

We thank John Van Stan and the two anonymous referees for their comments and useful suggestions, which helped us to rework our manuscript.

Response to referees:

John Van Stan

General comments

We extended the conceptualisation of OM biogeochemical processes in tree canopies by additionally elucidating the importance of the cortisphere processes on OM transformation and quality. We also added additional site information on forest structure metrics. With regard to the length of the sampling period, we added a reference addressing this issue.

Specific comments

0) The abstract is not detailed enough and most of the writing past line 7 is unclear. For example, is the TOM versus DOM comparison in lines 9-12 for all samples regardless of species?

→ For clarity, we reworked the abstract completely from line 7 onwards:

The ^{13}C NMR results, derived from 21 samples, point to pronounced differences in the composition of DOM and TOM in throughfall solution at the beech sites with TOM exhibiting higher relative intensities for the alkyl C region, representing aliphatic C from less decomposed organic material compared to DOM. Furthermore, TOM is composed of lower intensities for lignin-derived and aromatic C of the aryl C region resulting in lower aromaticity indices and a diminished degree of humification. Along the ecosystem compartments, differences in the structural composition of DOM and TOM under beech lessened in the order throughfall > stemflow \approx forest floor leachate. In contrast to the broadleaved sites, differences between DOM vs. TOM in throughfall solution under spruce were only less pronounced and spectra were overall dominated by the alkyl C region, representing aliphatic C. Explanations to the reported results might be substantiated in differences in tree species-specific structural effects, leaching characteristics or differences in the microbial community of the tree species' phyllosphere and cortisphere. However, the fact that throughfall DOM under beech showed the highest intensities of recalcitrant aromatic and phenolic C among all samples analysed, likely points to a high allelopathic potential of beech trees negatively affecting other organisms and hence ecosystems processes and functions."

What is meant by "fresh" POM in line 8?

→ since this wording appeared too unspecific, we left that out while rewording the abstract from line 7 onwards (s. above)

What is meant by a "tree species-related effect on the origin of OM composition [and] properties" – tree structural effect, tree-specific leaching characteristics, tree phyllosphere and cortisphere microbial community differences?

→ We reworked the abstract and some parts of the introduction, hoping to clarify the results/explanations (s. above and below).

More information is needed in the abstract, like:

(a) what is the list of OM characteristics and metrics derived from the ^{13}C NMR spec (see Table 1 and Table 2)?

→ We added more information on those characteristic spectroscopic measures.

(b) How many total samples were analyzed?

→ This information is given P15093L1 of the Discussion paper. However, we changed the sentence to: “In total, 21 samples were analyzed by solid state ¹³C NMR spectroscopy. Table 3 provides an overview over the sample consortium.” We also added this information in the abstract.

(c) Which results indicated species-specific throughfall, stemflow, or litter leachates may differ in allelopathic potential (see discussion P15096L27-&)?

→ Among all samples analysed, throughfall DOM under beech showed the highest intensities of aromatic and phenolic C (Table 4). Compared to throughfall DOM under spruce, the values for phenolic C were nearly twice as high and the ones for aromatic C by a factor of 1.5 to 1.7 times higher. Phenolic C forms part of polyphenols, secondary metabolites, being known for their allelopathic potential (Rice, 1984, Muscolo et al., 2001 and Hane et al., 2003). However, the striking high phenolic C signal in beech throughfall is just one suggestion to partly explain why for example the spreading of beech in Europe after the last glacial era was competitively that effective and persistent (Petraakis et al., 2011). Nevertheless, up to date, we are lacking of comparable measurements to assess the allelopathic potential of throughfall DOM composition of different tree species to prove our suggestion.

We reworked the entire “Results and discussion section” addressing that issue.

1) An overarching hypothesis formed regarding differences in TOM and TOM structural transformations between the two species (P15090L8-11) has two issues in its current form:

(a) it focuses solely on how leaf structural differences for *F. sylvatica* versus *P. abies* can alter OM vis-à-vis microbial communities and substrate interaction in the phyllosphere: this neglects the fact that these species also have quite different bark structures and branching architectures hosting microbial communities and substrates capable of interacting during the throughfall or stemflow process to alter OM.

