Responses to reviewer comments 'Long-range impacts of biomass burning on $PM_{2.5}$: a case study of the UK with a globally nested model' by Tan, D.Y.T. et al.

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Response to RC1

We thank the reviewer, Dr Tobias Osswald, for the time spent reading our manuscript and putting together these useful comments. We are pleased to read their assessment that this work is "quite important, well structured and of great scientific quality" and that "it should be published." Below, their comments are copied in italic. We respond on a point-by-point basis (in normal font), indicating the revisions we have made. All line numbers in our responses refer to the revised manuscript.

General Comments

This paper contributes to our understanding of the transport of BB emissions across the world, focusing on a country that has very few forest fires, but that still seems to be impacted by them. It is significant for the society at large, and points out that even countries like the UK, are unexpectedly affected by wildfire smoke, coming not only from Europe, but also Asia and North America. As such I consider the topic to be relevant. The authors focused on PM2.5, a valid choice, as this pollutant is well known to be a risk to human health and is associated with BB.

Response: We are pleased to read that the reviewer assesses our study to be relevant in both topic and choice of pollutant.

The method relies on a chemistry transport model (CTM) called EMEP4UK, which was run for a total of 2 years in a nested configuration, from global to national level, that represents a notable computational effort (How many cells in the horizontal directions were used for each of the domains? I find this information useful for the reader to get an idea, please include this in the text).

Response: Domains A, B and C (see Figure 1) contain 360×180, 199×169 and 369×447 grid cells, respectively. This has been added in L356 in Appendix B.

Several runs were carried out where each one considered the BB emissions from different regions of the world. A base run without BB emissions was also carried out. The differences in concentrations between the different runs allowed the authors to assess which regions of the world contributed the most to the air quality degradation in the UK. This is a smart and thorough method and a great advantage of CTMs in studying these phenomena. It allows the authors to consider higher-order effects taking part in the dispersion and chemical transformations of smoke in the atmosphere.

Response: We are pleased to read that the reviewer judges our methodology to be valid and thorough.

The methodology section often points to appendix B, where a deeper description of the model and its comparison to air quality stations is available. The EMEP4UK global model version used here had not been used elsewhere before, and so should in my opinion have a more prominent place, not in the appendix but in the methodology section. Its comparison to real world data is also very relevant for the paper.

Response: Whilst there could be merit in moving the content of Appendix B to the Methodology section – given that this configuration of EMEP4UK has not been used elsewhere – we have decided to retain it in a separate section for two reasons. Firstly, the content of Appendix B is long, so moving it would double the length of the Methodology section and substantially unbalance the sections of the paper. Secondly, while this novel modelling setup is fundamental to the results we present, we believe that moving Appendix B to the Methodology section would interrupt and distract from the main narrative of our paper, which is to highlight the impact of distant biomass burning (BB) emissions on regions with little BB activity.

There is one possible flaw in this methodology however, which was not pointed out. Plume rise is not being modeled at all, as is done for example in CHIMERE or other CTMs, where a plume rise height is calculated and a plume rise profile applied. Instead the smoke is assumed to be homogeneously mixed in the ABL as soon as it is released. I wonder if the authors can defend their methodology in this regard in their reply to this review? Weather this is possible or not I think that some considerations about this topic should be done in the paper.

Response: In considering this comment, we realised we stated the wrong BB emissions distribution mechanism in the Methodology section, for which we apologise. This has now been corrected in L105 to read "BB emissions are [...] evenly distributed from the surface up to 800 hPa". This approach loosely follows recommendations by Sofiev et al. (Simpson et al., 2012; Sofiev et al., 2009). The reviewer's comment still applies despite this correction. A plume height profile could not be applied because the BB emissions dataset used here (FINNv2.5) does not include pre-calculated plume and injection heights, nor does the EMEP4UK model have a functionality to calculate plume rise. Adding such a functionality would require significant changes to the model code, beyond the scope of this study. Importantly, however, our BB emissions are not all emitted at the surface but over altitudes that include to the top of the

