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Interactive comment on “Patterns of dissolved organic carbon (DOC) and nitrogen (DON) fluxes in deciduous and coniferous forests under historic high nitrogen deposition” by S. Sleutel et al.

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In our response below, the line numbers refer to a revised manuscript version that has been prepared in the meantime.

1. General comments

Referees #1 and #2 ask about the novel aspects in this study. Indeed DOC and DON leaching have been studied relatively frequent in forest ecosystems where atmospheric N deposition is high. However, and as we also indicated in the original paper, there are a number of reasons why this paper has particular value compared to other papers that have dealt with this topic. First, fluxes of matter, including DOC and DON, have

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been were measured at a large number of different strata, while many papers deal with fluxes (or often only concentrations) below the litter layer, and/or at the bottom of the soil profile, i.e. one or two ecosystem strata. Second, water fluxes (and associated solute fluxes, including DOC and DON) have been calculated very accurately using detailed soil physical data and a mechanistic water balance model, while many other studies use much simplified calculations of water and solute fluxes. Third, the impact of increased N deposition has been studied often in laboratory or field manipulation experiments, with mineral N fertilizer added to soil artificially. Because of the limited time scale of many of these experiments, a new equilibrium situation often will not be reached. In this research we could study the effect of additional N loading over the long term by including the forest edge treatment. The results of the comparison between deciduous and coniferous forests are discussed in the dedicated section 4.2.

2. Specific comments per referee

2.1. Referee #1

Title: abbreviations (DOC and DON) have been removed from the title. Abstract: Belgium has been added (line 7) Change matter to element: we think matter is a more correct term here, since we look at fluxes of compounds and not individual elements

Site description: more detail has been added and further clarifications have been made as requested by this referee, as follows: number of soil samples taken for the chemical analyses in Table 2: this information has been added (line 118) Thickness of the organic layers has been added (lines 130-131). The amount of organic C and N in the litter layer can be readily calculated from the data given in Table 3 (total dry matter and C contents of the forest floor layers given in Table 3). For the calculation of total C and N stocks in the mineral soil, measurements of bulk density are needed. We have made such measurements in 2005 for typical podsol profiles in the Ravels forest reserve, which yielded values of 1.41, 1.55, 1.15, 1.19, 1.33 and 1.44 for the A, E, Bh, Bs, BC and C horizons respectively. However, especially the measurements of bulk density in

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the Bh and Bs horizons are problematic because they are very thin layers. We have not included these bulk density measurements in the manuscript, because we think they are not essential. Still, total C stocks in the entire soil profile (down to 90 cm) have been calculated for these locations and were 134, 167 and 148 tons ha⁻¹ for the SB, CP and CPN respectively. We have not included these stocks because we think they are not necessary for the interpretation of results in this paper.

The amount of litterfall presented in Table 3 indeed is aboveground litterfall, and the way how we measured this has been added to the Materials and Methods section (lines 143-148).

Statistical analyses:

We agree that variance analysis was the correct choice for the statistical analysis, and we have now re-analysed the data accordingly. This resulted in slightly different statistical conclusions, namely the DIN and DON throughfall fluxes in SB and CP were no longer significant, while the DIN and DON throughfall fluxes in CPN were still significantly different from SB and CP. These changes have been made accordingly in the revised text.

Results:

Page 7143, l 17-19: last sentence was removed because there is discussion on this further in the paper

Page 7143, 21-24: this sentence was removed

Page 7144, 17-18: r values have now been added in this sentence (line 297)

Page 7145, 5-8: has been removed because because there is discussion on this further in the paper (section 4.2.)

Page 7145 last sentence – page 7146 first sentence: has been removed

Discussion:

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Differences in oxalate extractable Fe and Al concentrations and clay content: we do not agree that there were differences in clay content, the differences in Table 2 are very small and certainly within the analytical error of clay determination by the pipette method. There were indeed differences in oxalate extractable Fe and Al between the forest stands. When recalculating Fe_{ox} and Al_{ox} concentrations on a molar basis, and weighing molar concentrations with depth of the B + B/C horizons (assuming an overall bulk density of 1.5), SB tended to have a larger adsorptive capacity (58.6 mol Fe_{ox}+Al_{ox} m⁻²) compared to CP and CPN (36.3 and 37.3 mol Fe_{ox}+Al_{ox} m⁻², respectively). However, DOC retention in the mineral soil (which can be considered mainly as sorption) was 201, 185 and 265 kg DOC ha⁻¹ yr⁻¹ in SB, CP and CPN, respectively, and this does not relate directly to Fe_{ox} and Al_{ox} concentrations. Vandenbruwane et al. (2007) indeed found that while the DOC sorption capacity in the Bh horizon was 80-100 % saturated, it was only from 39-45 % and 16-18% saturated in the Bs and BC horizons, respectively. Apart from effective sorption to the mineral particles, DOC may also be retained by precipitation of metal-DOC complexes. These points are now also mentioned in the text (lines 500-512).

