nature portfolio

Peer Review File

Fire-derived phosphorus fertilization of African tropical forests

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

This manuscript presents convincing evidence that canopy complexity of forests of Central Africa plays a key role in the amount of phosphorus stored by these ecosystems. The age of the forest that determines tree structure and the canopy geometry acts as a trap for phosphorus for aged forests. The manuscript is clearly written, the arguments are convincing and I have only some minor points that could be clarified to improve the clarity of the points being made. I recommend that this manuscript be published with these improvements.

Major point: I could not find in the main text or in the Supplement how the ranges for the deposition fluxes were calculated and what uncertainties/variations were accounted for.

Minor points:

Line 81: The measurements you made do not cover whole of Central Africa, refer the reader to Figure S1 that presents the measuring site and shows the extent of the area covered by these measurements.

Line 84: The estimate that you indicate for the region from reference 24 (0.8 to 1.0 kg P ha-1 year-1) did not account for canopy structure. It is interesting to note that this estimate is well within the range of annual deposition of 5-, 12-, and 20-year-old forests ((1.55±0.67 kg P ha-1 yr-1, 1.45±0.28 kg P ha-1 yr-1, and 1.07±0.21 kg P ha-1 yr-1, respectively) that you measure.

Line 111 : You never define UAV, please indicate "Unoccupied Aerial Vehicles".

Line 142: For the BCCMD (black carbon column mass density) from MERRA2, you point the reader to Fig S2. I believe this is a typo and that you meant to point to Fig. 4 of the main text.

Lines 141 to 144: you point to seasonal variations of BCCMD to explain the variations of measured P fluxes. Note that dry deposition is linked to surface concentrations and not to the colmun load of BC. It would have been more physically correct to show maps of surface concentrations for the dry deposition.

Reviewer #2 (Remarks to the Author):

This manuscript deals with a relevant and challenging issue. The phosphorus dry and wet deposition in tropical forests is not easy to measure and correctly interpret the data. The sampling has significant complications for dry and wet deposition as well as for throughfall. The throughfall is a mixture of wet deposition, canopy leaching as well as dry deposition itself.

More problematic yet is the attribution of fire-associated emissions over forests. We always have long-range transport mixed with local or regional natural biogenic particles. Nutrient recycling is a crucial component in tropical forests, and this is not discussed in the manuscript. The combination of all these factors brings significant uncertainties that require rigorous statistical analysis.

The manuscript has some critical problems. The first one is the lack of a comprehensive statistical treatment of data. The primary data reported in table S1 do not provide experimental errors or standard deviations based on variability. In any modern scientific study, error analysis must be done integrated into the experiment. Tables are presented without standard deviations of the measurements. Plots such as Figure 1, Figure 2, and Figure 3 do not show error bars. Table S2 and S3 also do not include any measurement uncertainties or standard deviations. I think the lack of proper statistical treatment is unacceptable in a scientific study.

The manuscript uses DBC (Dissolved Black Carbon) as a proxy for biomass burning. DBC was quantified using the benzenepolycarboxylic acid (BPCA) method, described in reference 22. This method is a nonstandard method for DBC measurement, and, as this is critically important for this manuscript, it needs to discuss how well his technique provides biomass burning DBC. What about the un-dissolved BC component? The manuscript does not mention this component, and it is essential because BC can have a significant fraction of the mass being non-soluble. It would be essential to measure total BC and compare it with the DBC component. How brown carbon that can account for 20-25% of BC absorption affects their estimate of DBC?.

In the methods section, I think that sample collection has fundamental problems. Rapid degradation of solutions in rainwater samples is very well known, and we usually use Thymol or other biocides to limit algae and bacteria growth. The methods section indicates that Thymol was not used in this study. Also is typical to collect the samples in refrigerated rainwater collectors such as Eigenbrodt wet only rainwater collector (https://www.eigenbrodt.de/). This is necessary because if we leave rainwater or throughfall water for a few days in a tropical area, it will decompose very quickly and provide errors in the determination of P and other rainwater components. Just keep the throughfall sampler under the ground do not cool it down enough. Additionally, the study has not used wet only collectors, so the precipitation also included some dry deposition components. This is not even mentioned in the manuscript and needs a much better assessment. Please look at the precipitation protocols of DEBITS (Deposition of biologically important trace species), WMO or EPA, where this critical issue is thoroughly discussed.

The manuscript also calculates canopy roughness using a 3D canopy model to deal with a potential canopy trapping effect. The dry deposition velocity (vds) of particles over the forest depends on many factors, including vertical air movements and convection. The authors need a better discussion on how these highly uncertain properties affect their results.

The potential biomass burning component needs better discussion about the sources and transport characterization. Most of the fires in Africa are from savannah burning, not fires from pristine tropical deforestation. Of course, the site is affected by BC's long-range transport from savannah fires that are not discussed in the manuscript. A figure with the biomass burning influence regions for the site generated using FLEXPART or HYSPLIT (https://www.arl.noaa.gov/hysplit/hysplit/) could add insights on the biomass burning sources for the sampling site.

The manuscript also has some strange descriptions of deforestation versus slash and burning agricultural fields and charcoal production. Two examples in the text that raises confusion and show errors in the drivers of tropical deforestation:

Abstract line 22: Slash and burning agriculture is not a mechanism for the conversion of pristine tropical forests. Deforestation is the mechanism involving pristine forests. *Slash-and-burn agriculture will convert important areas of pristine forest to fallow-based agroecosystems and secondary forest*.

Line 58 - Curiously, the manuscript does not recognize deforestation as a mechanism affecting tropical forests. Only slash and burn agriculture and charcoal production. Congo is the third country in terms of deforestation, after Brazil and Indonesia. **"***The African continent represents nearly 65% of the global burnt area annually[17], in part due to the prevalence of slash-and-burn agriculture[18,19] and associated high rates of charcoal production[20]* " .** The 65% of the global burnt area is due mainly to savannah fires, not tropical forest fires. I think this must be corrected.

In the abstract and the final discussions, the authors extrapolate this single point measurement to the Congo Tropical forests. Of course, this should not be done since the forest is quite heterogeneous, and one single point of measurement in a million km^2 is not representative of the whole forest.

Based on the above discussion, I think the manuscript has serious issues that maybe could be corrected in a revised version. At least it requires a profound revision on methods, data analysis, and conclusions. As it stands now, I do not recommend this manuscript for publication in Nature Comm.

Reviewer #3 (Remarks to the Author):

This is a very interesting study of P input in tropical forests in regions with disturbances. They show that slash-and-burn practices export P (and probably other base cation nutrients) as fire derived dry

deposition to the surrounding forests in amounts that may lift P limitation. The study is thorough including 12 forest plots and 3 agricultural fields in tropical Africa (Congo) – an understudied region. It will make a valuable contribution to the understanding of tropical forests and the anthropogenic impact on these ecosystems.

The paper is well written and the methods describe in sufficient detail. However I have a few major issues to be addressed in a revision:

1) The effect of canopy roughness (Fig 2) which has a central role in the paper is not convincing since I do not think there is a relationship between P-input and the canopy roughness parameter when the agricultural fields are removed. Could some of the effect be masked by variable distances to burned source areas? The observations can as they appear now be taken as an indication not as prof the complexity increase P deposition.

Just some thoughts on the canopy roughness parameter. It is not my expertise but recently fell over the parameter 'rugosity' (se e.g. doi:10.1016/j.foreco.2003.09.001 and an example use of it in Hardiman et al 2013 https://doi.org/10.1016/j.foreco.2013.02.031), just wonder how it compares to what was done here and if alternative approaches have been considered.

2) Line 119-126: This is a kind of circular argument: that slash-and-burn remove canopy which then reduce P-input that is released by slash-and-burn. I am not sure this is an important issue. Reformulate….

3) Although the methods describe the deposition parameters in detail (throughfall, wet, dry etc.) the text and Fig. 1 could be more clear in defining what is discussed, particularly dry deposition which appear to be THF minus BP.

4) Some minor comments and removal of meaningless decimals on fluxes that vary can be found in the attached file.

Dear Dr Bauters,

First, please let me apologize again for the time it has taken to get back to you with a decision.

Thank you again for submitting your manuscript "Fire-derived phosphorus fertilization of African tropical forests" to Nature Communications. We have now received reports from 3 reviewers and, after careful consideration, we have decided to invite a major revision of the manuscript.

As you will see from the reports copied below, the reviewers raise important concerns. We find that these concerns limit the strength of the study, and therefore we ask you to address them with additional work. Without substantial revisions, we will be unlikely to send the paper back to review. In particular, Reviewer #2 raised a number of critical concerns regarding the methods, which will need to be justified and discussed more thoroughly. Further, this reviewer also stressed the need for error and uncertainty reporting. Reviewer #3 also raised some concerns about the relationship between P-input and canopy roughness that needs to be addressed.

We thank the editor for their deft handling of our manuscript and appreciate the challenges of finding reviewers these days. We are completely confident in our ability to address the concerns of the reviewers. Please find our detailed responses below. <u>To facilitate the review process, all line numbers refer to changes in the track-changed version of the manuscript, attached at the end of this rebuttal letter.</u>

REVIEWER COMMENTS

<u>Reviewer #1 (Remarks to the Author):</u>

This manuscript presents convincing evidence that canopy complexity of forests of Central Africa plays a key role in the amount of phosphorus stored by these ecosystems. The age of the forest that determines tree structure and the canopy geometry acts as a trap for phosphorus for aged forests. The manuscript is clearly written, the arguments are convincing and I have only some minor points that could be clarified to improve the clarity of the points being made. I recommend that this manuscript be published with these improvements.
We thank the reviewer for the constructive and positive assessment of our manuscript. We have now used the suggestions to create an improved MS version.

<u>Major point:</u> I could not find in the main text or in the Supplement how the ranges for the deposition fluxes were calculated and what uncertainties/variations were accounted for.
This is now explained in the first sentences of the section "Data analysis" and we have added a short sentence in the main text to make this clear to the readers. Additionally, we added a new 'uncertainties' paragraph to the end of the methods section to make clear to the reader what the potential uncertainties of these setups might be. <u>(L 69-76, L365-382)</u>

<u>Minor points:</u>
Line 81: The measurements you made do not cover whole of Central Africa, refer the reader to Figure S1 that presents the measuring site and shows the extent of the area covered by these measurements.

The reviewer is right, we now explicitly added this info in the reporting. (L94)

Line 84: The estimate that you indicate for the region from reference 24 (0.8 to 1.0 kg P ha-1 year-1) did not account for canopy structure. It is interesting to note that this estimate is well within the range of annual deposition of 5-, 12-, and 20-year-old forests ((1.55±0.67 kg P ha-1 yr-1, 1.45±0.28 kg P ha-1 yr-1, and 1.07±0.21 kg P ha-1 yr-1, respectively) that you measure.
Indeed, very interesting, especially given that these numbers were derived from modeling work! We now also stress this in the manuscript (L123-126): "Annual P deposition in the 60-year-old forest (3.1±1.4 kg P ha-1 yr-1; Fig. 2) was found to be twice that of annual P deposition in 5-, 12-, and 20-year-old forests, which exhibited similar values (1.6±0.7 kg P ha-1 yr-1, 1.5±0.3 kg P ha-1 yr-1, and 1.1±0.2 kg P ha-1 yr-1, respectively) which in turn relate well to simulated estimates of annual P deposition for the area[24]."

