bg-2023-38

We thank the associate editor for considering our manuscript, the comments of the reviewers and our answers to their comments. Below are our responses in blue, with the original editor's comments in black. We recall our answers to the reviewers (AC-1, AC-2) further down.

Thank you very much for uploading your detailed responses to the comments by the two anonymous reviewers.

The evaluation and comments by the reviewers were positive and it seems like you are willing to address these comments on a revised version of your manuscript.

I am particularly interested in seeing how you addressed the following important issues:

Reviewer #2 made a very pertinent comment on what would be the impact of temporal autocorrelations among trends of pH over time in different compartments of the hydrological system. This analysis is particularly relevant to your study given its long-term and large-spatial scale.

Beside the cross-correlations between precipitation pH and runoff water pH mentioned in AC-2, we now performed also auto-correlations of these time series. Without real surprise, they mainly showed an annual cycle, which justifies including the seasonality in our trend analyses by regressions.

In addition, reviewer #1 raises important questions regarding the impact of N deposition on the ecosystem. Again, given the long-term nature of this study I believe that it is important to address how this process could have influenced your results.

Indeed, the question of the impact of N deposition is addressed throughout the manuscript. In the revised version, we no longer only concentrate on the acidifying effect. We recall shortly previous findings about effects of N as a nutrient as seen in this experiment. This is important for the readers in order to better understand the context of the original results presented here.

AC-1

We thank Reviewer 1 for carefully reading and commenting our manuscript. Below are our responses in blue, with the original reviewers' comments in black.

The current manuscript addresses relevant scientific questions within the scope of the journal: testing the effects of increased N deposition on acidification processes in a well-buffered ecosystem. This is a concept not enough explore so far, which gives high scientific relevance to the paper. The hypothesis are clear, well described, and fully addressed through methods and assumptions, that are clearly explained.

We appreciate the reviewer's recognition of our manuscript's contribution to ecological research.

- line 14-15, Although it is known from previous studies, the text here it is not clear enough about the N addition. Please clarify if N treatment is 22 kgN greater than the ambient N deposition (C treatment), that was 12 kgN at the beginning of the study, or if it represented 22 kgN compared to 12 kgN of ambient deposition.

Indeed, these are 22 kg as treatment on top of 12 kg of ambient deposition. The abstract will be improved to make it clear that 22 kg is the additional input.

- line 139-145: It could be also clarified a little.

Especially the time course was not clear, with one-year of measurements preceding the start of the N treatment. The text will be amended to explain what we mean with this calibration period.

Deposition. *Although N deposition and cycling is not key for the study, some further data a discussion on this issue could improve the understanding or, at least, allow further interpretation of the results. Particularly, the nitrate concentration response in runoff could be related to N cycling dynamics that can be connected to saturation processes. Knowing that N leaching is addressed in previous studies, not much information would needed here, but some of it would be appreciated. Particularly, a historic evolution of N deposition comparing both treatments would be appreciated. Moreover, in this study, throughfall deposition (if available) would be of greater relevance than bulk deposition since it represents a direct input into the soil.*

It is certainly a good idea to recall here the main results obtained about nitrate leaching. In this regard, we can distinguish three main periods over the course of our experiment: (1) immediate response due to preferential water flow through the soil, (2) progressive N saturation and (3) several years of increased leaching after girdling, then felling part of the trees. These results are already published in more detail but will shortly be recalled when we discuss nitrate leaching in the present contribution.

As it also partly represents dry deposition, throughfall is indeed a valuable information. It is already given in the material and methods and will be added to the introduction and also to the abstract.

- line 15, line 92, line 130: the deposition value referred here is from the beginning of the study

N deposition did not change much over the course of the experiment, but it is indeed better to write explicitly that the given numbers are from the beginning of the experiment.

- line 130: total deposition and N retention in the catchments are also available from previous studies. The authors might consider if these data could be relevant here.

This point is also justified and can be answered in the same context as the 3 previous ones.

- lines 273-280: deposition data from other studies are referred here. Since it is stated that precipitation has not significantly changed in the valley, the changes in N concentration in rain are correctly assumed to be reflected in deposition values. However, some data on measured deposition trends would be appreciate here.

Indeed, even if the trend in precipitation in not statistically significant, it still slightly modifies the trend of the fluxes compared to the concentrations. We will include these trends in Tab. S1.

