.g. soil properties) and regional (e.g. atmospheric deposition) factors.

Referee 2: More fundamentally, I am not sure that the measurement of soil solution DOC provides an accurate estimate of the amount of DOC flowing out of terrestrial ecosystems (and supply of DOC to surface water). The leaching of DOC happens at specific moments of the year depending on hydric balance (precipitationevapotranspiration), soil type, plant activity etc. I am even sure that the DOC soil solution concentration can be inversely related to DOC leaching in some conditions. Just an example: soil solution DOC concentration is higher in summer than in winter, but DOC leaching only occurs in winter in France. This issue could explain why the present study fails to show clear overall trend in soil solution DOC at individual plot and soil depths. A warming-induced change of ecosystem water balance could also contribute to changes in DOC content in soil solution and surface water. Therefore, I suggest to present (in manuscript or in supplementary materials) the volume of water harvested in lysimeters or calculations of theoretical water balance (precipication-evapotranspiration).

We agree with the referee that we did not assess DOC fluxes flowing out from forest soils. Our time series analysis aimed to detect long-term changes of DOC concentrations that are not due to seasonal effects or dilution-concentration effects caused by fluctuations in soil water content. Therefore, we decided to apply both Seasonal Mann Kendall and Partial Mann Kendall using precipitation as a co-variable to remove the seasonality and dilution-concentration effects. This method allowed us to detect significant monotonic changes (increase or decrease) of DOC concentrations over a period of 10 years at least. Studies having shown temporal changes of DOC in surface waters also reported concentrations rather than fluxes (e.g. Worral & Burt 2004, Evans et al. 2005, Monteith et al. 2007, Dawson et al. 2009, all cited in the manuscript).

Using water volume collected by lysimeters to assess water fluxes was not possible, since these data were available since 2011 only. In addition, the volume collected by tension lysimetry depends on the suction applied to the system. To assess water fluxes at different soil depths, we would need to model the water balance at 118 for-

est sites, which is very challenging, since many input parameters (meteorology, soil, vegetation) would be required. This was beyond the scope of this study. A simple estimate based on the difference between precipitation and evapotranspiration, would add a substantial uncertainty to the calculation of DOC fluxes and therefore detecting long-term changes of DOC fluxes would be even more difficult. Because of large variations in soil water fluxes (e.g. Borken et al. 2011, Verstraeten et al. 2014, Meesenburg et al. 2016), it is more difficult to detect long-term trends of fluxes than long-term trends of concentrations in soil solution. Since most times series of DOC concentrations in soil solution do not indicate any long-term trend in our dataset, the chance of finding long-term changes in DOC fluxes are even lower.

References

- Borken, W., Ahrens, B., Schulz, C., & Zimmermann, L. (2011). SiteâĂŘtoâĂŘsite variability and temporal trends of DOC concentrations and fluxes in temperate forest soils. Global change biology, 17(7), 2428-2443.

- Meesenburg, H., Ahrends, B., Fleck, S., Wagner, M., Fortmann, H., Scheler, B., ... & Meiwes, K. J. (2016). Long-term changes of ecosystem services at Solling, Germany: Recovery from acidification, but increasing nitrogen saturation?. Ecological Indicators, 65, 103–112

- Verstraeten, A., De Vos, B., Neirynck, J., Roskams, P., & Hens, M. (2014). Impact of air-borne or canopy-derived dissolved organic carbon (DOC) on forest soil solution DOC in Flanders, Belgium. Atmospheric Environment, 83, 155-165

Referee 2: I am not really convinced by the relevance of removing the breakpoints. These breakpoints are not necessary the result of site disturbances (change of sensors etc) but could result from sudden change of atmospheric chemical composition or ecosystem functionning.

Monotonicity of time series is generally assumed when analyzing DOC data for temporal trends (Filella and Rodriguez-Murillo, 2014). However, it is rarely statistically tested and, thus, potential abrupt changes in the time series may be overlooked. This issue becomes important in temporal trend analysis since a breakpoint may cause changes in the direction of the trend and could lead us, for example, to classify a time series as constant, when in reality we may have averaged out separate periods with significant changes (de Jong et al., 2013). On the other hand, breakpoints may erroneously induce the detection of a significant trend in long-term time series due to artifacts (see Supplementary Material).The aim of our study is to analyze monotonic trends related to factors that have been measured within the ICP Forests database. Therefore, DOC time series were first analyzed using the Breaks For Additive Seasonal and Trend (BFAST) algorithm to detect the presence of breakpoints.