Line 111 : You never define UAV, please indicate "Unoccupied Aerial Vehicles".
We thank the reviewer for this suggestion, we have now added this to the new MS version. (L128)

Line 142: For the BCCMD (black carbon column mass density) from MERRA2, you point the reader to Fig S2. I believe this is a typo and that you meant to point to Fig. 4 of the main text.
Indeed, the reviewer is right, this was an error. We now corrected this in the new MS version. (L163)

Lines 141 to 144: you point to seasonal variations of BCCMD to explain the variations of measured P fluxes. Note that dry deposition is linked to surface concentrations and not to the colmun load of BC. It would have been more physically correct to show maps of surface concentrations for the dry deposition.
Excellent remark. The reviewer is right, we have now redone the analysis with the black carbon surface mass density, and made changes accordingly throughout the manuscript. As you see, this does not affect the story – the trend is highly similar. (New Figure 4, L163,166, 169)

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We thank the reviewer for the constructive and thorough assessment of the manuscript. The reviewer has good suggestions, but we also feel that we can defend our approach in other points. Below we respond on a point-by-point basis to the reviewer. We have changed the manuscript where we agreed with the reviewer and feel that this has greatly improved the manuscript quality.

The manuscript has some critical problems. The first one is the lack of a comprehensive statistical treatment of data. The primary data reported in table S1 do not provide experimental errors or standard deviations based on variability. In any modern scientific study, error analysis must be done integrated into the experiment. Tables are presented without standard deviations of the measurements. Plots such as Figure 1, Figure 2, and Figure 3 do not show error bars. Table S2 and S3 also do not include any measurement uncertainties or standard deviations. I think the lack of proper statistical treatment is unacceptable in a scientific study.

We are confused by this comment.

<u>Supplementary Tables:</u> Table S1; this is all plot-level data. The deposition data is accumulated deposition data (originating from 52 summed weekly samples). The true replicates are the triplicated plots per stage. Instead of summarizing the data per successional stage and showing standard deviations per successional stage, we feel it is much more informative to report the raw plot-level data. The same is true for Table S2 and S3: these are the raw data that underlie the figures and regression models we fitted for the paper: i.e. the individual sample values. We do not feel it is appropriate to summarize these data to plot level and give averages +/- standard deviations given Nature Publishing's policy to provide the raw data in supplementary files as much as possible.

<u>Main Figures:</u> For Figure 1, 2, and 3 we are left with the same confusion: these depict the regression models on individual point measurements. Rather than only showing boxplots (which are added on the side panels of the regression figure, including percentile whiskers), we show the raw data to leave the reader with a good sense of the spread. We are thus unclear as to what kind of error bars the reviewer suggests we should include for the points, since these are individual measurements. Maybe the confusion stems from Figure 1, where only values are given for the general successional stages ('5yrs', '12 yrs' etc.)? As explained in the methods section, the samples of the three replicated plots per stage were composited for DBC analysis, so again, these points represent individual measurements of composite samples.

To avoid that potential source of confusion, we have now explicitly stated within each figure caption what the individual points represent. For Figure 4, we have added error bars for the deposition data, since here this is shown on the successional stage-level. Additionally, R^2 values are reported specifically in the figures to give the readers an informed 'first-look' of what the goodness-of-fit is for the data, for Figure 1 and Figure 2. For Figure 3 we have now added a table (Table S4) with the respective R^2 values and model equations in supplementary, because the figure itself would become too busy.

The manuscript uses DBC (Dissolved Black Carbon) as a proxy for biomass burning. DBC was quantified using the benzenepolycarboxylic acid (BPCA) method, described in reference 22. This method is a nonstandard method for DBC measurement, and, as this is critically important for this manuscript, it needs to discuss how well his technique provides biomass burning DBC.

We thank the reviewer for her/his important questions regarding DBC. We have added the following text to emphasize that the BPCA method quantifies the condensed aromatic fraction of DBC and is the leading technique for quantifying DBC in aquatic systems, including throughfall and stemflow (L245-254):

"The BPCA approach to quantifying DBC involves chemothermal oxidation of condensed aromatic DOC compounds to benzenehexacarboxylic acid (B6CA) and benzenepentacarboxylic acid (B5CA) products. The B6CA and B5CA oxidation products are robustly measured and derive exclusively from pyrogenic sources (Kappenberg et al., 2016). Condensed aromatic DBC, as measured using the BPCA method, is ubiquitous in aquatic environments globally (Jones et al., 2020; Coppola and Druffel, 2016; Wagner et al., 2018; 2019a). DBC has also been quantified in throughfall and stemflow in longleaf pine forests that undergo regular prescribed burning (Wagner et al., 2019b). Therefore, we use the BPCA method as a proxy for carbon inputs from biomass burning in the current study."

References (also added to the manuscript now)

Jones, M.W., Coppola, A.I., Santin, C., Dittmar, T., Jaffe, R., Doerr, S.H., Quine, T.A. Fires prime terrestrial organic carbon for riverine export to the global oceans. Nat. Commun. 11, 2791, doi.org/10.1038/s41467-020-16576-z (2020).

Kappenberg, A., Bläsing, M., Lehndorff, E., & Amelung, W. (2016). Black carbon assessment using benzene polycarboxylic acids: limitations for organic-rich matrices. Organic Geochemistry, 94, 47–51. https://doi.org/10.1016/j.orggeochem.2016.01.009

Coppola, A.I., Druffel, E.R.M. Cycling of black carbon in the ocean. Geophys. Res. Lett. 43, 4477-4482, doi:10.1002/2016GL068574 (2016).

Wagner, S., Brandes, J., Spencer, R.G.M., Ma, K., Rosengard, S.Z., Moura, J.M.S., Stubbins, A. Isotopic composition of oceanic dissolved black carbon reveals non-riverine source. Nat. Commun. 10, 5064, doi.org/10.1038/s41467-019-13111-7 (2019).

Wagner, S., Jaffé, R., & Stubbins, A. (2018). Dissolved black carbon in aquatic ecosystems. Limnology and Oceanography Letters, 3(3),168–185. https://doi.org/10.1002/lol2.10076

Wagner, S., Brantley, S., Stuber, S., Van Stan, S., Whitetree, A., Stubbins, A. (2019) Dissolved black carbon in throughfall and stemflow in a fire-managed longleaf pine woodland. Biogeochemistry 146:191–207. doi.org/10.1007/s10533-019-00620-2

What about the un-dissolved BC component? The manuscript does not mention this component, and it is essential because BC can have a significant fraction of the mass being non-soluble. I would be essential to measure total BC and compare it with the DBC component. How brown carbon that can account for 20-25% of BC absorption affects their estimate of DBC?

We agree that undissolved, or particulate black carbon (PBC) is a potentially interesting component of deposition. However, we were solely interested in using DBC as a proxy for fire-derived depositional inputs rather than quantifying the total inputs of BC. Furthermore, the mass median diameter (MMD) of BC aerosols is ubiquitously at or below 0.2 micron (see rebuttal table below for some literature values). This size is well below the threshold for DBC in our samples that were filtered to 0.45 micron and thus supports our use of DBC as a proxy for quantity of fire-derived inputs. Lastly, given the pre-dominance of biomass burning as the source of combustion derived aerosols in the Congo and the long-range transport of aerosols to our study site, there is no reason to expect the proportions of DBC and PBC to vary substantially through time. We have added this also to the 'uncertainties' paragraph at the end of the methods section. (L365-382)

Regarding the brown and black carbon analytical overlap: naming conventions used to describe thermally altered carbon fractions have evolved alongside the methods used to characterize them (Hammes and Abiven, 2013). For example, researchers in the atmospheric community describe BC as light-absorbing, soot-like aerosols, whereas aquatic scientists typically describe BC as the condensed aromatic carbon fraction measured via BPCA analysis (see reply to previous comment). We now describe the analytical windows for the method used to quantify DBC in the current study to clarify exactly what we are measuring here. However, it is unknown how DBC as measured by BPCAs overlaps with brown carbon measurements the reviewer refers to here.

References

Hammes, K. & Abiven, S. Identification of Black Carbon in the Earth System. in Fire Phenomena and the Earth System: An Interdisciplinary Guide to Fire Science (2013). doi:10.1002/9781118529539.ch9

Particle Source	BC MMD (nm)	Reference
BB (long range transport)	120-160	Dahlkötter et al. 2014
BB (Asia)	177-197	Kondo et al. 2011
BB (Canada)	176-238	Kondo et al. 2011
BB (California)	177-209	Sahu et al. 2012
BB (Texas)	200-220	Schwarz et al. 2008
BB (Canada)	195	Taylor et al. 2014
BB (Wyoming)	170	Pratt et al. 2011
Urban Emissions (California)	120-160	Metcalf et al. 2012
Urban Emissions (Texas)	160-180	Schwarz et al. 2008

Rebuttal Table 1: *reported mass median diameter (MMD) of black carbon (BC). Adopted from Williams et al. 2019.*

Table references:

Dahlkötter, F., Gysel, M., Sauer, D., Minikin, A., Baumann, R., Seifert, P., et al. (2014). The Pagami Creek smoke plume after long-range transport to the upper troposphere over Europe – Aerosol properties and black carbon mixing state. Atmospheric Chemistry and Physics, 14(12). https://doi.org/10.5194/acp-14-6111-2014

Kondo, Y., Matsui, H., Moteki, N., Sahu, L., Takegawa, N., Kajino, M., et al. (2011). Emissions of black carbon, organic, and inorganic aerosols from biomass burning in North America and Asia in 2008. Journal of Geophysical Research Atmospheres, 116(8), 1–25. https://doi.org/10.1029/2010JD015152

Sahu, L. K., Kondo, Y., Moteki, N., Takegawa, N., Zhao, Y., Cubison, M. J., et al. (2012). Emission characteristics of black carbon in anthropogenic and biomass burning plumes over California during ARCTAS-CARB 2008. Journal of Geophysical Research Atmospheres, 117(16), 1–20. https://doi.org/10.1029/2011JD017401

Schwarz, Joshua P., Gao, R. S., Spackman, J. R., Watts, L. A., Thomson, D. S., Fahey, D. W., et al. (2008). Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions. Geophysical Research Letters, 35(13), 1– 5. https://doi.org/10.1029/2008GL033968

Taylor, J. W., Allan, J. D., Allen, G., Coe, H., Williams, P. I., Flynn, M. J., et al. (2014). Sizedependent wet removal of black carbon in Canadian biomass burning plumes. Atmospheric Chemistry and Physics, 14(24), 13755–13771. https://doi.org/10.5194/acp-14-13755-2014