ABL or higher. Furthermore, evidence indicates that while a more sophisticated injection height can improve model performance at a regional scale, this becomes less important in the case of long-range transport and moving towards climatological timescales, where meteorology becomes a more important factor (Field et al., 2024; Whaley et al., 2025). This suggests that a more sophisticated BB emissions dispersion mechanism is less important for our study, where we focus on the long-range, annual mean impacts of BB (though the domestic component to which we compare the long-range component may be more sensitive to this). The following sentence in the 'Study caveats' section has been added to acknowledge this issue (from L298): "Choice of the BB emissions dispersion scheme is also an important factor, particularly on a regional scale, though this has been found to be less important when considering long-range transport and longer time-scales (Field et al., 2024; Whaley et al., 2025), as is the case in this work."

In the discussion the authors went into a higher detail and assessed the secondary production of PM2.5, its chemical composition, the population-weighted average concentrations among others. I would prefer a more focused and direct approach, discussing only the results that are actually relevant for the main conclusions of the paper. Particularly in the discussion on sections 4.2 and 4.3 I struggled in understanding the concepts and the plots presented by the authors. When the authors mention area-weighted average, they mean what is the most intuitive average (on the other hand, an average value that was presented as such with no further description and that was not area-weighted would be misleading, would it not?), and so I would remove the term area-weighted. I would also remove most of the data and discussion about the population-weighted average. Though this can be mentioned once, as it may be interesting as a first-estimate of the impact on human health, it seems to me that this is not the focus of the authors, which is why there is no mention of this in the conclusions. If the authors do not agree with me on this then I would ask them to explain to me in their reply why this is relevant in this paper, and to what degree.

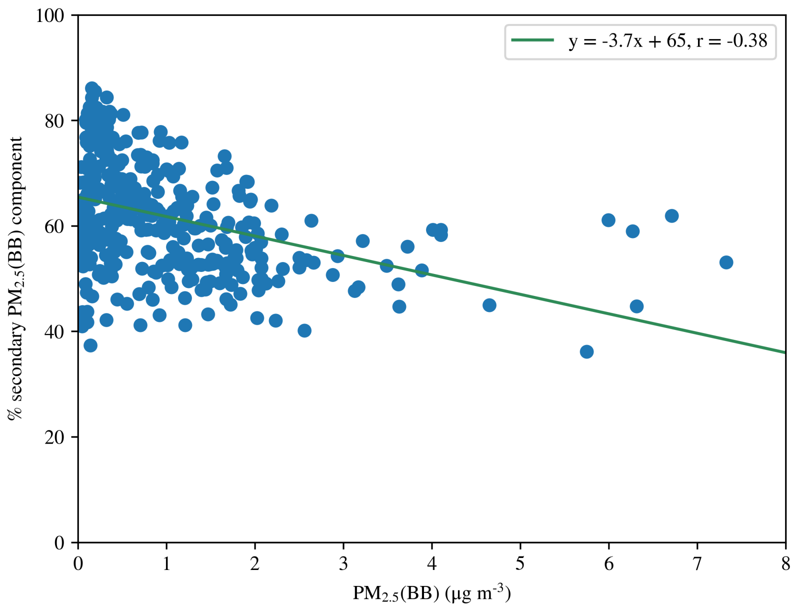
Response: The term 'area-weighted mean' has been removed, and is now simply referred to as the 'mean' - we agree that this is widely assumed to be the default method for calculating a spatial mean. In order to simplify Figure 4b, we have removed the red line representing the daily population-weighted mean, and any associated text in the caption and main text that referred to these. We have, however, retained the population-weighted version of the annual mean in Table 1, and the instances in the text where it is discussed (Section 4.2), because we believe the population-weighted annual mean provides useful information about the human health implications of this study. Firstly, the greater value of the population-weighted mean shows that larger amounts of $PM_{2.5}$(BB) coincide with the more densely populated areas of the UK; secondly, the population-weighted annual mean can be used to gauge progress against the WHO guideline for $PM_{2.5}$, and towards nationally and internationally agreed exposure reduction targets which are often based on the population-weighted mean.