Actually, for the litter layer in these three forest stands, we have made measurements of both litter quality (only chemical quality namely C/N, P, K, Ca, Mg, Na content, not biochemical quality) and of microbial biomass and composition of the microbial community by PLFA analysis. Litter quality in terms of elemental composition was higher for SB than for CP and CPN, with significant differences in total Ca and Mg content between SB and CP and total P content between SB and CPN (CP and CPN had comparable litter quality). However, measurements of biochemical litter quality (watersoluble C and N, lignin, polyphenols) and litter mineralization would be needed to discuss the influence on DOC and DON leaching, and these measurements were not available. Microbial biomass C and N did not differ significantly between the forest stands. The composition of the microbial community as measured by PLFA fingerprinting revealed only minor differences between the forest stands, namely significantly lower actinomycetes biomarkers in CPN compared to CP and SB, and significantly

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lower mycorrhizal biomarkers in CP and CPN compared to SB. A thorough discussion about the potential influence of these differences would require more in-depth measurements under controlled conditions, which was not within the scope of this study.

Denitrification will not have a significant role in the forest ecosystem that we studied. Our study was limited to fluxes leaving the soil profile at a depth of 65 cm. The highest groundwater level at these sites is deeper than 1 m below the soil surface. Moreover, these are very sandy soils (around 95% 50-2000 μm). Denitrification is therefore unlikely in the soil profile that we studied. It may be important for nitrate removal once it has moved into the saturated zone, but this will again depend on the reduction capacity of this zone (which will be mainly determined by the content of Fe bearing minerals such as pyrite), and this was not the scope of our study. In the paper of Tietema & Verstraten (1991), groundwater levels were periodically very high, explaining the importance of denitrification (in their research this was remarkably mainly N_2O).

Page 7152, 25-26: We have reformulated this observation accordingly (lines 525-526)

Figure captions

Abbreviations of SB, CP and CPN have been explained

Fluxes in the figures are mean fluxes and this has now been mentioned in the caption

Referee #2

General comments:

First general comment: see above

DON leaching only 2-5 kg N ha⁻¹ yr⁻¹: We think that this comment is already addressed in the original manuscript. We agree that other authors have found larger DON leaching losses from forest soils, and that some authors found increasing DON losses with increasing N deposition loads. We also acknowledge this fact in the original manuscript (see e.g. Introduction, page 7136). However, many studies looked only at leaching of

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DON from the forest floor, while we analysed fluxes leaving the soil profile at 65 cm. Pregitzer et al. 2004 and Fang et al. 2009 (mentioned in the manuscript) found a large increase in DON leaching out of the mineral soil in N addition experiments or under large ambient N deposition. However, other authors found no increase in DON leaching in N addition experiments (e.g. Currie et al. 1996). Our results show only a small increase in DON leaching when N deposition was higher. The comparison with the unpolluted forests was based on catchment measurements rather than measurements at the bottom of the soil profile, and therefore we have removed the sentence on the unpolluted forests from the manuscript.

Necessity of appendix: The appendix is not essential for understanding and interpreting the results of this paper, but we still think that it has sufficient value to keep it in. The parameters that have been measured and calibrated can not often be found in literature and we think that others may be very interested in comparing their model parameter values with values that were obtained here. But again, if the editor also finds this appendix to be superfluous, it can be removed.

Specific comments:

1) Title and abstract: Belgium has been added in the Abstract 2) Page 7136: these indeed report about the same experiment, and we have now removed McDowell et al. 1998 because they only reported on leaching below the litter layer, which is less relevant in the point we want to make. 3) Page 7136 line 23: has been changed accordingly 4) Page 7138 line 7: has been changed accordingly

Referee #3

Comments about experimental design and statistical evaluation:

Indeed what we have done is replicate within “treatments” (within individual stands). This indeed may be considered as pseudo-replication, but the reason for this choice obviously was the difficulty in finding additional forest stands of birch and pine stands

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on the same soil type, and with the same deposition and similar deposition gradient. We acknowledge this now more explicitly in the revised manuscript (lines 152-155, lines 273-276). While the use of replicates within “treatments” is not optimal, it is a choice that is often made because of practical reasons, as explained above.

Page 7135 lines 14-16: this has been reformulated to stress that forest floor is the main source of DOM

Page 7142 lines 12-13: we agree and refer to the answer to the comments of referee 1.

Page 7144 lines 2-4: the statement about relation was not very precise, what we meant was that DOC concentrations were influenced by weather conditions during the sampled fortnight and the preceding fortnights, and this has been reformulated accordingly (line 282-284).

Page 7146 lines 22-23: the DON/DIN ratio in the bulk precipitation in our study was around 1/2, i.e. a DON/TN ratio of around 1/3, which is not an exceptional value. The organic N in bulk deposition is of a completely different nature compared with DON in soil. Studies show that while the percentage contribution of organic N to total N loading varies from site to site and with measurement methodology, it is consistently around a third of the total N load, with a median value of 30% (Neff et al. 2002), which matches the DON/TN in bulk deposition measured here.

Report C pools of O horizon in Table 2: these are reported already in Table 3 dedicated to the forest floor layer.

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