Pratt, K. A., Murphy, S. M., Subramanian, R., Demott, P. J., Kok, G. L., Campos, T., et al. (2011). Flight-based chemical characterization of biomass burning aerosols within two prescribed burn smoke plumes. Atmospheric Chemistry and Physics, 11(24), 12549–12565. https://doi.org/10.5194/acp-11-12549-2011

Metcalf, A. R., Craven, J. S., Ensberg, J. J., Brioude, J., Angevine, W., Sorooshian, A., et al. (2012). Black carbon aerosol over the Los Angeles Basin during CalNex. Journal of Geophysical Research Atmospheres, 117(8), 1–24. https://doi.org/10.1029/2011JD017255

In the methods section, I think that sample collection has fundamental problems. Rapid degradation of solutions in rainwater samples is very well known, and we usually use Thymol or other biocides to limit algae and bacteria growth. The methods section indicates that Thymol was not used in this study. Also is typical to collect the samples in refrigerated rainwater collectors such as Eigenbrodt wet only rainwater collector (https://www.eigenbrodt.de/). This is necessary because if we leave rainwater or throughfall water for a few days in a tropical area, it will decompose very quickly and provide errors in the determination of P and other rainwater components. Just keep the throughfall sampler under the ground do not cool it down enough. Additionally, the study has not used wet only collectors, so the precipitation also included some dry deposition components. This is not even mentioned in the manuscript and needs a much better assessment. Please look at the precipitation protocols of DEBITS (Deposition of biologically important trace species), WMO or EPA, where this critical issue is thoroughly discussed.

We agree with the reviewer that using thymol is generally a good practice for these kinds of setups. However, in this case, we designed the project specifically without adding thymol, because we are basing all our findings on total phosphorus (TP) concentration of the sample. The reviewer is right that the elemental species composition of the sample might change through microbial activity: i.e. relative shift in N species composition, DOC decomposition etc. However, <u>total phosphorus concentrations</u> of the samples are not likely to be altered by microbial activity: the total P amount in the bottle will not change through microbial activity. Additionally, we explicitly wanted to avoid adding unnecessary organic chemicals to the samples. Thymol contains an aromatic ring, which might contaminate our future planned FT-ICR MS analyses for the samples (Bauters et al. 2018). We considered using mercury, but the risk for environmental contamination (the labs in the DRC are not equipped for the disposal) was simply too high. Putting the collectors in the ground and covering them was not only done to cool them down as much as possible, but also to avoid sun light (algal growth).

For the automated collectors, please bear in mind that these field sites are remote and there is no grid power in the forest. Additionally, the more the setup relies on advanced and expensive technologies (incl. solar panels, powered collectors, etc.), the more problems we have with maintaining and protecting it from theft and intermittent failures. We have operated one ISCO automated sampler (for peak discharge) at this site for 1 year now and it has been stolen once (but we recovered it) and the batteries have been stolen twice. You can imagine the difficulties of trying to maintain 120 collectors (15 x 8) with similar theft rates. Hence, we have learnt the hard way that low-tech approaches are preferred to ensure continued on-site monitoring.

*Regardless, we now acknowledge specifically in the manuscript that there might have been some microbial decomposition because of our 1-week sample storage in the field, but state explicitly also that this we do not expect this to affect the **total phosphorus concentrations** of the samples. We have now explicitly acknowledged this, along with the wet vs. dry only collectors, in the new 'uncertainties' section at the end of the methods section. (L365-382)*

The manuscript also calculates canopy roughness using a 3D canopy model to deal with a potential canopy trapping effect. The dry deposition velocity (vds) of particles over the forest depends on many factors, including vertical air movements and convection. The authors need a better discussion on how these highly uncertain properties affect their results. The potential biomass burning

component needs better discussion about the sources and transport characterization. Most of the fires in Africa are from savannah burning, not fires from pristine tropical deforestation. Of course, the site is affected by BC's long-range transport from savannah fires that are not discussed in the manuscript. A figure with the biomass burning influence regions for the site generated using FLEXPART or HYSPLIT (https://www.arl.noaa.gov/hysplit/hysplit/) could add insights on the biomass burning sources for the sampling site.

For the dry deposition velocity: Indeed, high resolution wind speed and direction data would be needed to simulate the actual (high resolution) dry deposition. However, given that our study is looking deposition at low temporal resolution (accumulated, weekly), and given that the proximity of the plots controls for any significantly divergent wind conditions over the landscape, we feel that the structural component (canopy roughness/rugosity) can be isolated as the dominant driver for the dry deposition. This is also why we focus on this in the manuscript and why the statistical fits perform so well. Given these factors, we do not find it necessary to delve into actual high resolution dry deposition velocity for this study.

Considering the dry deposition velocity: we now added a short section on this in the new 'uncertainty paragraph'. For the source of the biomass burning-derived deposition: we agree with the reviewer that this was not discussed explicitly enough in the previous version. In fact, we have exploited the combination of HYSPLIT and fire incidence in Africa in previous work on the same site (but only for pristine forest and a different monitoring period; Bauters et al. 2018). While this is indeed informative, we don't feel that we can do a better job with this dataset than in the previous work on identifying sources, since it would basically just repeat the same analysis for a different paper. Additionally, through substantial storage of aerosol constituents in the canopy, as we show, the signal becomes obscured over the year. However, we now more explicitly refer to the findings of this earlier work and have added a new Figure S3 in supplementary material along with a new methods section, in which we apply the same methodology (HYSPLIT vs. Fire incidence per season) to show to the reader how biomass burning aerosols could be sourced and transported across the continent. (L351-363, new Figure S3, L108)

References:

Bauters, M. et al. High fire-derived nitrogen deposition on central African forests. Proc. Natl. Acad. Sci. **115**, *549–554 (2018).*

The manuscript also has some strange descriptions of deforestation versus slash and burning agricultural fields and charcoal production. Two examples in the text that raises confusion and show errors in the drivers of tropical deforestation: Abstract line 22: Slash and burning agriculture is not a mechanism for the conversion of pristine tropical forests. Deforestation is the mechanism involving pristine forests. Slash-and-burn agriculture will convert important areas of pristine forest to fallow-based agroecosystems and secondary forest. Line 58 - Curiously, the manuscript does not recognize deforestation as a mechanism affecting tropical forests. Only slash and burn agriculture and charcoal production. Congo is the third country in terms of deforestation, after Brazil and Indonesia.

We thank the reviewer for pointing out this unclear use of terminology. In our original manuscript, we were mainly focusing on slash-and-burn and charcoal production, since these are the main drivers for deforestation in the Congo basin, as opposed to South America and South-east Asia, where commercial logging and commodity plantations are a far bigger threat for pristine forests (see Curtis et al. 2018). In central Africa, deforestation, i.e. conversion of pristine forest to other land use types, is predominantly caused by smallholder practices such as slash and burn, and charcoal production, as has been shown by several studies (Curtis et al. 2018, Tyukavina et al. 2018, Tyukavina et al. 2013).

We have now rephrased our statements in several parts of the manuscript to avoid confusion.(L21-28, L60-62)

References:

*Curtis, P. G., Slay, C. M., Harris, N. L., Tyukavina, A. & Hansen, M. C. Classifying drivers of global forest loss. Science (80-.). **1111**, 1108–1111 (2018).*

*Tyukavina, A. et al. Congo Basin forest loss dominated by increasing smallholder clearing. Sci. Adv. **4**, (2018).*

*Tyukavina, A. et al. National-scale estimation of gross forest aboveground carbon loss: a case study of the Democratic Republic of the Congo. Environ. Res. Lett. **8**, 044039 (2013).*

"The African continent represents nearly 65% of the global burnt area annually[17], in part due to the prevalence of slash-and-burn agriculture[18,19] and associated high rates of charcoal production[20] " . The 65% of the global burnt area is due mainly to savannah fires, not tropical forest fires. I think this must be corrected.

We fully agree with the reviewer on this point. We have now changed this in the new MS version. (L61-62)

In the abstract and the final discussions, the authors extrapolate this single point measurement to the Congo Tropical forests. Of course, this should not be done since the forest is quite heterogeneous, and one single point of measurement in a million km² is not representative of the whole forest. Based on the above discussion, I think the manuscript has serious issues that maybe could be corrected in a revised version. At least it requires a profound revision on methods, data analysis, and conclusions. As it stands now, I do not recommend this manuscript for publication in Nature Comm.

We agree with the reviewer that the previous manuscript version might have overstated the results in some parts. We have now changed this in the new MS version and better qualified our extrapolation (L94, L30, L37, L38). Nevertheless, we want to stress that the main finding of manuscript is the mechanism by which more complex and mature forests capture dry deposition. Although the reviewer states that the Congo forests are quite heterogeneous, recent research has shown that the forests of the whole Congo Basin can be classified in three rather homogenous types (Réjou-Méchain et al. 2021 Nature). Additionally, forest heterogeneity per se (in terms of species distributions etc.) should not matter for P deposition. Rather, it is the combination of forest structure and the cocktail of biomass burning aerosols and its spatial (and temporal) fluctuations that will ultimately determine the magnitude of P deposition. The forest structure link is clearly exploited in our manuscript (in a mechanistic way), and wanted to show via Figure 4d that biomass burning aerosols are omnipresent in large parts of the basin. Hence, although we indeed tone down the overgeneralization in the new MS version, we feel confident that the mechanisms that underpin our observations hold for a much larger region.

Reference:

Réjou-Méchain, M. et al. Unveiling African rainforest composition and vulnerability to global change. Nature (2021). doi:10.1038/s41586-021-03483-6

In our responses and revised manuscript, we feel we were able to further clarify our approach in several parts of the paper's methodology and amend our analyses and conclusions.

Reviewer #3 (Remarks to the Author):

This is a very interesting study of P input in tropical forests in regions with disturbances. They show that slash-and-burn practices export P (and probably other base cation nutrients) as fire derived dry deposition to the surrounding forests in amounts that may lift P limitation. The study is thorough including 12 forest plots and 3 agricultural fields in tropical Africa (Congo) – an understudied region. It will make a valuable contribution to the understanding of tropical forests and the anthropogenic impact on these ecosystems. The paper is well written and the methods describe in sufficient detail.

We thank the reviewer for the constructive and positive assessment of our manuscript. We have now used the suggestions to create an improved MS version.