Also in the discussion a particular episode in April was pointed out. During that month there was an abnormally high concentration not only of PM2.5 (BB), but also *from other sources. I am curious to know what was the reason for this. What caused the episode of bad air quality from other sources and where did the BB emissions come from? I do not think that more than a sentence should be added to the text, as this is not the main point of the paper, however, being an abnormal peak, I think that a concise (but still deeper than the current one) explanation of it would be better.*

Response: The elevated April episode of $PM_{2.5}$ pollution (non-BB sources) is the result of meteorological conditions that increase long-range transport of $PM_{2.5}$ and its precursors from continental Europe, and reduce dispersion, similar to spring episodes in 2003 (Vieno et al., 2014) and 2014 (Vieno et al., 2016). The modelled episode has a large particulate nitrate component, which is often found to peak in early spring due to easterly air flow (Abdalmogith and Harrison, 2005; Charron et al., 2013) and increased agricultural emissions of ammonia at this time of year (Vieno et al., 2016). The original text from L219 has been amended and expanded to read as follows. "The $PM_{2.5}$(BB) contribution is superimposed on an already elevated episode of $PM_{2.5}$ pollution caused by the easterly air flow conditions that increase long-range transport of $PM_{2.5}$ and its precursors from continental Europe into the UK, together with reducing dispersion, as per analyses of previous spring-time episodes (Vieno et al., 2014, 2016). Particulate nitrate in particular is often found to peak in early spring in the UK due to easterly air flow (Abdalmogith and Harrison, 2005; Charron et al., 2013) and increased agricultural emissions of NH_3 at this time (Vieno et al., 2016). The majority of the $PM_{2.5}$(BB) component during this episode is associated with BB in the model's European domain (predominantly from eastern Europe and the western areas of Russia also included in that domain)."

In line 229 I could not understand how the authors could see from the plots the tendency they mention. Could I have this explanation in the reply to this review, and if necessary also in the paper?

Response: Our statement is based on the observation that, across the full year of data, the top of the purple shading in Figure 4d, which represents the proportion of $PM_{2.5}$(BB) that is primary, is approximately correlated in their respective lower and higher values with the blue line in Figure 4b, which depicts the $PM_{2.5}$(BB) concentration. The corollary is therefore that the proportion of secondary $PM_{2.5}$(BB) tends to be greater when $PM_{2.5}$(BB) concentration is lower. This observation is most evident during the winter months (January, the first half of February and a lot of November and December) where $PM_{2.5}$ is lowest (Figure 4b) and Figure 4d shows that the secondary component is generally higher than in warmer months. The scatter plot below between daily percentage secondary contribution and daily mean $PM_{2.5}$(BB) concentration confirms this anti-correlation.



To further support our statement in the paper, we have amended the sentence in question (L233) to read: "Figures 4b and d show a tendency for the lowest concentrations of $PM_{2.5}$(BB) to have a larger proportion of secondary aerosol (confirmed by a scatter plot of daily percentage secondary contribution vs daily mean $PM_{2.5}$(BB), not shown)."

In section 4.3 an interesting extrapolation of the data to other years is carried out. However I would not put too much emphasis on it as it may have significant errors due to the presented assumptions and may also propagate errors inherent to the main methodology for 2019. For this reason I think that the text here should be more concise (about half the characters). Also in this section one of the assumptions that is correctly identified is that the proportions of the emitted pollutants stay constant (I believe the issue here is because other pollutants can impact the PM2.5 production or destruction, correct?), and then in parentheses "there is no reason why these proportions should vary substantially". Possible reasons would be changes in fire intensity and fuel load. I would say that there are reasons for why this could change substantially. However, the authors' assumption is also reasonable, since they are accepting that the extrapolation has its limits, but is still as good of an estimate as possible. I would therefore remove the sentence within parentheses.

