Reply to interactive comment on "Imbalanced phosphorus and nitrogen deposition in China's forests" by Anonymous Referee #1

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This is a potentially publishable article about an interesting topic (N:P ratios in deposition in China), but there are a few points that need modification and/or justification before the paper can be accepted.

Reply: Thank you. We have revised the manuscript according to your suggestions.

Major point: How can you deduce dry deposition from throughfall and precipitation concentrations? This needs to be better explained or justified, and the following points are where I get confused.

Reply: Before going into the detailed questions we like to give a general overview of our assumptions, which we have now also included in the manuscript. Canopy-captured dry deposition accumulates during periods without precipitation and it is washed down during precipitation events. Here we calculated canopy-captured dry deposition as the difference between bulk deposition and the estimated total deposition. In our original manuscript, total deposition was estimated based on throughfall concentrations and total (bulk) precipitation but in the revised manuscript we now multiply it with the sum of throughfall and stemflow water fluxes. To estimated the sum water flux of throughfall and stemflow, we establish a new database of observed throughfall and stemflow (including data from 28 forested stands from 26 sites, see Figure R1 as below) and explored an empirical equation between precipitation and the sum of throughfall and stemflow (TS =0.88×precipitation-64.6, R^2=0.97). The sum of

throughfall and stemflow water fluxes for each forested site was then estimated from the available bulk precipitation data based on this empirical equation. We realize that our estimate of canopy-captured dry deposition is lower than total dry deposition because a proportion of dry deposition is already included in bulk deposition and our estimate of total deposition is likely underestimated due to the neglect of canopy uptake (Reddy and Majmudar, 1983; Draaijers et al., 1996; Sparks, 2009) and stemflow enrichment (N & P concentrations in stemflow are generally higher than that in throughfall). We now mention this explicitly in the discussion and we will include detailed information on the method and uncertainty in the method section in the revised manuscript.

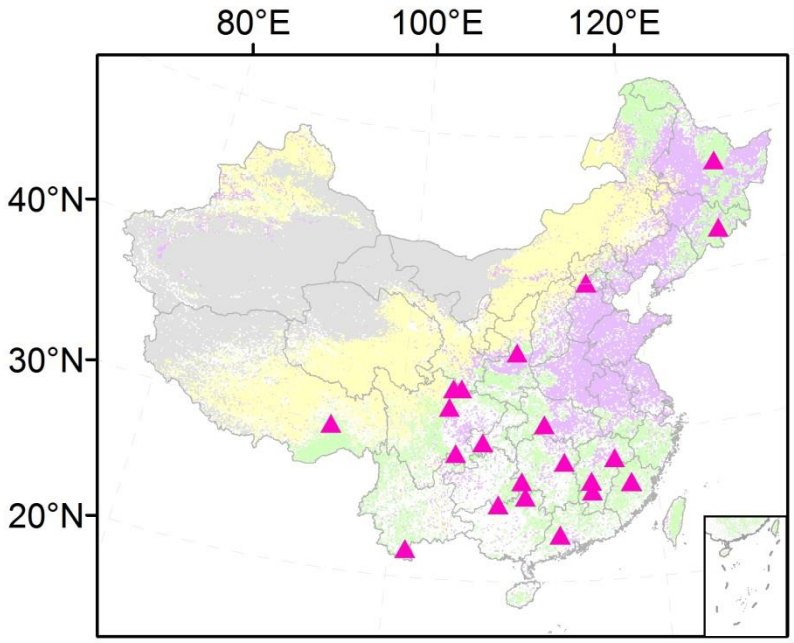

Figure R1. Distribution of 26 sites with measured data of throughfall and stemflow.

"Phosphorus and N fluxes in the bulk precipitation and throughfall were estimated according to the volume-weighted mean concentration and annual precipitation." So you are taking your measured concentrations in each precip and throughfall and multiplying by annual precip? But not all the precip comes out in throughfall does it?

Reply: This is a correct comment. Indeed not all precipitation comes out in throughfall and we now do not use total (bulk) precipitation any more but the sum of throughfall and stemflow water fluxes estimate (see above). On an annual basis, total deposition can be estimated as the sum of N/P fluxes in throughfall and stemflow as well as canopy exchange (see Figure R2 as below). Due to a lack of measured data on canopy exchange and stemflow

concentrations, we approximately estimated total deposition by multiplying the volume-weighted mean N/P concentration in throughfall with the sum of throughfall and stemflow water fluxes in the revised manuscript. Our results generally underestimate total deposition because concentrations in throughfall usually are much lower than those in stemflow and tree foliage can take up tree foliage can take up a small proportion of soluble N and P in rainwater and gaseous N (part of dry deposition) (Reddy and Majmudar, 1983; Draaijers et al., 1996; Sparks, 2009). More detailed information on the method and its uncertainty will be included in the revised manuscript.