However I have a few major issues to be addressed in a revision:
1) The effect of canopy roughness (Fig 2) which has a central role in the paper is not convincing since I do not think there is a relationship between P-input and the canopy roughness parameter when the agricultural fields are removed. Could some of the effect be masked by variable distances to burned source areas? The observations can as they appear now be taken as an indication not as prof the complexity increase P deposition.
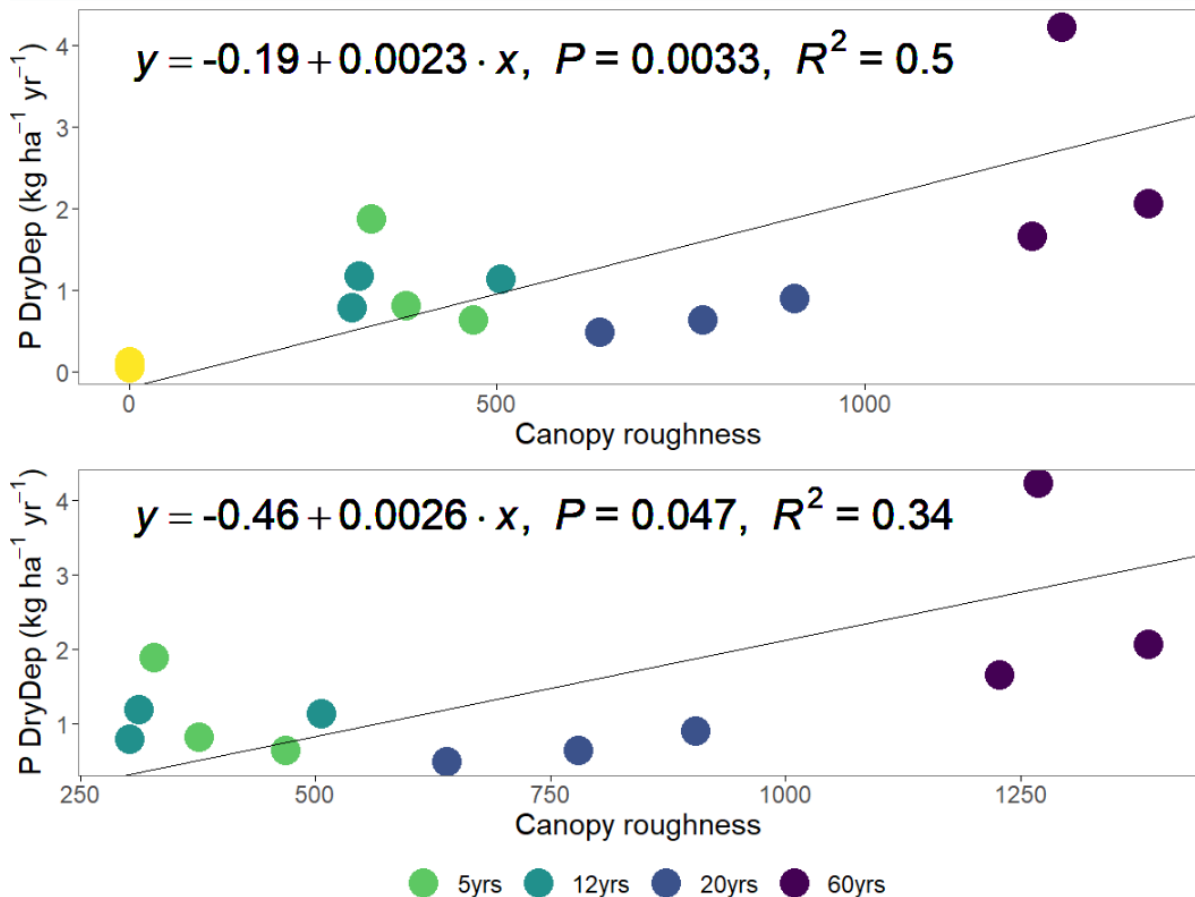Just some thoughts on the canopy roughness parameter. It is not my expertise but recently fell over the parameter 'rugosity' (se e.g. doi:10.1016/j.foreco.2003.09.001 and an example use of it in Hardiman et al 2013 https://doi.org/10.1016/j.foreco.2013.02.031), just wonder how it compares to what was done here and if alternative approaches have been considered.

We thank the reviewer for this excellent suggestion. In fact, yes, we considered including 'rugosity' in the original manuscript version. The paper referred to (Hardiman et al.) used a lidar (i.e. vertical canopy profile) based proxy, that integrates the full 3D 'heterogeneity' of the canopy structure. For our sites, these LiDAR measurements are not available, nor does it seem necessary when considering physics behind dry deposition. This is because the lower canopy layers should play a relatively minor role for the deposition of particles on the canopy surface layer. However, Hardiman et al. derived their method from Parker and Russ (reference below), who only considered the upper canopy surface 'vertical height variability'. This is an excellent metric to compare to our 'roughness' parameter. In some ways, it is a simplified version of our 'roughness index: it is just a proxy for the overall variability in height along the canopy transects (cf. Figure S2 in the original manuscript file). Originally, we felt that our roughness parameter was a better 'mechanistic' parameter to include, because it is a proxy for the likelihood that a particle encounters a positive height difference (an obstacle) as it passes over the canopy. However, it seems like rugosity – a much simpler index – might also be worth including for simply being much easier to grasp. Hence, although we decided not to include this in the original manuscript version, we feel that many readers might be left with the same question of the reviewer, so we now also include this in the revised version. (Changed Figure 2, L131-133, L320-324)

Regarding the agricultural fields potentially driving the relationship: actually, the relationship holds without the fields (see Rebuttal figure 1 below). Nevertheless, we generally downturned our conclusions and reformulated these relations (although rugosity again confirms a good fit in our opinion) as being an indication for rather than hard proof. (L30, L32, L131-137, L182)

References

Parker, G. G. & Russ, M. E. The canopy surface and stand development : assessing forest canopy structure and complexity with near-surface altimetry. For. Ecol. Manage. **189**, 307–315 (2004).

The equations shown in the figure:

$$y = -0.19 + 0.0023 \cdot x, \quad P = 0.0033, \quad R^2 = 0.5$$

$$y = -0.46 + 0.0026 \cdot x, \quad P = 0.047, \quad R^2 = 0.34$$

Legend: 5yrs, 12yrs, 20yrs, 60yrs

Rebuttal Figure 1: Canopy Roughness as a predictor for P dry deposition. This figure shows the linear fits with (upper panel) and without (lower panel) agricultural fields. Note that the effect estimate hardly changes with omission of the agricultural fields.

2) Line 119-126: This is a kind of circular argument: that slash-and-burn remove canopy which then reduce P-input that is released by slash-and-burn. I am not sure this is an important issue. Reformulate….

We thank the reviewer for pointing this out. We have now indeed reformulated this in the new MS version. This was too speculative for our own data, but the statement actually just tries to provide the reader with an 'outlook' of our observations, based on similar work by Lawrence and colleagues. We believe this is now better formulated in the new version. (L142-152)

3) Although the methods describe the deposition parameters in detail (throughfall, wet, dry etc.) the text and Fig. 1 could be more clear in defining what is discussed, particularly dry deposition which appear to be THF minus BP.

We thank the reviewer for pointing this out. We now more clearly state in the main text as well as in the figure caption what we considered 'dry deposition' and how this was calculated and added explicit pointers on several places to remind the reader of this. We also acknowledge in the new 'uncertainties' section that our dry and wet deposition quantification are not exact because we did not use 'wet only' collectors. (L69-76, L103-104, L154-155)

4) Some minor comments and removal of meaningless decimals on fluxes that vary can be found in the attached file.

We thank the reviewer for these comments. This will improve the readability and consistency of the manuscript. We have made following edits as suggested:

- *Removed decimals L96*
- *Added 'dry' L101*
- *Removed decimals L123, 124, 125*
- *Deleted space L190*
- *Removed decimals L215*
- *Removed decimals L264*
- *Deleted space L2*

1 **Title**: **Fire-derived phosphorus fertilization of African tropical forests**

2

3 **Authors**: Marijn Bauters[1,2,*], Travis W. Drake[3], Sasha Wagner[4], Simon Baumgartner[1,5], Isaac

4 Makelele[1,6], Samuel Bodé[1], Kris Verheyen[2], Hans Verbeeck[2], Corneille Ewango[7], Landry

5 Cizungu[8], Kristof Van Oost[5], Pascal Boeckx[1]

6

7 [1] Department of Green Chemistry and Technology, Ghent University, Ghent, 9000, Belgium

8 [2] Department of Environment, Ghent University, Ghent, 9000, Belgium

9 [3] Department of Environmental Systems Science, Swiss Federal Institute of Technology, ETH

10 Zurich, Zurich, 8092,Switzerland

11 [4] Department of Earth and Environmental Sciences, Rensselaer Polytechnic Institute, Troy,

12 New York, USA

13 [5] Earth and Life Institute, UCLouvain, Louvain-la-Neuve, 1348, Belgium

14 [6] Department of Biology, Université Officielle de Bukavu, Bukavu, DRCongo

15 [7] Faculty of Renewable Natural Resources Management, University of Kisangani, B.P.O.

16 2012, Kisangani, DR Congo

17 [8] Soil science laboratory, Faculty of Agronomy, Université Catholique de Bukavu, Bukavu,

18 DR Congo

19 * Corresponding author: Marijn.Bauters@UGent.be

20

~~The Congo basin's~~Central African tropical forest~~s is~~s face ~~facing~~ increasing anthropogenic pressure~~s~~, particularly in the form of deforestation and land-use conversion to agriculture. The long-term effects of this transformation of ~~, converting substantial areas of p~~pristine forests ~~to~~to fallow-based agroecosystems and secondary forests ~~secondary forests~~through biomass burning-associated practices. Slash-and-burn ~~agriculture will convert substantial areas of pristine forest to fallow-based~~ ~~agroecosystems and secondary forest.~~. The long-term effect of such anthropogenic ~~activities~~ on biogeochemical cycles that drive forest functioning are poorly understood. Here, we show that biomass burning ~~activities~~ on the African continent result~~s~~ in high phosphorus (P) deposition on ~~equatorial forest~~an equatorial forest~~s~~ via fire-derived atmospheric emissions. Furthermore, we show that deposition loads increase with forest regrowth age, likely due to increasing canopy complexity, ranging from 0.4 kg P ha^{-1} yr^{-1} on agricultural fields to 3.1 kg P ha^{-1} yr^{-1} on old secondary forests. In forest systems, canopy wash-off of dry P deposition increases with rainfall amount, highlighting how tropical forest canopies act as dynamic reservoirs for enhanced addition of this essential plant nutrient. Overall, the magnitude of the observed P deposition load at the study site challenges the P-limitation status ~~of the second-largest tropical forest~~ of this forest ~~on Earth~~ and demonstrates the importance of canopy trapping as a pathway for nutrient input into forest ecosystems.