Response: Yes, the reviewer is correct in their interpretation that the spectrum of pollutants emitted from BB can impact the $PM_{2.5}$(BB) at long distance; for example, the extent of secondary NH_4NO_3 production is impacted by BB emissions of CO, VOCs and NO_x. We thank the reviewer for pointing out potential reasons for why the proportions of these emissions could vary from year to year. We have followed their suggestion to remove the text within the parentheses.

Regarding the length of Section 4.3: the aim of this section is to put the year of study (2019) in the context of other years, to understand whether or not 2019 was an anomalous year for $PM_{2.5}$(BB) over the UK. We agree that there should not be an over-emphasis on the accuracy of this extrapolation, but we need to ensure the reader is clear about the assumptions that underpin our extrapolation, so it is essential we include these. The rest of this section is devoted to explaining the algorithm used, again so the reader can clearly understand what we have done, and a comparison of our results to literature. The latter is helpful because it allows us to demonstrate that our estimation for one of the years is consistent with previously published data, providing some confidence in our methodology. We believe that Section 4.3 is an important and integral part of our work and do not think that its length is out of balance with other sections. We have experimented with moving some of this section to the Methodology but prefer that the description of the extrapolation to other years is kept alongside the resulting data to avoid extensive cross-referencing.

In the conclusion the authors point out how relevant it is to mitigate anthropogenic emissions, as these will have an impact in PM2.5 (BB), the latter being a significant driver of air quality in the UK. This section once again points out the relevance of this work. In line 324 it is pointed out that the importance of PM2.5 (BB) is likely to increase in regard to total PM2.5. This is because there will be more burning and less or equal anthropogenic emissions. However, could the decrease in SIA counterbalance the increase in BB emissions? Could this effect be so strong that the relative impact of BB emissions to anthropogenic emissions on air quality remains the same? I don't expect that this can be answered without further calculations, but I would like to know the opinion of the authors in reply to this review.

Response: This is a highly relevant question and one that occurred to us as well. Some sensitivity experiments were carried out using the global configuration of EMEP4UK as part of the work for a previous paper (Tan et al., 2025). In that paper, experiments were carried out in which FINNv2.5 emissions were reduced. Over the areas of the UK where NH_4NO_3 conditional on BB is significant, there was an approximately linear response between reductions in total $PM_{2.5}$(BB) and reductions in the NH_4NO_3 conditional on BB. This indicates that the BB emissions were the limiting factor for NH_4NO_3 formation conditional on BB. If anthropogenic emissions of NO_x and/or NH_3 are reduced, we expect the BB emissions to remain the limiting factor down to a threshold, below which the anthropogenic emissions then become the limiting factor. We suspect a large reduction in NO_x and/or NH_3 would be required to reach this threshold, since this area of the UK is both NO_x- and NH_3-rich. However, more sensitivity experiments would be required to verify this and to determine the aforementioned threshold value, which would require substantial computational effort and goes beyond the scope of this study. We will retain this thought for further research into this topic and thank the reviewer for this insight.

In our revised paper, we have replaced 'would' with 'may' in L321 as it is not clear how large a reduction in local anthropogenic NH_3 and/or NO_x emissions is required for this to start reducing the SIA component that is conditional on BB emissions. In addition, to acknowledge that further research is needed, we have expanded the final paragraph (L332) to read: "..., whilst conventional anthropogenic sources will likely remain static or decrease further. (It remains unclear, however, if, or to what extent, any potential associated reductions in the SIA component conditional on BB may mitigate the increase in the non-SIA component.)”.

Technical Aspects

There were other, more technical aspects of the paper where I would like to point out some issues I found and that I think should be addressed:

Line 123: The phrase that begins here is ambiguous, hard to understand. Could it be that the authors are trying to fit two sentences into one? Could you explain what is meant here on your reply and rephrase in the text.

Response: As suggested we have split the sentence into two so that each sentence now describes each post-model run subtraction in turn. The text now reads as follows (L123). “Concentrations conditional on BB in the European domain illustrated in Figure 1 were calculated by subtracting the NEBB model run from the BASE model run. Concentrations conditional on BB in the UK were calculated by subtracting the NUBB model run from the BASE model run.”