We agree that removing breakpoints using the BFAST technique may remove time series that show abrupt changes not only due to artifacts (collector replacement, etc.,), but also due to natural causes (meteorological conditions, extreme events), forest management (changes in soil condition, thinning, etc.), sudden change of atmospheric chemical composition or ecosystem functionning. Nevertheless, many breakpoints are the consequence of technical issues or even inconsistencies in the database. The ICP Forests soil solution dataset has a great potential for analysis of large scale trends, but at the same time it may also contains some inconsistencies. The BFAST analysis proved to be effective at removing breakpoints caused by some dataset errors and thus the most defective time series were left out.

Although the investigation of the potential causes of the abrupt changes (breakpoints) in the individual time series can indeed provide a very valuable information, we do not count with the site-level information necessary for that purpose. To attribute the different causes of the breakpoints at site scale, we would need information of the management and climate history at each particular site, which is not available at the time of writing the present manuscript. Consequently, we cannot be sure of the origin of each breakpoint, and thus we decided to leave out all the time series showing abrupt

changes to avoid erroneous detections of significant trends. Moreover, in this way, we are confident that the trends found in the time series that we analyzed are not a consequence of local factors. The alternative is to study each time series individually to identify the local (or regional) factors causing abrupt changes at plot scale, and this task is beyond the scope of this study, but is a very interesting topic to be addressed in a follow-up paper.

References

- Filella, M. and Rodriguez-Murillo, J. C.: Long-term Trends of Organic Carbon Concentrations in Freshwaters: Strengths and Weaknesses of Existing Evidence, Water-Sui, 6, 1360-1418, 2014.

- de Jong, R., Verbesselt, J., Zeileis, A., and Schaepman, M. E.: Shifts in Global Vegetation Activity Trends, Remote Sens-Basel, 5, 1117-1133, 2013.

Referee 2: The terminology used in the manuscript is often not clear. The term “trend” is vague and does not specifically refer to change with time. The terms “trend slope”, “trend direction” and “relative trend slope” are even more difficult to understand.

To avoid any confusion about the term “trend”, we could add a small paragraph in the Method section explaining the different temporal components of time series analyses. Time series can be typically decomposed into random noise, seasonal and trend components (e.g De Livera et al., 2011).

We also suggest to explain the meaning of “trend slope”, “trend direction” and “relative trend slope” in the Method section. However we would prefer to keep the same terms in the whole manuscript. They frequently appear and we would need much more words if we had to explain them in each occurrence.

Reference

- De Livera, A. M., Hyndman, R. J., & Snyder, R. D. (2011). Forecasting time series with complex seasonal patterns using exponential smoothing. Journal of the American

Statistical Association, 106(496), 1513-1527.

Referee 2: The terms“depositions” and “troughfall” are interchangeably used, I suggest you to use only one of the two terms.

We used throughfall deposition of sulfate and organic nitrogen, except in Table 3 where bulk deposition was provided for three sites because throughfall deposition was not available. This was marked by an asterisk in the table. To avoid any confusion, we will use only throughfall deposition in the text.

Referee 2: The term “fertile soil” is weak and, as usual, does not refer to measurable variable. The fact that tree growth is high does not necessary mean that the soil is fertile. The tree growth is often linked to forest dynamics and age (tree growth of old forests is typically slow irrespective of soil characteristics; tree growth after forest disturbance (drought events, storm etc) is typically high because tree mortality allows the recruitment of seedling with fast growth rate).

We agree with the referee that tree growth is not necessarily related to soil fertility. We suggest to reformulate chapter 4.2.1. This chapter aims to discuss the relationship between forest productivity (in our case only stem growth is available) and DOC in soil solution. A number of factors such as climate, soil water availability, soil fertility, tree age and competition between neighboring trees can influence tree growth.

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