(b) it neglects to mention that interspecific differences in phyllosphere and cortisphere microbial community structure/function may also play a critical role in altering TOM travelling through the canopy along hydrologic flow paths.

→ Thank you for these suggestions. We added a new section on the effects of differences in bark traits on OM composition:

“Interspecific differences in bark traits and cortisphere properties, e.g. bark texture and colour, influence the microclimate and arthropod fauna (Nicolai, 1986) the plant epiphyte (Wyse and Burns, 2011) and microbial community (Andrews and Harris, 2000) consequently affecting the amount and composition of OM as well. For example, enhanced bark microrelief increases the contact time of stemflow solutions with bark surfaces leading to an enrichment of aromatic hydrocarbons (Levia et al., 2012) likely due to the enhanced degradation and release of soluble lignin compounds (Guggenberger et al. 1994).” (P3 L65)

2) A key introductory component supporting the need for this research is that POM might be particularly relevant to other nutrient cycles (specifically through microbial decomposition processes [P15089L11]). But, the authors don't cite any studies in support of this statement, nor do they provide explicit explanation of how POM's C:N ratios (or myriad other microbially-attractive qualities) support this statement. Please make this connection more explicit and substantiated by citations as it is a piece of the foundation supporting “why” your work is necessary.

→ To support the “why” for our work we included additional facts on the amount of extra incoming OM by the POM fraction and the impact of the C-to-N ratio of organic matter on degradation processes. Therefore we changed the introduction part as follows:

“Lamersdorf and Blank (1995) revealed a significant additional input of organic carbon and nitrogen to the soil with throughfall (TF) via the particulate fraction (> 0.45 µm), amounting to 11–15% for C and 14–21% for N relative to the total annual input by litterfall and TF (< 0.45 µm), respectively. These results are corroborated by recent findings for POM and DOM in TF of beech, spruce and pine

forest stands (le Mellec and Michalzik, 2008; le Mellec et al., 2009, 2010), where the additional input of C and N by particulate organic matter represented between 20–30% of the total organic C and 10–20% of the total N fluxes with throughfall. The low C-to-N ratios of 12–14 for POM as reported by Lamersdorf and Blank (1995) furthermore suggest that this material might be particularly relevant to promote microbial decomposition processes (Fontaine et al., 2004), while wider C-to-N ratios of > 30 tend to decelerate decomposition rates (Pérez-Harguindeguy et al. 2000).” (P2 L43)

3) Citation issues in introduction:

P15088L22: the statement “only a few have investigated the dynamics” is lacking necessary citations – please cite which few studies have done so.

→ We added the lacking citations (Carlisle et al., 1966; Sollins et al., 1980; le Mellec et al., 2009, 2010).

P15089L16-18: the statement “The chemical nature of mobile OM is : :” currently lacks citation. Please provide.

→ we added Guggenberger and Zech (1994) and the fundamental work on humic substances by Schnitzer and Khan (1972) and Wardle et al., (1998) addressing other ecosystem processes induced by altered DOM composition

P15089L28: The citation Levia et al., 2012 is incorrectly cited, as the authors’ statement discusses cation cycling and the citation does not investigate cation cycling – only DOM. If the authors wish to stick to that research group’s work for a citation, a more appropriate reference would be Van Stan et al. (2012, The effects of phenoseason and storm characteristics on throughfall solute wash-off: : , Sci. Tot. Environ., 430: 48-58) for throughfall ion work, or Levia et al. (2011, Atmospheric deposition and corresponding variability of stemflow chemistry: : , Atmos. Environ., 45: 3046-3054) for stemflow ion work.

→ Thanks a lot for this clarification. We decided to add both references on ion cycling as suggested by the reviewer as well as Gersper and Holowaychuk (1971) and Levia et al. (2012) addressing changes in DOM.