Tropical forests store a substantial amount of the Earth's terrestrial carbon and host a large share of global biodiversity. Although nitrogen (N) is a potentially limiting plant nutrient early-on in tropical forest succession[1,2], it is a virtually infinite resource that can be fixed biologically from air[3–5]. In contrast, phosphorus (P) has been identified to be the main limiting nutrient in many old-growth tropical forests rooted in strongly weathered soils[6,7]. As such,

these forest ecosystems are dependent upon trace inputs of P and cations from the atmosphere (e.g., as dust or biomass-derived emissions) rather than inputs from soil weathering[8,9]. In light of the dependency of tropical forests on these atmospheric P inputs, it is hypothesized that forest canopies may function as a 'trap' for atmospheric constituents[10,11]. As they regrow, secondary forests exhibit increased canopy complexity (e.g., surface area or roughness), which enables more efficient trapping of atmospheric constituents[12]. Furthermore, repeated deforestation can reduce atmospheric P inputs to a level that induces a long-term negative ecosystem P balance[13]. As such, the loss of this canopy trap through repeated clearing might also reduce cumulative P inputs to an extent where some forests completely lose the capacity to naturally recover from disturbances[14]. Understanding the biogeochemical and physical interactions of canopy trapping and atmospheric P deposition along secondary forest succession is vital for tropical landscapes, which face increasing anthropogenic pressures. These effects are particularly relevant in Africa, where secondary forests are expected to increasingly dominate the landscape as a result of ongoing slash-and-burn agricultural practices[15,16]. The African continent represents nearly 65% of the global burnt area annually[17], ~~in part due to the prevalence of slash-and-burn agriculture[18,19] and associated high rates of charcoal production[20]~~mainly via a high incidence of grassland and savanna fires[18]. This biomass burning leads to high fire-derived nitrogen deposition loads on central African forests[19], but the deposition of potentially limiting nutrients, such as P, and the interaction with canopy trapping along forest regrowth trajectories, is unknown.

We quantified atmospheric P deposition along a forest successional gradient in the central Congo basin to estimate the magnitude of biomass burning-derived P deposition and to assess the importance of canopy trapping in regulating the delivery of P to the forest floor. For this, we set up permanent precipitation collectors in 40 by 40 m monitoring plots along a

successional gradient comprising five stages: agriculture, 5-year-old, 12-year-old, 20-year-

72   old, and 60-year-old secondary forests, near Kisangani. Per successional stage, we set up three

73   plots, and each plot was equipped with eight rainfall collectors, which were sampled weekly.

74   Nutrient loads in open field and throughfall was calculated by multiplying the rainfall volume

75   with the total nutrient concentration per plot, per week. We used open field precipitation as a

76   'wet' deposition endmember, and calculated net throughfall loads to quantify dry deposition.

77   To date, it has proven difficult to quantify net nutrient deposition in forest ecosystems, as

78   canopy uptake/leaching effects are difficult to separate from dry deposition effects in

79   throughfall samples. Here, we used dissolved black carbon (DBC; measured as

80   benzenepolycarboxylic acid molecular markers[20]) as a tracer for biomass burning-derived

81   aerosols in throughfall samples. The burning of biomass releases nutrients and produces black

82   carbon, a heterogeneous mixture of charcoal, soot and other thermally altered forms of

83   organic carbon[21]. We related DBC deposition to P deposition (both wet and dry) and then

84   compared results to canopy complexity in forests along a successional gradient (varying in

85   age from 5 to 60 years since agricultural abandonment).
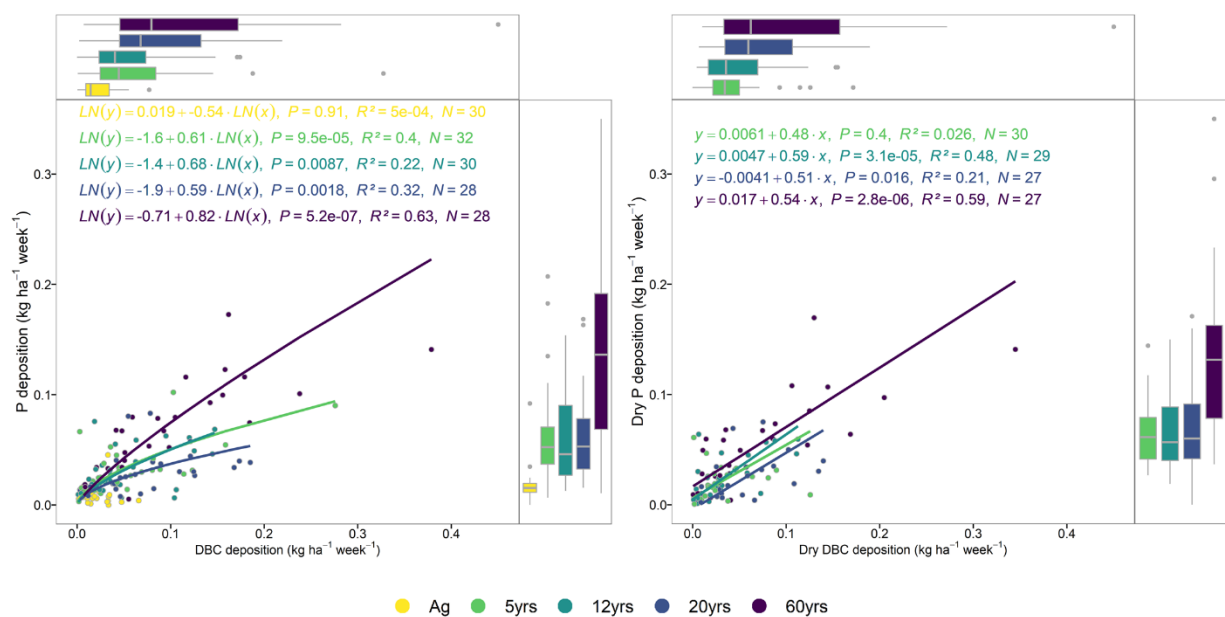
Fig. 1. Weekly total phosphorus deposition versus dissolved black carbon deposition (DBC deposition) (left) and

dry P deposition versus dry DBC deposition (right) along successional stages of central African forests:

3

89   agricultural field (Ag), 5- (5yrs), 12- (12yrs), 20- (20yrs) and 60-year-old forest (60yrs). Boxplots show the

90   median, the 25th and 75th percentile and the 1.5 interquartile range spread. Points represent individual, -weekly,

91   measurements of composite samples (n=8) per plot for each successional stage.

92

93   **High P deposition on central African forests is sourced from biomass burning.** Our data

94   show that open field wet deposition ~~in~~ at our site in central Africa (Fig. S1) is 0.43±0.09 kg P

95   ha^{-1} yr^{-1}, while 60-year-old forests are subjected to an average total deposition load (~~i.e.~~i.e.,

96   wet and dry deposition) of 3.1~~08~~±1.4~~2~~ kg P ha^{-1} yr^{-1}. These measured P loads are substantially

97   higher than what has been previously estimated for the region (0.8-1.0 kg P ha^{-1} yr^{-1};[22]), but

98   similar to total P deposition measured on the shores of Lake Victoria (1.8–2.7 kg P ha^{-1} year^{-1}

99   [,23]). We analyzed DBC in pooled samples (see methods) to determine whether deposited P

100 measured in throughfall (Fig. 1) was indeed related to biomass burning. Since dry P

101 deposition scales linearly with DBC dry deposition along the forest chronosequence (Fig. 1b),

102 it indicates that 1) the excess P input measured as net throughfall under forest canopies is fire-

103 derived, and 2) net P throughfall loads (i.e., the throughfall P minus the open field P

104 deposition) are derived entirely from biomass burning, rather than canopy leaching. A

105 pyrogenic source for throughfall P is consistent with previous studies that suggest an

106 estimated >50% of global atmospheric P is derived from combustion[24]. Africa is also known

107 to be a hotspot for biomass burning and constitutes over half of the global carbon emissions

108 from fire[18], with the fire incidence being most abundant in the savanna zones (Figure S3).

109 Additionally, localized P emissions from biomass burning have been shown to be up to 20 kg

110 P burnt ha^{-1} [25], owing to the rather high P emission factors from tropical forest and savanna

111 fires[26].

112
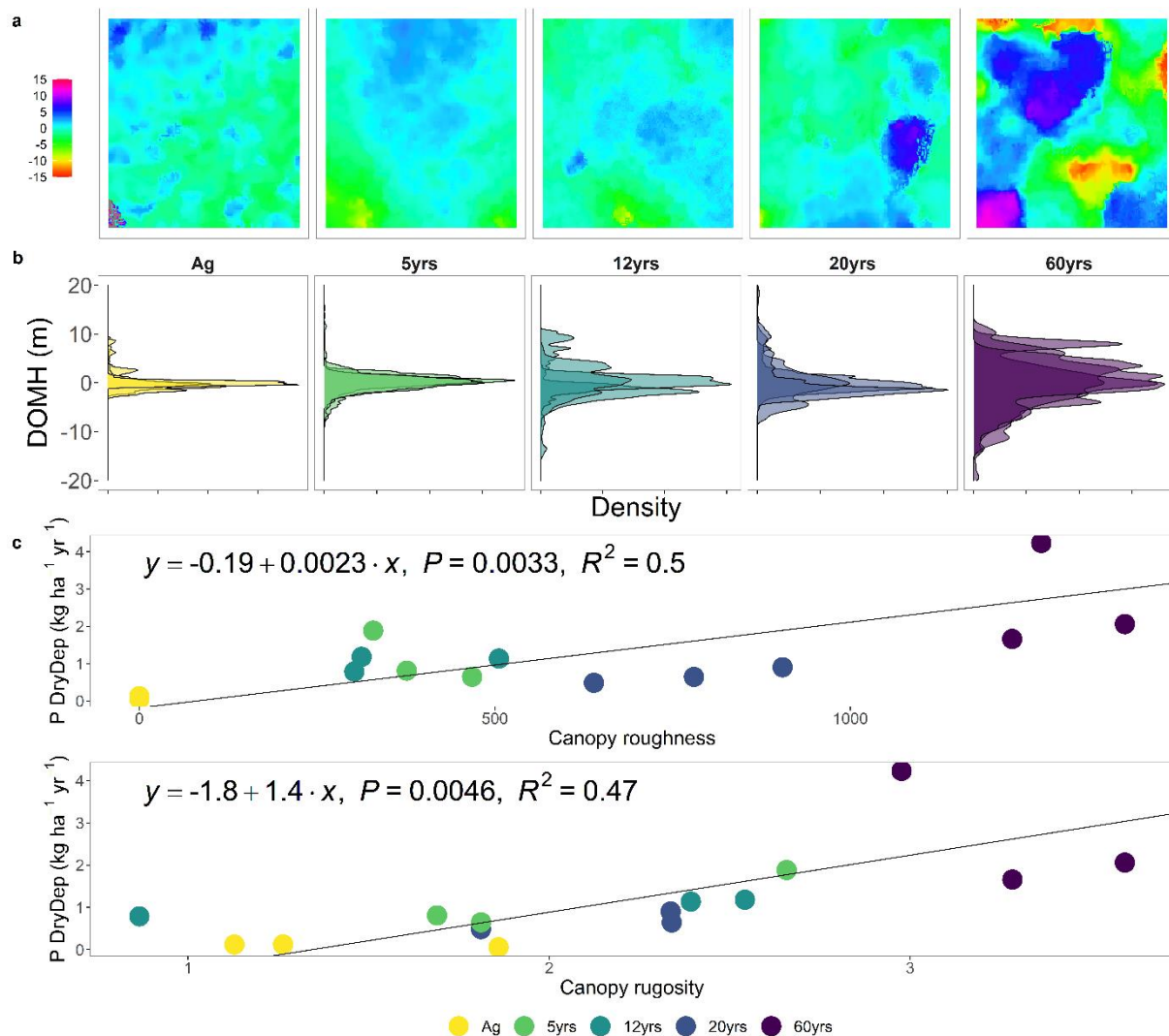
Fig. 2. Canopy complexity and annual phosphorus <u>dry</u> deposition along a successional forest stages: agricultural field (Ag), 5 (5yrs), 12 (12yrs), 20 (20yrs) and 60-year-old forest (60yrs). (a) Canopy height models show the deviation in canopy height (m) from the mean of a 40 by 40 m plot per successional stage. (b) Density distributions for three plots per successional stage displaying the deviation of mean height (DOMH, m) from the plot's mean. (c) Canopy roughness and rugosity, based on Unoccupied Aerial Vehicle-derived structure for motion and vegetation height, versus the annual P dry deposition. <u>Points in (c) represent the plot-level measurements for annual P deposition and canopy roughness and rugosity.</u>