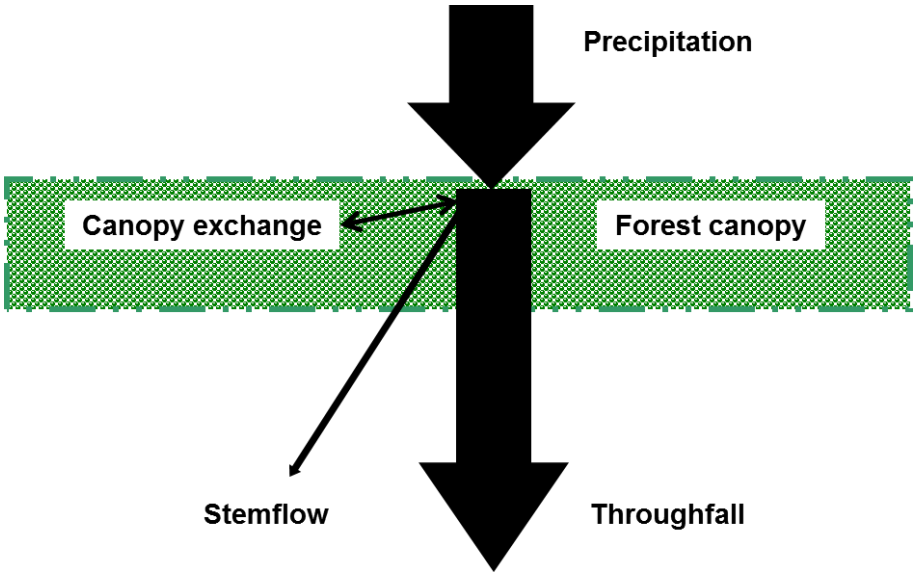

Figure R2. Partition of nitrogen or phosphrus flows when rainwater passes through forest canopy.

Canopy captured dry deposition was estimated as the difference between throughfall deposition and bulk deposition." How can you get dry deposition from the difference in two wet deposition numbers? Later on you try to use this: "Based on the P enrichment in throughfall versus bulk precipitation, the mean canopy captured dry P deposition was estimated as high as 0.46 kg P ha-1 yr-1 in China's forests, being significantly higher than bulk deposition (0.38 kg P ha-1 yr-1) (paired t-test, df = 32, $p < 0.01$)." So I think you need to be a bit clearer: maybe make a skematic of your assumptions?

Reply: Canopy-captured dry deposition is defined as the amounts of dry deposition captured and accumulated in forest canopy during the period without precipitation events. Therefore, we calculated the canopy-captured dry deposition as the difference between bulk deposition and the estimated total deposition (as described above). The new result indicate that mean canopy captured dry P deposition was estimated to be 0.40 kg P ha^{-1} yr^{-1} in China's

forests, being comparable to bulk deposition (0.38 kg P ha^{-1} yr^{-1}) (paired t-test, df = 32, p = 0.36). We will include more detailed information on the method in the method section and discussed uncertainties of our results in the revised manuscript.

Again you say something here that doesn't make sense to me: "Although throughfall may still underestimate total deposition due to canopy uptake (Draaijers et al., 1996; Sparks, 2009), it is a better proxy of total N deposition than bulk deposition because bulk deposition only accounts for 63% of throughfall deposition."

Reply: The last part of this sentence may have caused confusion but our point is that even though throughfall may underestimate total deposition it is a better proxy of total N deposition than bulk deposition that makes a much larger underestimation. We now changed the sentence to: "Although our estimate based on annual sum of throughfall and stemflow water fluxes and total N concentrations in throughfall is still lower than the factual total deposition because tree foliage can take up a small proportion of gaseous N (part of dry deposition) and soluble N in rainwater (Draaijers et al., 1996; Sparks, 2009) and the stemflow N flux is likely underestimated, it is a better proxy of total N deposition than bulk deposition." In addition, the uncertainties of our estimates are also discussed in the revised Discussion section.

Are you only concerned with soluble P?

Reply: Our analysis focused on total P concentrations, which were measured after digestion and include dissolved inorganic forms, dissolved organic forms and particulate forms. This is mentioned in the text of the method section.