4) Study site description is lacking some details that are necessary, in my opinion, for contextualizing/interpreting results (even though a previous site description exists in Fischer et al. [2010]). Specifically, the authors lack a comparison of stand structure for the plots to substantiate that the interspecific structural differences described in **P15090L8-11** are present. Although a basic description of the *F. sylvatica* plots is supplied (P15091L13-17), the *P. abies* plantations are not described: : : are they also similarly-aged or -sized compared to the *F. sylvatica* plots? How do stand characteristics (density, basal area, height, dbh, etc) between *P. abies* and *F. sylvatica* compare?

→ In an additional Table 2, we compiled stand structure data presented by Schwarz et al. (2014) and reworked the paragraph from P15091 L 15 onwards:

“These forests are comparable with regard to tree age and tree species composition and are similar with regard to stand density, mean diameter at breast height, basal area and forest management (Table 2). In the Hainich-Dün exploratory we additionally collected samples from three Norway spruce (*Picea abies* (L.) H. Karst.) plantations (Table 2). We only choose spruce sites of the Hainich-Dün site, because the Schorfheide-Chorin exploratory only provides pine (*Pinus sylvestris* L.) stands and the spruce stands of the Schwäbische Alb differ in tree species composition (Schwarz et al., 2014). (P4 L112)

Why were *P. abies* plots only chosen at one observatory when, according to Table 1, there were plots available in the Schwäbische Alb site? Doesn’t the sole selection of *P. abies* sites from Hainich-Duen diminish the authors’ aim for comparison of these species across different environmental conditions? I believe these questions could be answered (briefly) to the benefit of the manuscript while still leaving some details to be found in Fischer et al. (2010).

→ We add explanations to the text (see added text above).

5) Can the authors please provide some further details regarding their methods:

P15092L10-12: What were the sample collection procedures, more specifically? E.g., what type of bottles were used to collect/store the water? How long were samples permitted to remain in the field after a storm (this has a big impact on what DOM character will be observed)? Were bottles acid washed and triple-rinsed with sample prior to collection?

→ For the synchronisation of the field sampling campaigns in all three study regions performed by three different working groups, we stick to a fixed routine sampling interval of 14 days.

Concerning the impact of the time on sample composition, a field incubation study by Michalzik et al. (1997) reporting on the chemical composition of bulk and throughfall solutions, exhibited, that the throughfall composition (concentrations of N species and DOC) exposed during August was rather stable over the first two weeks of field incubation before DOC concentrations started to drop. However, since we are not aware of any study testing temporal changes of the structural character of DOM and especially TOM during field exposure, it is speculative and therefore difficult to comment. The bottles were acid washed and triple-rinsed with sample prior to collection.

P15092L19: Are the “cooling boxes” simply refrigerators? Regardless, at what temperature were the samples stored?

→ The samples were stored on ice in a cool box until further processing in the laboratory. (P5 L141)

6) Results: Figure 1 was absent: : : Not sure if it is something I did during the download or if Figure 1 is really just not there: : : So, I took the authors at their word during descriptions of the ¹³C NMR spectra.

→ Figure 1 was not absent (Please compare with the downloaded pdf file of the Discussion paper, Fig. 1 is found on the last page P15107)

7) Discussion:

(a) The introduction links TOM structure in canopy-derived hydrologic fluxes to canopy-based microbial communities, yet the discussion generally avoids this topic. Is this because no bulk precipitation samples could be analyzed via ¹³C NMR spec?

→ Your suggestion is partly true, since we were technically not capable to perform ¹³C NMR analysis to the bulk precipitation, we therefore do not know the DOM/TOM composition of atmospherically deposited onto the canopy and what is “internally” within the canopy produced or transformed.

However, we restructured the Results and Discussions section partly addressing this issue.

(b) Depending on how long samples were allowed to sit in the field, could the authors please provide a discussion of how this would affect the character of their DOM and POM measurements?