Aerosol trapping increases with forest age and canopy complexity. Annual P deposition in the 60-year-old forest (3.1~~08~~±1.4~~2~~ kg P ha^{-1} yr^{-1}; Fig. 2) was found to be twice that of annual P deposition in 5-, 12-, and 20-year-old forests, which exhibited similar values (1.6~~55~~±0.7~~67~~ kg P ha^{-1} yr^{-1}, 1.5~~45~~±0.3~~28~~ kg P ha^{-1} yr^{-1}, and 1.1~~07~~±0.2~~1~~ kg P ha^{-1} yr^{-1}, respectively) <u>which in</u>

5

126 turn relate well to simulated estimates of annual P deposition for the area [24]. To further

127 examine the role of forest age and structure on P deposition, we quantified canopy

128 complexity, using high resolution 3D point clouds derived from Unoccupied Aerial Vehicle

129 (UAV) imagery, as canopy 'roughness'. ~~-~~ Canopy roughness is linearly related to the dry

130 deposition velocity of aerosol particles [27,28]. Canopy complexity encompasses both the

131 structural variability of the canopy, as well as the vegetation height. Additionally, we used

132 canopy rugosity, ~~i.e.~~ the standard deviation of vertical variability in the upper canopy layer, as

133 a ~~more~~ simple structural metric related to canopy complexity. As hypothesized by Powers et

134 al.[12], our data ~~show~~ suggest ~~indeed~~ that more complex canopies, that is, with ~~—~~ higher canopy

135 roughness and ~~canopy~~ rugosity, ~~—~~ trap more P via dry deposition (Fig. 2). This ~~, which~~

136 highlights canopy trapping as a potentially~~n~~ important ecosystem-level mechanism for

137 nutrient acquisition. Our data also suggest that forests trap nutrient-rich aerosols with

138 increasing efficiency as they recover from disturbance. The first 20 years of forest succession

139 exhibited a plateau in the structural variability of the canopy (see λ in Table S1), which is

140 mirrored by consistent P dry deposition loads. However, both canopy complexity and P dry

141 deposition loads increased in 60-year-old forests. The observed variability in canopy trapping

142 across the successional gradient, which lead ~~has~~ ~~leading~~ to three-fold differences in P input to

143 the forest floor ~~, across the gradient~~ ~~—~~ might have important ramifications for the nutrient

144 cycles of central African forests. Indeed, ~~, since~~ reduced canopy cover also ~~reduces locally~~

145 reduces the local atmospheric inputs of nutrients to the soil. ~~The localized removal of older~~

146 ~~and more complex canopies via intensified slash-and-burn and deforestation practices may~~

147 ~~result in the overall reduction in atmospheric P inputs to the Congo-basin forest floor.~~ Thus,

148 long-term absence of a canopy ~~disturbance , could result in negative ecosystem feedback,~~

149 ~~where reduced input of limiting nutrients~~ might thereby enhance a long-term negative

6

150 ecosystem nutrient balance, could and constraint future forest recovery, that should be

151 considered in land use management, much likeas described by Lawrence and colleagues[13].

152

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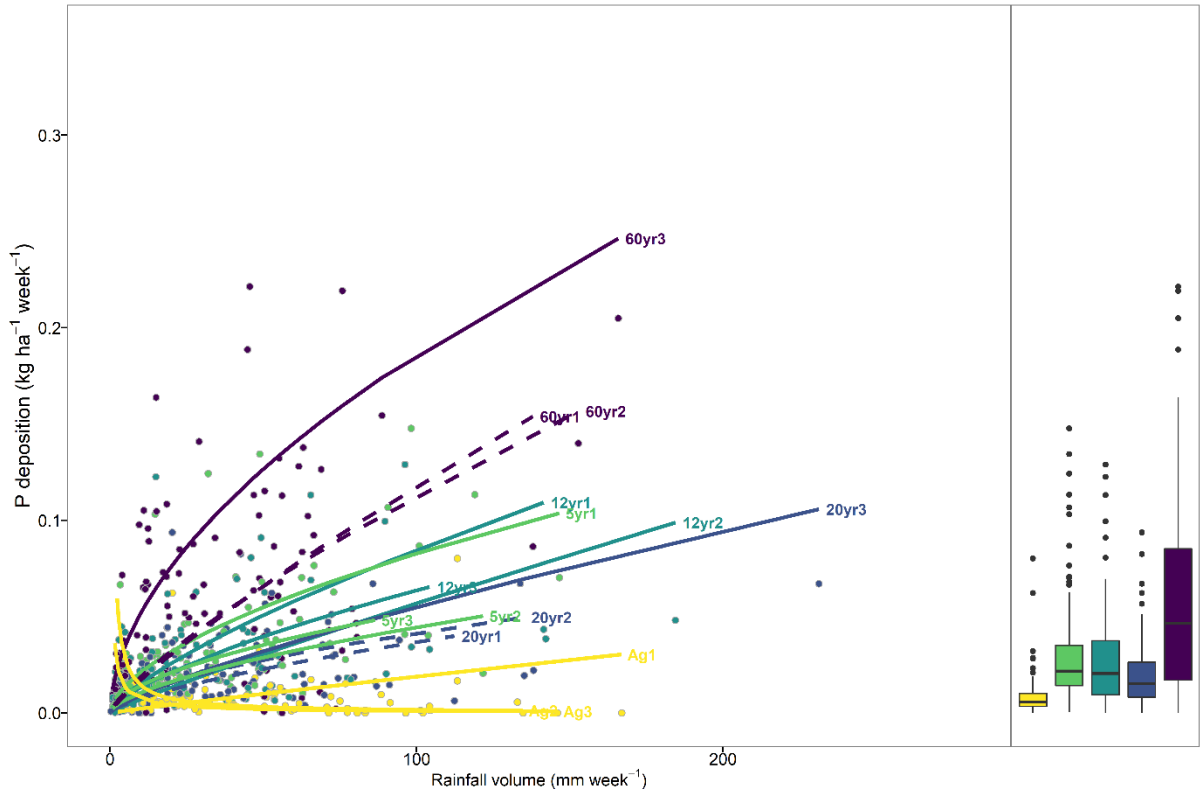
Fig. 3. Weekly rainfall amount (open field rainfall volume for agricultural fields, throughfall volumes for the forest stages) versus weekly phosphorus (P) deposition, in agricultural field (Ag), 5- (5yrs), 12- (12yrs), 20- (20yrs) and 60-year-old forest (60yrs). Solid lines show significant log-log fits via standardized major axis regression, while dashed lines indicate insignificant regressions (P-value>0.1). Boxplots show the median, the 25th and 75th percentile and the 1.5 interquartile range spread for the weekly P deposition loads. Points in represent the individual weekly measurements of composite samples (n=8) per plot for rainfall volume and P deposition.

161

**Forest canopies as temporary reservoirs for P deposition.** We used simulated black carbon surface mass density concentration (BCCMDBCSMC) from the MERRA2 model (Fig. 4dS2), to assess whether temporal P deposition patterns could be explained by temporal variations of black carbon in the atmosphere. However, we did not find a significant relationship between

7

166 the rate of P deposition and the BCSMC BCCMD over the site. Instead, weekly P throughfall

167 amounts – or post-dry-deposition P wash-off – were shown to be correlated with weekly

168 rainfall on a log-log basis across each site (back transformed relation shown in Fig. 3, Table

169 S4). While the BCSMC BCCMD temporal pattern, which is consistent with the aerosol

170 optical depth at 550 nm via MODIS' Terra satellite, show that the intensity of dry deposition

171 varies over the year (Fig. 4), significant correlations between P throughfall and rainfall

172 suggest that wash-off of dry deposition is controlled by the amount of rainfall available to

173 wash particles from the canopy rather than the timing of their arrival on the canopy. In other

174 words, P input to lowland forest floor is a transport-limited process (Fig. 3). As such, canopies

175 act as dynamic reservoirs where dry deposition accumulates over time, replenishing the forest

176 floor with an amount of deposited P in proportion to the amount of rainfall. In fact, our data

177 show that this is not only valid on a rainfall event basis, but also that canopies can act as year-

178 round stores for nutrients.

179

180 Overall, we found that central African forests receive P deposition loads that are substantially

181 higher than expected from model simulations and are sourced by biomass burning on the

182 continent. Additionally, canopy complexity is a major drivercouldis likely be a major driver

183 of nutrient inputs, where older and more complex canopies more efficiently trap atmospheric

184 constituents. This dry deposition trapping increases the bulk 'open field' deposition from 0.4

185 kg ha^{-1} yr^{-1} to 3.1 kg ha^{-1} yr^{-1} in old secondary forests, which means that forests are about

186 eight times more effective at capturing atmospheric P compared to non-forest ecosystems.