It is important to clarify these points. If you are going to argue that from two wet deposition numbers you can deduce dry deposition and total deposition, then maybe you want to verify at a point where you have all three numbers to show that your approach works? Your paper could be publishable with only wet deposition numbers, but I think you just need to be clear about what your results are.

Reply: Thank you for your suggestions. More detailed information on the method and uncertainty has been included in the revised manuscript (also see replies above).

Secondary points: In the comparison to the model results there are two issues:

i. "Anthropogenic sources have been traditionally thought to make only a small contribution to P deposition (Okin et al., 2004; Mahowald et al., 2008). In contrast, our results emphasize that anthropogenic sources near large cities

can have significant impacts on the spatial patterns of regional P deposition. The urban hotpot of P deposition might be derived from intensive combustion-related emissions near urban areas (Wang et al., 2015) and a short-distance transfer of P-containing aerosols from P-rich farmland soils (Anderson et al., 2006).” The problem with this conclusion is that it contradicts the observations here that the highest P deposition comes from regions close to the arid sources of dust: so your gradient suggests the dominant sources are dust related. Even using the smaller anthropogenic source of Mahowald et al., 2008, anthropogenic sources should dominate near cities in China (Figure 5h: Mahowald et al., 2008). So I would actually argue that your data supports Mahowald et al., 2008’s budgets quite well. In contrast, from Wang et al., 2015, you shouldn’t be seeing a P deposition gradient across China (Figure 3) from the arid regions, but only the combustion hot spots. You could actually be a bit quantitative and do the comparison against the model output from these two models, to discriminate between them? See if there is a statistically significant difference in the comparison?

Reply: Our results show significantly higher P deposition nearby than far from semiarid regions and this indeed supports Mahowald et al., 2008's conclusions that large-scale gradients of P deposition are dust dominated. At regional scale, either far from or nearby the semiarid regions, our results indicate that P deposition showed a significant power-law increase with closer distance to the nearest large cities and this supports Wang et al., 2015's conclusion that anthropogenic contribution to P deposition can be substantial. Overall, our results don't contradict the conclusions either by Mahowald et al.(2008) or Wang et al. (2015). Moreover, the modelling results usually have a very coarse resolution and are not suitable for quantitative comparison with our datasets. We have revised the text to make this point more clear.

ii. Both the model of Mahowald et al., 2008 and Wang et al., 2015 only include long range transported P, so less than 10um or 20um particles. Your results, in contrast, should be including all P (unless you are only looking at soluble P, which should then not be compared to Wang et al., 2015 and only the soluble P in Mahowald et al., 2008). In forests, most of the P is in much larger mode particles, locally generated, according to previous observational synthesis (and the observational studies within them; e.g. Tipping et al., 2014). So it is likely you have a mismatch in the P deposition you are comparing (see Mahowald et al., 2008; Neff et 2009; Brahney et al., 2015 for more discussion on how to compare observational P fluxes across sizes to models or not). Notice that Wang et al., 2015 is very careful about the size of their emitted combustion P particles, but not at all careful about the size of the particles in the deposition P dataset they use (Tipping et al., 2014; which is dominated by large local generated primary biogenic particles, according to that study), compared to the particles that they are modeling (long range

transported particles <20um). They actually use the mismatch in particle size and the resulting mismatch in deposition magnitude to deduce the extra-large combustion source they postulate, which makes that study problematic in its conclusions.

Reply: In the discussion section, we mostly compared our results with other assessments based on observations (e.g. Tipping et al., 2014, Vet et al., 2014). We have already recognized that different studies may focus on different components of P and therefore we are very careful when making comparison with other results.

Minor points:

Section 3.1: I am not sure what I am supposed to get out of knowing that the data follows a log-normal distribution for P or N or N:P. Isn't this what we expect? I think this would be better illustrated with a figure of the distribution, if you think it's important enough to discuss?

Reply: The description of datasets (e.g. arithmetic mean for a normal distribution and geometric mean for a log-normal distribution) should be according to their distribution. Therefore, we tested the distribution of the datasets at the beginning.

"Spatial patterns of P deposition via bulk precipitation and throughfall showed large heterogeneity" This is a factor of 4. Globally, the deposition varies by several orders of magnitude, so I'm not sure I would call this a large spatial hetereogeneity. Maybe just say the values vary by a factor of 4.

Reply: We have done so.

Some English errors, especially in the few last lines of the introduction, should be fixed.

Reply: We have checked and corrected all the linguistics errors in the manuscript.

References:

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