→ Please compare with our answer given under 5)

(c) P15096L3-6: Please discuss why the aromatic C-region intensities for FF leachates of Sanderman et al. (2008)’s mixed redwood stand were so much higher than observed for your Spruce sites.

→ In the paper by Sandermann et al. (2008) it is not clearly stated when the solutions for solid state ¹³C NMR spectroscopy were sampled. We assume that it is a composite sample collected throughout the rainy season (representing most of the annual rainfall of 1,450 mm) covering a period from Dec 2005-May 06 (see p. 185, Soil water sampling and Fig. 4), while our samples were collected over 14 days in August/September. According to the results of Kaiser et al. (2001) on “Seasonal variations in the chemical composition of dissolved organic matter in organic forest floor layer leachates of old-growth Scots pine (*Pinus sylvestris* L.) and European beech (*Fagus sylvatica* L.) stands in northeastern Bavaria, Germany”, diverting sampling period and season might partly explain the differences in DOM composition in concert with differences in soil properties and tree species composition (Redwood/Douglas fir), and especially with differences in climate and hydrology (Mediterranean vs. Temperate). In this context, enhanced hydrological flux generation (e.g. throughout rainy seasons)

was shown to increase the aromaticity of DOM by the intensified degradation of soluble lignin compounds as observed by Guggenberger et al. (1994) and Levia et al. (2012). Given that background information we shortly addressed that question in the reworked results and discussion part (P9 L260)

Editorial comments:

Abstract, P15088L7-8: The phrase “echoed in structural differences” is unclear, please revise. The verb “echo” literally means “repeated” or “reverberated”, but can more symbolically mean “shared characteristics with”. How does *F. sylvatica* leaf surface-derived POM “repeat” or “have shared characteristics with” total organic matter (TOM) structure differences? **The abstract was completely reworded leaving this irritating phrase out.**

Moreover, TOM differences in what (species? landscape? source)? **The reworked abstract is hopefully more specific answering that question. (P1 L7)**

Introduction, P15088L22: Please replace “but” in the statement “but none the character of water-bound TOM: :” with “and”. **We replaced “but” by “and” (see also below).** Also, please insert a verb into the statement between “none” and “the”. Perhaps the statement could read “and none investigated the character of water-bound TOM: :”? **We changed the sentence as follows: [] ..., only a few investigated the dynamics of particulate OM (Carlisle et al., 1966; Sollins et al., 1980; Le Mellec et al., 2010) and none the dynamics of water-bound total OM (TOM) including the particulate OM fraction (POM; 0.45 µm<POM<500 µm). (P2 L33)**

Introduction, P15090L12: In the statement “fresh beech leaves exhibit” – do you mean to say “fresh beech leaves leach”?

→ We reworked the whole section:

“In this context, beech trees potentially exhibit the highest amounts of phenolics in leaves (Bussotti et al., 1998), bark (Dübeler et al., 1997), wood (Mämmelä, 2001) and roots (Weissen and Praag, 1991) compared to co-occurring species. Phenolic compounds released from living or dead plant material have been shown to function as allelochemicals, affecting associated species (Rice, 1984; Wardle et al., 1998). An interspecific allelopathic potential on other organisms may result in net changes in ecosystem processes and functions (e.g. herbivory, decomposition and nutrient mineralization) (Wardle et al., 1998). However, the release of phenolic or other allelopathic compounds from living plant material via throughfall and stemflow and the effects on ecosystem processes and function is still insufficiently understood.” (P3 L77)

Methods, P15092L134:

Two issues: The verb tense “were” should be “was”. And, the terms “count” and “terminated” could be replaced with clearer terms, like “release” and “over” respectively.

→ We rephrased the sentence: “... developed, external inferring events such as the release of pollen by coniferous trees was over (mid May to mid June) and...”(P5 L136)

Results, P15095L5-6: The statement “as of SF samples in general” is unclear. Do you mean that there are no studies reporting on stemflow ¹³C NMR spec-derived TOM characteristics and metrics? Please clarify.