187 Local P deposition is a spatially heterogeneous process controlled by both on-site biotic

188 factors (e.g., canopy complexity), as well as regional-scale determinants (e.g., continental fire

189 regimes). Our estimate of total P deposition in older forests of the Congo basin is three times

190 the amount that would be needed to sustain tropical forest growth in steady state across the

tropics (1.1 kg ha^{-1} yr^{-1},[29]). Hence, the magnitude of these deposition loads raises the question

whether the widespread assertion of P limitation in old-growth central African forests

holds[30,31]. At present, biomass burning on the African continent is slightly lower than it has

been over the last millennia[32], which suggests that the magnitude of annual P deposition has

remained high over the same duration. However, given the likely increase in biomass burning

related to anthropogenic activities, P deposition is likely to increase in the coming decades.
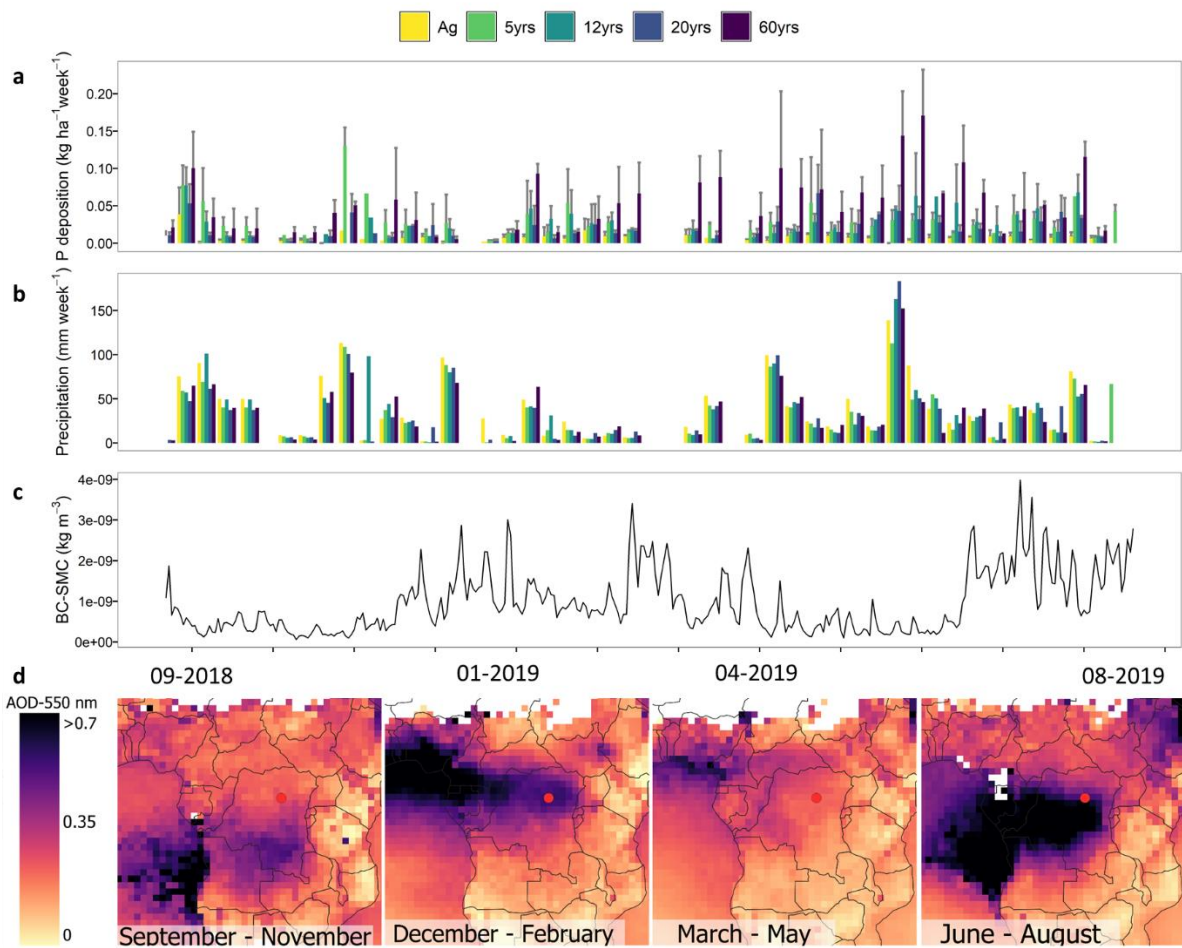
Fig. 4. Temporal trends for open field (Ag), 5 (5yrs), 12 (12yrs), 20 (20yrs) and 60-year-old forest (60yrs) over
the monitoring period for phosphorus (P) deposition (mean ± standard deviation) (a), weekly precipitation (b),
black carbon column surface mass density concentration (BC-SMD SMC) (c) above our site as extracted from
MERRA2. Aerosol Optical Depth (AOD) at 550 nm, signaling biomass burning-derived aerosols detected over
the study period by MODIS' Terra satellite at the larger region around the study site (red circle) (d).

Methods

Study site. The study was carried out in post-agriculture forests at different growth stages near the forest reserve of Yoko (N00°17′; E25°18′; mean elevation 435 m a.s.l.), situated between 29 to 39 km south east of Kisangani, in the Democratic Republic of the Congo. We set up 18 (40 by 40 m) plots, set out in triplicate along 5 successional stages (15 plots): 5, 12, 20, 60 years old secondary forest (respectively 5 Y, 12 Y, 20 Y , 60 Y). Additionally, soils were also characterized in 3 agricultural plots (AG). We interviewed owners, farmers and local experts to determine the time-since-disturbance of all plots. Tree height measurements were recorded at the plot level for 20% of individuals of each diameter class. The climax vegetation in the region is classified as semi-deciduous tropical. Climate falls within the Af-type following the Köppen-Geiger classification[33]. Annual rainfall ranges from 141~~8~~ ~~7.5~~ mm to 1915~~.4~~ mm with mean monthly temperatures varying from 23.7°C to 26.2 °C. Throughout the year, the region is marked by a long and a short rainy season interrupted by two small dry seasons December-January and June-August. Soils in the region are highly weathered Oxisols, being poor in nutrients, with low pH and dominated by sandy texture.

Sampling and sample analysis. Throughfall and bulk precipitation was collected weekly using polyethylene (PE) funnels supported by a wooden pole of 1.5 m height to which a PE tube was attached and draining into 5 L PE container. A nylon mesh was placed in the neck of the funnel to avoid contamination by large particles. The container was buried in the soil and covered by leaves to avoid the growth of algae and to keep the samples cool. We installed eight throughfall collectors in each plot as two rows of four collectors, with approximately 8 m distance between all collectors. On every sampling occasion, the water volume in each collector was measured in the field, and recipients, funnels and mesh were replaced, rinsed with distilled water. A volume-weighted composite sample of the devices per plot was made. All samples were stored in a freezer immediately and sent in batch to Belgium for chemical analysis. The volume-weighted composite samples were first filtered using a nylon membrane filter of 0.45 µm before freezing. Total phosphorus was measured by inductively-coupled plasma atomic emission spectroscopy (ICP AES, IRIS interpid II XSP, Thermo scientific, USA). Although we acknowledge ~~that~~ the potential for microbial activity in the collectors during a one-week, dark, *in situ* ~~preservation~~ storage of the samples ~~is suboptimal, we judge that~~ the use of total phosphorus concentration ~~after~~

233 ~~filtering is not likely to change a lot through microbial activity~~and lack of algal growth allow for complete

234 phosphorus recovery.

235

236 After analysis, the samples from the replicate field sites per forest stage were pooled into 'weekly' forest type

237 samples, and these were subsequently analyzed for dissolved black carbon (DBC). In short, the pooled water

238 samples were acidified to pH 2 and analyzed for dissolved organic carbon (DOC) concentration via high-

239 temperature catalytic oxidation on Shimadzu TOC-L total organic carbon analyzer following established

240 methodology[34]. DOC was isolated from the water samples by solid phase extraction (SPE) following Dittmar et

241 al.[35]. Briefly, SPE cartridges (Varian Bond Elut PPL, 1g, 6 mL) were conditioned sequentially with methanol,

242 ultrapure water, and ultrapure water acidified to pH 2 using concentrated HCl, then passed through the SPE

243 cartridges by gravity. SPE cartridges were dried under a stream of high purity N_2 gas. DOC was eluted from the

244 SPE cartridge with methanol (SPE-DOC) and stored at -20°C until further analysis. DBC was quantified using

245 the benzenepolycarboxylic acid (BPCA) method as detailed in Wagner et al. [20]. The BPCA approach to

246 quantifying DBC involves chemothermal oxidation of condensed aromatic DOC compounds to

247 benzenehexacarboxylic acid (B6CA) and benzenepentacarboxylic acid (B5CA) products. The B6CA and B5CA

248 oxidation products are robustly measured and derive exclusively from pyrogenic sources [36]. Condensed aromatic

249 DBC, as measured using the BPCA method, is ubiquitous in aquatic environments globally [21,37–39]. DBC has

250 also been quantified in throughfall and stemflow in longleaf pine forests that undergo regular prescribed burning

251 [40]. Therefore, we use the BPCA method as a proxy for carbon inputs from biomass burning in the current study.

252 ~~The BPCA approach to quantifying DBC involves chemothermal oxidation of condensed aromatic DOC~~

253 ~~compounds to benzenehexacarboxylic acid (B6CA) and benzenepentacarboxylic acid (B5CA) products.~~

254 ~~Briefly~~To analyze our samples for BPCAs, aliquots of SPE-DOC (~0.5 mg-C equivalents) were combined with

255 concentrated HNO_3 in flame-sealed glass ampoules and heated to 160°C for 6 hours. The resultant BPCA-

256 containing residue was dried and re-dissolved in mobile phase for subsequent analysis. Individual BPCAs were

257 separated and quantified using an HPLC system (UltiMate 3000, Thermo Fisher, Germany) (CV < 5%). Sample

258 DBC concentrations were calculated using the established power relationship between DBC (μM-C) and the sum

259 of B6CA and B5CA (nM-BPCA) using the equation from Stubbins et al. [41]: $[DBC] = 0.0891 \times ([B6CA +$

260 $B5CA])^{0.9175}$; n = 351, R = 0.998, p < 0.0001. We report the concentrations of B5CA and B6CA with DBC to

261 facilitate comparison with other datasets (Table S2).

262

Canopy 3D model. UAV surveys were carried out in Yoko on February 9, 2020. The Yoko survey consisted of 5 flights due to the dispersed locations of the 15 inventory plots and in total covered an area of 30~~21.7~~ ha (Fig. 23). Two UAV platforms were used in the surveys: (i) a consumer-grade DJI Mavic 2 Pro. This UAV was equipped with a Hasselblad L1D-20c camera (20 megapixels, 5184×3456 pixels, ca. 77° FOV). The onboard GNSS supports GPS and GLONASS. (ii) a customized DJI Phantom 3 Advance. We removed the DJI camera-gimbal system and mounted a GoPro Hero 3 camera (12 megapixels, 4000×3000 pixels, with 2.92 mm F/2.8 123° HFOV lens) and connected the camera to a RTK/PPK (Real-Time Kinematic and Post-Processing Kinematic) enabled GNSS receiver to determine camera exposure position at centimeter-level. In brief, the Mavic camera provides higher image quality but lower GNSS accuracy, while the GoPro camera provides accurate positioning with PPK solution but had lower resolution of images. The combination of both platforms provides high positional accuracy as well as high-quality images[42]. A 90% forward image overlap and 80% side overlap were used during the flights for both UAV/camera systems. The flight height was set at 180 m from the ground level, providing an average Ground Sampling Distance (GSD) of ca. 0.04 m px-1 for Mavic and 0.10 m px-1 for GoPro. During the flights, a Reach RS (Emlid Ltd) base station was mounted on a tripod placed in an open area within the surveyed region to provide correction data for PPK georeferencing. The coordinate of the base station was determined using the c. 8h average value of the single solution throughout the survey. The images of the GoPro were georeferenced using the RTKLib © software in PPK mode. This setup provides meter-level absolute accuracy but centimeter-level precision (relative accuracy). The images were then processed using the Pix4D Mapper software (https://www.pix4d.com/) where both image types were combined in a single SfM-workflow. The outputs (i.e. 3D point cloud, DSM and RGB mosaics had a centimetric precision (RMSE estimated at c. 0.04m)[42].