Line 125: The population-weighted means seem out of place here, and indeed they are not really part of the sensitivity study. However, as previously noted, this variable should probably be omitted altogether.

Response: All text regarding daily population-weighted means has been removed, see response to the related comment about this under ‘General Comments’.

Figure 2: This way of showing the numbers of the regions is not the most intuitive, but it does its job. A legend box with colored squares next to the numbers would be more natural.

Response: Figure 2 has been amended according to this suggestion.

Line 142: Areas outside... This is already in the figure description, and is not relevant for the main text in my opinion.

Response: This sentence has been deleted.

Figure 4: I found this figure confusing, I may be misinterpreting some things. However I would like to see a new version that I can understand more easily. Perhaps changing labels and the description of the figure will suffice.

Response: Changes have been made based on the individual points raised below, as well as minor edits to the captions of Figure 4 and Table 1, and to the main text, which we believe now make Figure 4 easier to understand.

Figure 4a: What is meant by Total in the y-axis? How is this different from the concept of Mean in the y-axis of figure 4b? I found overall the usage of the word "total" confusing in figure 4. I also fail to understand where I can see the non-BB PM2.5. Are the blue and gray areas stacked upon each other? Also in the text line 144 is confusing for the reasons mentioned above.

Response: The y-axis in Figure 4a is the daily mean $PM_{2.5}$ concentration. The word 'total' was intended to emphasise that this is the $PM_{2.5}$ from all sources, not just associated with BB. The mean was calculated in the same way as the blue line in Figure 4b (i.e. standard area averaging over the UK) so, yes, the blue bars in Figure 4a represent exactly the same data as the blue line in Figure 4b. To avoid any ambiguity, we have re-named the y-axis in Figure 4a to read "Mean $PM_{2.5}$".
The non-BB components are the grey bars, which are stacked on top of the blue bars, so the $PM_{2.5}$ from all sources is the blue and grey bars together. We have amended line L145 (and similarly the figure caption), to state this explicitly: "... and all other contributions to $PM_{2.5}$ in grey stacked on top ...".

Figure 4b: What do the percentiles represent? How were they calculated? Are they calculated from the daily means of each cell? Is the mean for the domain done at each time-step and the percentiles calculated from those values? Is it something else? I would also remove the red curve.

Response: The daily means for all grid cells over UK land were averaged to give the UK daily mean values represented by the blue line in Figure 4b. This is for each day, not each model time step (hourly). The percentiles for each day are derived from the set of daily means across all the grid cells that are over UK land. For both the daily means and percentiles, only the grid cells covering the UK were used, not what is defined as the UK domain (which includes the Republic of Ireland and some ocean). To improve clarity, we have added the following to L150. "... and the 5th to 95th percentile envelope (light shading) of the daily mean $PM_{2.5}$(BB) values across all the model grid cells over UK landmass". The caption of Figure 4 has been changed similarly. The red line in Figure 4b has been removed, as per our response to previous comments relating to these data.

Figure 4f: the chosen gray and purple are hard to distinguish at first glance

Response: The grey colour representing '2018 BB' has been changed to a paler shade of grey, to enhance the distinction between the two colours. The corresponding colour in Figure 5 has also been updated.

Line 181: remove the sentence that starts with "To the author's knowledge..."

Response: We would like to keep this sentence, to explain why we have not compared this value to others published in the literature.

Line 186: Why 31/42? Is this relevant? The uncertainty of this value is probably *more than 1% and so I think that 74% is an adequate enough representation of this fraction, and easier to grasp.*

Response: Here we were trying to give an example calculation of how the 74% was derived from the values in Table 1. We have added words to this sentence so that it now reads "...constituting a proportion of 31/42, or 74%, of..." (L186).

Line 280: "Error bar" is a term more widely used for the graphical representation. perhaps "margin of error", or some other term would be more appropriate.

Response: The term "error bar" has been changed to "uncertainty range" (L285).

Response to RC2

We thank the reviewer, Dr Jie Zhang, for the time spent reading our manuscript and putting together these useful comments. Below, their comments are copied in italic. We respond on a point-by-point basis (in normal font), indicating the revisions we have made. All line numbers in our responses refer to the revised manuscript.