→ Your interpretation is right and for clarification we reworked the statement:

“The discussion generally lacks of comparable data on TOM composition. To the best of our knowledge, there are no studies available reporting on solid state ¹³C NMR spectroscopy-derived TOM characteristics in TF, SF and FF leachate samples under beech or other tree species.” (P9 L267)

Anonymous referee # 2

General comments

GC 1 I would have appreciated a list of abbreviations to help navigating the text, such as BP, FF; ALB, TF, HAI etc.

→ We're now providing a list of abbreviations.

GC 2 P 15090/L07-16 contain interesting hypotheses and I implicitly assumed they would be re-visited in the discussion or conclusion, but there they are not addressed explicitly. Can you extend on these?

→ We followed your suggestions.

GC 3 In Fig. 1 - Please add a line, marking zero, to the graph of the differences between filtered and unfiltered samples. It is difficult to make out where the difference is positive or negative.

→ We added the lines in Fig. 1.

GC 4 Fig 1, Table 2: The first lines in Section 3.2 say that differences in DOM and TOM diminished from TF to SF and FF. I understood this as implying an order, that is $TF > SF > FF$, and refers again to the differences in filtered and unfiltered samples shown in Fig. 1 and Table 2? The following sentences and also the first paragraph in the conclusions seem to further develop this suggestion. But in Fig 1 and also Table 2 the (absolute) differences between filtered and unfiltered samples seem to go in a different order: $TF > FF > SF$. This needs to be clarified.

→ That is a tricky question and the order between SF and FF varies with the chemical shift regions being of interest and with the ratios (of aromaticity or of alkyl C/O-alkyl C) derived from them. If we only look at the chemical shift regions (now Table 3) then your impression is right. If we look at the aromaticity % or the alkyl C/O-alkylC ratio for SF and FF at the Schorfheide site (now Table 4), then differences appear more pronounced for the SF composition. However, since the order between compositional differences between SF and FF is less definite, we changed the order to $TF > SF \approx FF$. (P9 L268)

GC 5 Results vs. discussion: Much of section 3.1 reads like a results section, while 3.2 reads like a discussion section. In 3.1 some literature is cited for comparison, i.e. whether the observed differences were to the same direction in other studies, but there is no discussion of the potential mechanisms. Only one paragraph includes discussion on mechanisms, but only for a particular item (P 15095 L7-20), that is the possibly influence of insect infestation on the results.

I propose transforming section 3.1 into a results section (while I think it is ok to keep the references to the literature for comparing the measurements) and section 3.2 into an interpretation section (or simply "discussion"). For this, move the paragraph P 15095 L7-20 into (now) section 3.2. If you go with this suggestion, mechanisms would only be discussed in section 3.2 and the heading could be adjusted accordingly to help orientation for the reader. The current name of section 3.2 is misleading as "differences in structural C composition" are also a topic in section 3.1.

→ We understand your concerns and therefore we merged section 3.1 with 3.2, add subtitles for each ecosystem compartment, and insert discussions on mechanisms potentially driving the results and differences we observed. However, for the sake of clarity while presenting numerous NMR data, cited literature for comparison and potential mechanisms, we stick to a combined "Results and discussion" section.

Detailed comments

DC 1 P15089-L13: Sentence should start better with "Although"

→ We changed it to "although". (P3 L53)

DC 2 P15089-L26: Better "Previous work" or "Previous studies"

→ We changed it to “Previous work”. (P3 L72)

DC 3 P15089-L26-28: I find this sentence far fetched to motivate the need to study the influence of tree species on properties of DOM. In fact it would read better if the sentence was erased.

→ According to reviewer #1 we reworded this sentence adding some references as well:

“Previous work has demonstrated that tree species and spatial and temporal patterns of environmental conditions such as chemical soil properties influence the input and cycling of cations (Gersper and Holowaychuk, 1971; Finzi et al., 1998; Levia et al., 2011; Van Stan et al., 2012) and DOM in temperate forests (Levia et al., 2012).

