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## Responses to Referees' Comments

### Summary for the Editors

Insightful and constructive comments from both Referees helped us to improve the manuscript. Below, we summarize the main changes in the revised manuscript in order to address Referees' major concerns.
Following Referees' suggestion:

1. In the Abstract, we clarified those terms that were not clear.

2. In Section "Methodology", we clarified the description of sensitivity simulations and our choice in terms of spin-up and run period (Sect. 2.2).

3. In Section "Results":

   (a) We completed model evaluation by (a) comparing ERF from NASA ModelE2-YIBs with IPCC AR5 values, and (b) by computing the linear correlation Pearsons coefficient (Pearsons $R$), the Pearsons R squared ($R^2$) and the root-mean-squared error ($RMSE$) between model output and global datasets (i.e., MODIS AOD and the FLUXNET-derived GPP product) (Section 3.1).

   (b) We re-organized Section 3.2 to discuss separately changes in aerosol pollution (now Sec. 3.2), surface solar radiation (now Sec. 3.3) and surface meteorology (now Sec. 3.4), and, at last, changes in land carbon fluxes (plant productivity in Sec. 3.5, and isoprene emissions in Sec. 3.6). In each section, we firstly present changes at the global-scale, afterwards we focus on changes in five key regions: eastern North America, Eurasia, north-eastern China, the north-western Amazon Basin and central Africa.

   (c) To clarify discussion of results at regional scale and different mechanisms, we inserted Table 5 that presents, for each of the five key regions, absolute and percentage changes in annual average surface radiation, canopy temperature, GPP and isoprene emissions. For the five key regions, absolute and percentage changes in seasonal averages are reported in the Supplementary Material (Table S3 and Table S4).

   (d) We discussed more in details changes at the leaf level in Section 3.4.

4. We indicated more clealy the main limits of our approach, including the opportunities and challenges to linking aerosol-driven changes in surface meteorology to the changes in land carbon fluxes.

In the following, we address Referees' comments separately in a point-by-point response.

## Responses to Referee #1's Comments

We thank Referee #1 for their time and consideration. We closely considered the insightful and constructive comments from Referee #1. Referee #1's comments have helped us to improve the manuscript.
Referee #1's comments are quoted in italics. Authors' answer follows referee's comment in regular font.

### Response to Specific Comments

**1. Additional experiments***: It would be quite interesting to see the model response to coupled changes in the aerosol emissions, e.g., 50% increase in industrial vs. 50% decrease in biomass burning, and vice versa. I think this would add another dimension to the paper, but shouldnt limit the papers acceptance if the model simulations are too time-consuming to perform.*

**Authors**: Referee #1 suggests exciting ideas for future research. We do plan to explore the impacts of other changes in aerosol emissions according to possible future scenarios, i.e., increases in some sectors and simultaneous decreases in other sectors. This follow-on work will use the full land carbon cycle version of NASA ModelE2-YIBs. This paper is already rather long in its goal to examine the effects of all anthropogenic pollution emissions, biomass burning aerosols and industrial aerosols on plant productivity. Moreover, additional simulations are way beyond the scope of the present study because we have already used up the computational resources allocated for this paper.

**2. Abstract***: There are couple of instances where a little more explanation is needed so that a reader can clearly understand the full details of the study. For example, on a first read Im left wondering what "complex canopies" are, and exactly what you mean by "cooling in the Amazon Basin". It is clear when you read the manuscript further, but not when you read the abstract alone. I suggest the authors just take a little more time to carefully improve the abstract.*

**Authors**: Following Referee #1's recommendations, we clarified the Abstract by: (a) replacing "complex canopies" with "forested canopies" (pag. 1, ll. 9), (b) removing the term "cooling" and by clarifying the explanation of mechanisms operating over tropical biomass regions (the Amazon Basin and central Africa) (pag. 1, ll. 15–17), and (c) stating upfront the feedbacks accounted for in the model framework (pag. 1, ll. 4–6):
"The model framework includes all known light and meteorological responses of photosynthesis but uses fixed canopy structures and phenology." (see Point 8 below).

**3. Page 25440, lines 3***: Section number missing.*

**Authors**: Section number (2.1.1) has been inserted (pag. 5, ll. 152).

**4. Page 25441, lines19–23***: I had to read this twice first time around. I would bullet point these different simulation types and indicated more clearly that different emissions are removed (e.g., "all anthropogenic emissions including biomass burning are removed . . . " and ". . . biomass*

*burning emissions only are removed . . . " etc.)*

**Authors**: To clarify the paragraph that describes the sensitivity simulation types, we follow Referee #1's suggestion and use an itemized list where, for each simulation, the removed emissions are more clearly stated (pag. 7, ll. 209–213).