Canopy roughness and canopy rugosity calculation using canopy 3D model. To show a potential canopy trapping effect, we related the canopy 3D properties to the dry deposition loads in the different plots, using the UAV structure-from-motion 3D product. It has been shown that dry deposition velocity (v_{ds}) of particles is linearly related to the surface roughness length (z_0) [27,28], and a simple parameterization for various surfaces – across various biomes - has rendered following relation[27]:

$$v_{ds} = 0.617 \, log(z_0) + 1.77$$

Hence, comparing across the successional stages of our forest succession, the canopy trapping hypothesis should manifest through a consistently different v_{ds} along the chronosequence, if the canopy complexity varies in z_0. As

293   such, we aimed to determine z_0 along the chronosequence, in order to determine the v_{ds}, to see if this related to

294   the differences in dry deposition loads. Hence, z_0 was determined following Lettau[43] and De Vries et al.[44]:

295    $$z_0 = CH\lambda \ ,$$

296   where C is a constant – assumed 0.5 [44], H is the average obstacle height and λ is the obstacle density of the

297   roughness elements. H was derived based on field inventories and field-based measurements of tree height. In

298   turn, λ was determined following de Vries et al. and Li et al. [44,45] as:

299    $$\lambda = \frac{\sum \Delta y}{\sum \Delta x} \ for \ \Delta y > 0$$

300

301   In other words, λ integrates the positive Δy divided by the Δx over a certain transect of the canopy height model

302   of a certain experimental plot. This was done on 32 transects in the north-south direction of the plots, as well as

303   32 transects in the east-west direction for every plot. However, the calculation of λ over height transects is prone

304   to instrument noise in the canopy height models. To filter out this noise, the transect data can be smoothed by

305   applying a rolling means of a certain size, i.e. including a certain number of neighbor points along the transect.

306   To determine the best size of the rolling mean, the calculated lambda value can be plotted against the number of

307   measurements that are included in the moving average. The resulting slope of lambda versus the size of the

308   moving average has two components; a first steep descent, followed by a less steep stable slope. It is accepted

309   that the first steep descent is caused by filtering out unwanted noise in the data, while the second part is the

310   actual λ (Fig S3). Instead of detecting a breakpoint in this curve per plot, we qualitatively inspected the different

311   curves, and selected a general number of measurements to be included in the rolling average (19 points in total,

312   i.e. 9 points on both sides of the point itself) to determine λ, to avoid that an inconsistent rolling mean would

313   drive differences in the λ estimation across plots. The drone flights were performed six months after the end of

314   the monitoring period. We don't expect this to affect the forest canopy structure, but the agricultural field were

315   heavily overgrown already by then, compared to the bare fields they were at the beginning of the monitoring

316   period. For this reason we divided the standard deviation of the agricultural plots by three in Figure 2, for

317   visualization purposes, assuming a linear increase of variability over time, and targeting the average (after 6

318   months of monitoring) situation in the monitoring period.

319

320   Additionally, we calculated canopy rugosity[46]. In essence, this is a much simpler proxy that ~~simply~~ estimates the

321   standard deviation of the vertical variation of the upper canopy layer. For this, we used the transects that were

322   generated based on the CHM of the plots, as described above, ~~and used those transects~~ to calculate the standard

323 deviation of canopy height. The final 'rugosity' parameter is an average of the standard deviation over all the

324 different transects per plot.

325

326 **Data analysis.** We calculated weekly throughfall fluxes of P by multiplying the concentration of P with the

327 weekly recorded rainfall volume per plot. We aggregated this data to annual fluxes by calculating a weekly

328 average and multiplying by 52 weeks per year, to account for some missing values – in case a throughfall

329 collector was stolen, or sampling was logistically impossible that week (on one occasion). Up-to-date, it has

330 proven difficult to quantify net nutrient deposition in forest ecosystems, since most studies are based on

331 throughfall collection. This method uses rainfall collectors set-up in open-field, paired with collectors under the

332 forest canopy (i.e. throughfall collection). The measured deposition load in the open field is typically considered

333 'wet' deposition, while the throughfall collectors are considered to be the sum of wet deposition, dry deposition

334 and canopy leaching, minus canopy uptake of nutrients. Contrary to other direct quantification methods of dry

335 deposition[8,47], this method is attractive for its simplicity but also the potential to fully include canopy complexity

336 effects on the dry deposition budgets. However, the downside of the method is that it is complex to disentangle

337 dry deposition, from a canopy process such as leaching and uptake. There have been a variety of theoretical

338 models that attempt disentangling both exogenous and endogenous effects in throughfall deposition loads, but

339 these show varying success and results (for an overview, [48]). In this study, we approached this differently by

340 combining the deposition loads of a proxy for biomass burning (DBC) with the P deposition loads. Theoretically,

341 subtracting the weekly wet deposition, i.e. the deposition loads as recorded in the open field, from the throughfall

342 deposition load in the different forest types, results in the weekly net canopy effect, i.e. either canopy

343 uptake/leaching or the added dry deposition effect of the canopy. We did this for both DBC deposition loads as

344 well as P deposition loads, and subsequently assessed if the P deposition could be consistently predicted using

345 only DBC deposition loads across the plots. We performed standardized major axis (SMA) regression between

346 DBC deposition load and P deposition load, and between rainfall volume and P deposition. Where assumptions

347 of the model fit were violated, we logtransformed both variables and performed a log-log SMA regression, but

348 these were backtransformed for visualization purposes. SMA regressions were fitted using the 'smatr' package[49],

349 using the robust estimation method. All analyses were carried out with the R software[50].

350

351 **Wind trajectories and satellite data.** To show how biomass burning aerosols might be sourced throughout the

352 year across the African continent, we crossed a burnt area dataset with backwards wind trajectories ending at the

site, ~~much like was done in~~similar to previous ~~resource~~work at the same site [51]. The fire pixel dataset covering the entire African continent was obtained from NASA's MODIS Collection 6 NRT[52]. The backwards wind trajectories were generated using the Hybrid Single Particle Lagrangian Integrated Trajectories (HYSPLIT) model provided by the National Oceanic and Atmospheric Administration Air Resources Laboratory (NOAA ARL) [53–56] with the reanalysis 2.5 degree archive meteorological dataset. For the entire study period, we generated one trajectory at noon~~,~~ for every day, ending above the study site pixel at 500 meters above ground level, going back one week. Instead of displaying all the trajectories separately, we constructed a~~n~~ convex hull around the trajectories, to effectively show the potential source areas to be crossed by winds (Figure S3). Additionally, we extracted the hourly simulated Black Carbon Surface Mass Concentration (BCSMC) from above our study site from the Modern-era Retrospective Analysis for Research and Applications version 2 (MERRA-2) from the Goddard Earth Observing System Model, version 5 [57].

Uncertainties. This study reports some of the first *in situ* measured P deposition loads for central Africa. However, due to logistical constraints and local conditions, we had to design the monitoring setup in a robust and low-tech manner. These adaptations for the long-term monitoring might bring about some additional uncertainties we want to report here. First: the rainfall and throughfall collectors we used in the field did not have cooling (no grid power available, and safety issues in maintaining technological setups in the field), nor did we add preservation chemicals to the collection bottles (due to the environmental risks involved with the disposal of mercury, and the contamination of future analyses - thymol). Nevertheless, we judge that the consequences of this are minor for total phosphorus concentration in the samples.~~– For t~~The DBC quantification, by definition, was performed on filtered samples and thus~~: as we filtered the samples prior to analysis, we~~ did not include particulate BC (PBC)~~in these analysis.~~. However, we use DBC merely as a tracer for (dissolved) total P sourced by biomass burning. Given the pre-dominance of biomass burning as the source of combustion derived aerosols in the Congo (consistent source), ~~and~~ the long-range transport of aerosols to our study site, and the sub 0.2-micron size fraction of these aerosols, we are confident that ~~there is no reason to expect the proportions of~~ DBC ~~and PBC to vary substantially through time~~is a reliable tracer for combustion-derived inputs. Finally, we use 'wet' and 'dry' deposition throughout this manuscript, while in fact our wet deposition was not collected with 'wet-only' collectors. In practice, the open field depositional loads will also have dry deposition collecting on the collector surface. Hence, our 'wet' deposition estimates are slightly overestimating wet deposition, while the 'dry' deposition estimates are slightly underestimating actual dry deposition.

383

Data availability

The authors declare that all data supporting the findings of this study are available within the paper and its
supplementary information files.

387

Acknowledgements

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Author contributions

M.Bauters, T.W.D and P.B. conceived the study. M. Bauters and K.V.O. carried out the drone flights. K.V.O.
processed the drone imagery for the 3D canopy reconstruction. M. Bauters, C.E. and I.M. oversaw and carried
out field work, and set up the permanent sampling units. M. Bauters, T.W.D., S.Bodé and S.W. analyzed the
dissolved black carbon in the composite samples. M. Bauters, I.M., P.B. and S.Bodé analyzed the phosphorus in
the sample set. I.M., C.E., L.C., M. Barthel and S. Baumgartner provided logistical support for the field work.
M.Bauters wrote the paper, with significant contributions from T.W.D., S.W., K.V., H.V., P.B. and K.V.O.

410

Competing interests

The authors declare no competing interests.

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Additional information

Supplementary information is available for this paper at XXX.

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Correspondence and requests for materials should be addressed to M. Bauters

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REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

This manuscript has improved as the authors took into consideration the remarks put forward but the reviewers. There are still two points that need to be clarified before a possible publication of this work: 1) As indicated by reviewer 2, the rain collectors are not collecting only wet deposition but a quantity linked to total deposition. This should be more clearly indicated in the text since any later attempt for modellers to separate dry and wet deposition is not possible.

2) Line 163 the authors indicate that Figure 4d represents surface concentration of Black Carbon (BC) when the Figure as it stands shows Aerosol Optical Depth @ 550nm. The authors should either correct the text or update the Figure.

Reviewer #3 (Remarks to the Author):

In this revised version of the ms all the issues and concerns raised by the reviewers have been sufficiently addressed and the ms was improved. The point I raised on rugosity has been nicely incorporated. I agree with the authors response to Reviewer #2's methodological concerns, that these concerns have minor influence and do not compromise the results.