(P3 L72)

DC 4 P 15094-L3: I believe it is meant “were being” instead of “were be” or simply erase “be”.

→ We corrected it to “were assigned”. (P6 L173)

Anonymous referee # 3

General comments

The authors have submitted a potentially interesting ms about water-transported organic compounds in, among others, throughfall, stemflow, and forest floor leachates.

To this purpose they have used three sites across Germany, each having stands with two tree species, namely common beech and Norway spruce.

→ That is not correct. There are no Norway spruce stands in the Schorfheide-Chorin exploratory. However, we address this issue in the last paragraph of the Introduction as well as in the Methods section 2.1 Study sites. (P5 L116)

The authors have not given any clearly stated aim but mention in the Introduction that this is the first study to evaluate the relative sizes and the chemical composition of the main flows of dissolved organic matter and organic matter in suspension.

→ This is not correct. In the original Introduction we did not mention that this “is the first study to evaluate the relative sizes and the chemical composition...”. In the revised version we reworked the Introduction as suggested by the other two reviewers, now clearly stating our aims: “Therefore, we present the first study investigating the composition of DOM compared to TOM by solid state ¹³C NMR spectroscopy.

We tested the hypotheses that (i) the structural composition of DOM in filtered and TOM in unfiltered forest ecosystem water samples differ along ecosystem compartments and (ii) between tree species, exemplarily tested between beech and spruce at one of our study regions.” (P4 L98)

Thus, in the Introduction they give a very good motive for the study.

They also present two hypotheses namely that the chemical composition differ between total organic matter and dissolved organic matter as well as between the two tree species common beech and Norway spruce. In conclusion, the study is potentially valuable and can give new information.

When it comes to presenting their study in a manuscript the authors have not done a very good job (see also below). The main lines they sketch in the Introduction dissolve in the first section in Results.

In fact, the present ms gives me the idea of a project report. I would recommend that the paper is restructured (please see below). The aim(s) need to be clearly stated and a linguistic revision is needed. I would recommend that the authors should be encouraged to resubmit their paper.

→ To sharpen our aims, we restructured the Introduction and the Results and Discussion section. The paper was already revised by native speakers.

My advice would be to present an ms that gives focus onto one site and a comparison between spruce and beech, alternatively to use just the beech data for three sites. Either of these alternatives may give the impression of a finished study.

Specific comments

The authors describe their sites and the measurements carried out at them. Still, when studying tables 2 and 3, figure 1, and the para beginning on line 24 on page 15092 it appears that the authors have not enough data to make a good comparison of the two tree species. With spruce missing from two out of three sites there is no good possibility to develop thoughts about hypothesis (ii).

→ We like to emphasize that every throughfall (TF) sample analysed by ^{13}C NMR consists of a pooled sample made up of 60 individual TF solutions collected by 60 throughfall samplers at each exploratory (see P5 L139 in the Methods section). Even though we included only the three spruce sites at the Hainich-Dün exploratory, tree-specific properties are spatially well represented and the comparison of beech vs. spruce provides a good first insight into the interspecific differences in DOM and TOM composition.

“Individual samples of BP (3 sites x 5 replicates = 15), TF (3 sites x 20 replicates = 60), SF (3 sites x 3 replicates = 9) and FF leachates (3 sites x 3 replicates = 9), were pooled to one volume-weighted sample per sample type, sampling date and exploratory.”

→ In the Methods section 2.1 Study sites, we give reasons why we were only able to sample the spruce sites at Hainich-Dün exploratory. “In the Hainich-Dün (HAI) exploratory we additionally collected samples from three Norway spruce (*Picea abies* (L.) H. Karst.) plantations (Table 2). We only choose spruce sites of the Hainich-Dün exploratory, because the Schorfheide-Chorin exploratory (SCH) only provides pine (*Pinus sylvestris* L.) stands and the spruce stands of the Schwäbische Alb exploratory (ALB) differ in tree species composition (Schwarz et al., 2014).” (P4 L114)

→ Additionally, we clarified our hypothesis with regard to the tree species comparison (see above).