Summary

In this study, Tan et al. used a novel, globally nested atmospheric model to quantify the long-range impact of global biomass burning (BB) on PM2.5 pollution in the UK. The research reveals that in 2019, BB contributed a significant 0.99 μg m^{-3} *to the UK's annual mean PM2.5, accounting for 10% of the total concentration and 20% of the WHO's annual guideline. Crucially, the study demonstrates that this impact is overwhelmingly due to long-range transport, with 73% of the BB-conditional PM2.5 originating from outside of Europe, primarily from Russia, Asia, and boreal North America. Furthermore, the analysis shows that over half of this pollution is secondary aerosol formed during transport, including ammonium nitrate created when oxidants from distant fires interact with the UK's own domestic emissions. The findings highlight the insufficiency of regional-only air quality models and underscore the critical importance of considering global sources in national pollution policy, suggesting that local emission reductions can play a role in mitigating pollution from distant sources. The manuscript is well-structured, clearly written, and the figures are of high quality, effectively communicating the key findings. The conclusions are well-supported by the evidence presented. This study is a significant contribution to the field and will be of high interest to the atmospheric science community and air quality policymakers. I recommend its publication after the authors address a few minor points for clarification and discussion.*

Response: We are pleased to read the reviewer's very supportive comments on the quality and significance of our paper, and for their recommendation that it be published following response to some minor points of clarification.

Minor comments

1. Regarding the BB emissions inventory uncertainty mentioned in Section 4.4, the authors should consider expanding this point. Given that FINNv2.5 is often a high-end estimate compared to inventories like GFED, a brief discussion on how this might affect the magnitude of the calculated 0.99 μg m^{-3} *contribution would be valuable. Additionally, a brief note on how potential differences in the spatial patterns between inventories could influence the source-region attribution would strengthen the paper's conclusions.*

Response: We agree that it would be useful to have a comparison to model simulations that use other emission inventories such as GFED or GFAS, which are likely to differ in the spatial pattern, timing and magnitude of emissions, thus affecting both our annual mean results and source apportionment. We have expanded on this in L295: "It will also vary with the choice of anthropogenic and BB emissions datasets (which may have different spatial patterns, timings and magnitudes of emissions)."

We would not, however, like to speculate (qualitatively or quantitatively) on exactly how changing the BB emissions dataset might impact our results. There is some evidence indicating that BB inventories with higher emissions (but similar temporal and spatial emission patterns) may lead to higher concentrations of particulate matter (Tan et al., 2025; Vongruang et al., 2017). However, other research suggests that inventories with larger net BB emissions do not always translate to higher concentrations (Scheffler et al., 2025). It is also important to note that Vongruang et al. (2017) and Scheffler et al. (2025) consider episodic events and consider a smaller spatial scale, which is fundamentally different from our study on annual, long-range effects - and therefore may not be applicable. Ultimately, considerable additional model runs would be required to confidently make even a qualitative statement about the impact of a different BB inventory. We envision that in the future the 'HTAP3 Fires' project (Whaley et al., 2025) will provide more clarity on this question, as one of its objectives is to better understand the effect of differing BB inventories on modelled pollutant concentrations.

2. Consider replacing the term " 'brute force' model experiments" (Line 165) with a more formal alternative like "direct perturbation simulations" or "simplified source-receptor experiments."

Response: The term 'brute force experiment' is frequently used when describing modelling setups similar to ours (for example Bartík et al. (2024); Burr and Zhang (2011); Kelly et al. (2015)) and we would therefore like to retain this terminology. However, to avoid ambiguity, we made an addition to L164 so as to provide the reader with an explanation of what we mean here by 'brute force': " – in which all relevant BB emissions are switched off in a given model perturbation run."

Congratulations on a very well-executed and important piece of research. Hopefully, these minor suggestions are intended to help the authors finalize an already excellent manuscript for publication.

Response: Thank you!

Literature cited in the responses

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