Nitrate leaching. *lines 297-299: This is just a question. It is stated that "NO_3^- can have an acidifying effect if leached together with base cations, leading to their replacement by H^+ or Al^{3+} on the exchange complexes". This effect is observed in N treatment in the increase of NO_3 leaching, that is related to the strong increase in soil acidity on the mounds. Should it be also related to an increased leaching of base cations that matches the NO_3 leaching but that cannot be observed in the current graphics?*

Ca^{2+} is the dominant cation in runoff water. It can be seen (Fig. 6) that Ca^{2+} concentrations decrease more in the control catchment than in the N-treated catchment. Additional NO_3^- leaching from the treated catchment can partly (only partly) explain this difference in Ca^{2+} trends. A short indication about this will be added here in the discussion.

Figure 1. *I understand that trenches in catchment 1 are present in the same way as in catchment 2 (around the entire perimeter) but it is not drawn in the picture.*

On its upper part, catchment 1 (control) is delimited by a natural water divide. As the contour lines drawn on the graph are barely visible, a short explanation will be added to the legend of the figure.

Please consider the following suggestions. *(Detailed comments not repeated here)*

All these comments are useful and will be taken into account. Only the 4th one (about decomposition) does not contain any particular action to be taken. The sentence was thus just considered with a particular attention within the overall language editing of the whole manuscript.

bg-2023-38-AC2

We thank Reviewer 2 for carefully reading and commenting our manuscript. Below are our responses in blue, with the original reviewers' comments in black.

The paper by Baer et al. monitored changes in rainfall, soil, and runoff chemistry (pH, acidity and ions) in the preAlps during more than 20 years during which actions were taken to abate atmospheric pollution in Europe. They also added N to plots to simulate an increase in N deposition. This is an important contribution to understanding the long-term effects of increased N deposition in forest catchments, as well as of the importance of abatements measurements. As such, the current study is a highly valuable contribution to understanding patterns of N deposition and its consequences for the biogeochemistry of European forests.

We much appreciate the overall positive assessment of the reviewer.

On the negative aspect, more care should have been taken to avoid incomplete sentences, colloquial expressions, and grammatical errors and typos. Thus, the paper should be carefully revised in this aspect.

The manuscript will be given a thorough revision to correct for grammatical errors, typos, redundancies and incomplete sentences while avoiding and replacing colloquial expressions. The text will also be revised by a third party (non author) competent in both English language and scientific writing.

Moreover, some questions remain untested. For example, one of the most intriguing results is the increase in the pH of rainfall across time, indicative of reduced acid rain due to political action, but the continuous decrease of soil pH and pH in runoff water across the same time period.

It is indeed not an obvious but an important result. In the discussion we consider the cause-to-effect relationships and their timing. It is especially important to distinguish between fluxes and buffering mechanisms. We could add here in a very short manner: it is not because there are less acidifying inputs that there is no longer any acidification. The discussion will be made more explicit in this regard.

I wonder whether there is any temporal correlation among these variables. Or in other words, are shifting pH values across time in rainfall, soil and runoff coupled or decoupled? Are these couplings/decouplings maintained across soil horizons and experimental treatments? The same can be asked for other variables like total acidity, etc. Analyzing these types of questions would, in my opinion, increase the novelty of the paper beyond what could be considered as a very informative and highly valuable report by providing more novel insights.

As shown by our results, precipitation pH, soil pH, soil exchangeable acidity and runoff pH have each a different time-course over the duration of the experiment. At a multi-year time-scale, they are clearly decoupled. Nevertheless, we gladly retain the reviewer's question to examine whether pH measurements in precipitation and in runoff water are correlated in the shorter term. After excluding the cold-season data that may be affected by a snowpack and after removing the long-term trends, we did not find any significant short-term correlation. This is not really surprising if we consider the large buffering capacity of the soil. We will add this new aspect to the revised manuscript. In terms of soil pH and exchangeable acidity, we have too few time points to try any such statistical approach and have to stay with the contrasting long-term trends of these time-series.

Other than that, I consider this paper as a very valuable contribution to the field. (Detailed comments not repeated here)

Most of these suggestions will be directly implemented. It is true that chemical equations are not commonly shown in ecological research articles. Nevertheless, we consider them useful because readers in this field often don't remember exactly what these reactions are and which (quantitative) consequences they have.

The sentence on lines 168-171 is about the replicated plot design. It is always a bit difficult to explain that we used two statistical designs for a single experiment: a paired-catchment design (with the advantage of being able to measure export fluxes in runoff water) and a replicated block design (with the advantage of the replications). We believe that everything will be more understandable by adding here a reference to Fig. 1, on which both designs are visible.