With the background presented in the Introduction I would expect that Results and Discussion starts with the main lines, for example amounts of bulk precipitation, amounts of stemflow as well as of DOM and POM plus forest floor leachates in both types of stands, thus data giving an overview. What I see is a detailed description of C-NMR spectra, which per se is all right as a subsection but which would fit in a bit later in the paper.

→ It was not the objective of this study to present flux data of DOM and TOM, but data on the chemical structure and properties of DOM compared to TOM. This was clearly stated in the title of the ms, in the first line of the abstract as well as in the introduction (P 15091 L 1). Since it was not possible to analyse bulk precipitation by solid state ^{13}C NMR spectroscopy (please see P 15092 L26), we focussed on throughfall, stemflow and forest floor leachate. However, based on the comments and useful suggestions of the other two reviewers we reworked the Abstract and Introduction to further clarify the aims and motivation of our study.

Further, this section (3.1) is intended to give the properties of the compounds identified but in part it is not clear to me what is data for spruce and what is beech. The text is not clearer as regards what refers to TOM and to DOM (hypothesis i) as the authors use the terminology 'filtered' and 'unfiltered'. Earlier, in Methods and Introduction they define the fractions by particle size (less than

and bigger than 45 µm). It would be better and clearer to the reader to keep to the once defined terminology.

→ Although the first paragraph of the Introduction provides an overview over the terminology, we changed the wording of hypothesis i):

We tested the hypotheses that (i) the structural composition of DOM in filtered and TOM in unfiltered forest ecosystem water samples differ along ecosystem compartments and (ii) between tree species, exemplarily tested between beech and spruce at one of our study regions.” (P4 L98)

The authors write in the hypothesis para (last para in te Introduction) that they have an hypothesis about chemical recalcitrance and allelopathic potential. That hypothesis is not even approached in the text, except for some comments to references.

→ We understand your concerns and therefore we present additional information in the Introduction as well as in the Results and Discussion part.

The number of abbreviations and acronyms is a bit high and some terms are introduced which are not really necessary. Please remove such ones as PE for polyethylene and PU for polyurethane – they are not really used. In line 24 (last line) on p 15089 the list of 'DOM and POM in TF, SF and FF' is a bit hard on the reader.

→ Since “PE” for polyethylene is used five times within this paragraph we like to stick to the abbreviations.

Additionally, we will provide a list of abbreviations as suggested by Reviewer #2.

Page 15090 lines 4 thru 6. The authors mention two species and draw a conclusion about 'coniferous' vs 'broadleaved' ?

→ This part is shifted to the Results and Discussion section. There we clarified the terms by exchanging 'coniferous' by spruce and 'broadleaved' by beech. (P7 L216)

Details

Key words are missing.

→ We're now providing key words.

Abstract. Use the full names of the two species ideally with the Latin name after. E.g. common beech (*Fagus sylvatica*). Further, often chemical compounds are introduced. Thus '....carbon (C).

→ We followed your suggestion.

Line 2. Define each term separately – thus total OM (TOM), consisting of DOM and particulate OM (POM)....

→ We reworked the abstract and defined each term separately.

Introduction.

Line 18. Introduce carbon (C) and nitrogen (N) before you use the chemical denominations.

→ We followed your suggestion.

Page 15089, line 10. Normally written C-to-N ratios

→ We changed to C-to-N ratios.

Same page line 15. What is OM in solution(?) – it seems to be in some kind of contrasts to DOM in the line above? Or do you simply mean OM suspended in water?

→ For clarification we changed it to “TOM in solutions”. (P3 L54)

Page 15089 lines 19 thru 25. Split up that long sentence

→ We reworded the sentence to:

“In this context, precipitation patterns (Mercier and Lindow, 2000) and canopy architecture (Levia and Frost, 2006; Levia et al., 2012) basically alter the release of OM with throughfall. Atmospheric deposition (e.g. pollen, inorganic N deposition, Lamersdorf and Blank, 1995) and increased nutrient leaching from damaged leaves and immobilization processes within the canopy mediated by phyllosphere microorganisms (Guggenberger and Zech, 1994; Stadler and Mueller, 2000) appear to influence the chemical qualities of OM from the forest canopy.” (P3 L77)

Page 15090, line 7 the percentages 30-40% and 17-20%. Percent of what?

→ Percent of “rainfall interception rate values”

“Additionally, spruce canopies exhibit higher rainfall interception rate values of 30-40 % compared with 17-20 % for beech canopies (Rothe et al., 2002).” (P7 L218)

Same page line 9 'time'

→ We corrected it to “time”. (P7 L220)

Methods

Section 2.1 The sentence starting with 'The three regions.....' Fits better at the end of the para

→ We reworked this paragraph (see above), ending with the reasons why we were only able to sample the spruce sites at Hainich-Dün exploratory. (P5 L116)

Section 2.2.

First line; 'exploratory'? That word is an adjective in English. If the authors mean 'site' please write that.

→ It is more than a “site”. It is a region/ landscape in which several forest sites are located. The concept of the “Biodiversity Exploratories” as a scientific platform for biodiversity research is elucidated in the paper by Fischer et al. (2010) and received international recognition. Overall, we are instructed by the steering committee of the “Biodiversity Exploratories” to use the term “exploratory” in a general sense, like e.g. “observatory”. We therefore like to keep this word.

Set up should be setup.

→ We corrected it.

Line 2. Five replicates from each of the three open areas?

→ That is correct.

Line 4 '...collectors of the 'same type'... same type as what?

→ We used the same open funnel samplers (diameter 0.12 m) for collecting throughfall (TF) as well as bulk precipitation (BP). We collected BP in 5 replicates and TF in 20 replicates.

“... bulk precipitation (BP) was collected in five replicates from three open areas using 2 L polyethylene (PE) open funnel samplers (diameter 0.12 m). TF was sampled using rain collectors of the same type...”

Section 2.3

First para, first lines. Introduce the abbreviations ALB, HAI and SCH in the text. The same para, line 18 (3x3=9), respectively... why respectively?

→ We left the word “respectively” out.

Line 21. 'bottled'? Does that mean that you took an unfiltered sample? Third para (p 15093) line 20.

→ That is correct. We took an unfiltered solution to analyse TOM by solid state ^{13}C NMR spectroscopy.

Sensitive? Do you mean 'sensitivity'

→ We mean the adjective "sensitive" and not the noun.

Results and Discussion

First para (p 15094) line 3 delete 'be'

→ We corrected it to "were assigned". (P6 L173)

Line 9 'differed remarkably' - from what?

→ "Unfiltered and filtered TF samples from the beech sites differed remarkably with regard to C composition." (P6 L181)

Section 3.2, 2nd para, line 19 and 20. 'Enzyme activity' ?? It is important for such a discussion to make clear what enzymes that were inhibited.

→ We followed your suggestion. (P7 L194)

Conclusions

Last line '....research has to be extended spatially and to different tree species.' That phrasing is a bit surprising for a conclusion in this paper. Having read the Introduction and Methods I would expect that to be what this paper deals with. Three regions across Germany and two tree species.

→ Beech and spruce forests are the most common forest types in Germany and they are also representative for large regions in Central Europe. They provide important ecosystem services (e.g. nutrient and carbon cycling, biodiversity), which are worth to study. Since information on the structural composition of DOM and TOM in forest ecosystem water samples is generally scarce, we conducted this first orientating study including the three regions (or Exploratories) across Germany. That is why we also state that "...research has to be extended spatially and to different tree species."

Table 3 and in other places, e.g. Fig 1. 'unfiltered' is abbreviated nf? Why not uf?

→ We changed the abbreviations to "fil." for filtered and "unf." for unfiltered.