**5. Section 2.2***: I can see why understand why a long simulation is needed, but a little more justification on why the first 12 years (& not 10 or 15 years) are discarded, and why only for example the last 20 years (& not 30 years) are used.*

**Authors**: To clarify our choices in terms of spin-up and run period, we added the following sentences: "The global atmospheric oxidant-aerosol composition usually takes about 2 years to spin-up, while the atmospheric dynamics and land-surface climate takes about 10 years to reach steady-state due to an imposed aerosol radiative forcing. Therefore, we discard the first 12 model run years as spin-up. The remaining 20 model run years are averaged for analysis. Twenty model years of data are necessary such that any aerosol-driven variable differences between the control and sensitivity simulations are statistically significant relative to internal climate model variability." (pag. 7; ll. 224–230).

**6. Section 3.2, Page 25445, line 2***: I may be interpreting Table 1 wrong, but simNObb has a notable effect on SOA ACB reducing it from 1.37 to 1.14.*

**Authors**: Referee #1 is correct. Removal of biomass burning reduces SOA ACB by 17%. Hence, we completed our comments on Table 1 and highlighted the contribution of biomass burning emissions to SOA ACB by 17% (pag. 10, ll. 329–331).

**7. Section 3.2.2***: I think that it would be prudent to discuss changes in leaf temperature actually here (& included figure S11 or S12 in the main manuscript) in preparation for the following discussions on isoprene emissions. Or maybe have a new section 3.2.3 instead.*

**Authors**: Following Referee #1's suggestion, we modified Figure 5, which previously showed changes at the surface in atmospheric temperature (SAT) and relative humidity (RH), with a new version of Figure 5 that presents changes in transpiration efficiency (i.e., a proxy of canopy conductance), RH and canopy temperature. Relying on these new graphics, in the tropical regions we find evidence for a bio-meteorological feedback, which we discussed in detail in Sec. 3.4. By enhancing plant productivity, anthropogenic pollution aerosols increase canopy transpiration. In the north-western Amazon Basin, biomass burning aerosols drive the largest increase in transpiration efficiency, resulting in the largest decrease in canopy temperature (pag. 14, ll. 458–467). Annual changes in SAT are still commented on in the manuscript (pag. 14; ll. 442–451), and shown in Figure S3 the Supplementary Material.

**8. Section 4***: I appreciate the authors discussion on the studys limitations. But I think not allowing leaf phenology to respond to the changes in aerosol emissions is a significant issue. Canopy inputs are changing and thus ecosystems will respond accordingly. Is there anyway this effect can be quantified in this system set-up or has its likely magnitude been quantified in a similar study. I think this problem should also be directlymentioned in the abstract as well.*

**Authors**: While we agree with Referee #1 that interactive leaf phenology and land carbon allocation may influence the vegetation response to aerosol emissions, we emphasize that this

was a deliberate choice on our part for this first study that focuses on GPP and isoprene responses only. The feedbacks between vegetation and the atmospheric aerosol composition are extremely complex and challenging to disentangle. We decided it was useful and sensible to quantify the GPP and isoprene sensitivity to pollution aerosol emissions in a model framework that includes all known meteorological and light responses of photosynthesis, but that uses fixed canopy structures and phenology i.e. no feedbacks from dynamic carbon allocation or phonological timing allowed in this first study. **The goal is to provide a benchmark for future research that will add in the dynamic LAI and phenology feedbacks**. The model framework does include feedbacks from the meteorologically-altered vegetation carbon, water and energy fluxes on the atmospheric aerosol composition. We assert that the only real way that dynamic carbon allocation would potentially substantially influence the GPP response to aerosol emissions is through LAI changes, which would be a positive feedback, implying that our current results are underestimates and/or at the low end of the range of sensitivities. To our knowledge, there is only one existing published study that applies prognostic LAI [1]. However, that study did not isolate the effects of prognostic LAI. Our recent work with the YIBs land carbon cycle model has suggested that changes in the growing season length between 1982–2011 due to all global change drivers has little impact on regional carbon uptake and BVOC emission fluxes [2]. Therefore, it seems unlikely that the aerosol-induced rapid-adjustment meteorological changes described in detail in Section 3.3 would have substantial impacts on phenological timing and growing season length. Even if they did, the impacts on annual average GPP would apparently be minor. That said, our follow-on research is applying similar simulation methodology to assess the impacts of aerosol emissions on GPP, isoprene and other land carbon and water fluxes using a fully dynamic land carbon cycle model with interactive carbon allocation and prognostic phenology. Since the reviewer has raised important interest, we will endeavor to isolate the effects of individual global change drivers in this future research.
In the Abstract, we add the following sentences:

- "The model framework includes all known light and meteorological responses of photosynthesis but uses fixed canopy structures and phenology." (pag. 1, ll. 4–7).
- "Future research needs to incorporate the indirect effects of aerosols and possible feedbacks from dynamic carbon allocation and phenology." (pag. 1, ll. 19–20).

We add 2 sentences to existing explanatory paragraph in Section 2.1.1 (pag. 6, ll. 187–197):
"Linkages between vegetation and atmospheric aerosols are extremely complex. This version of the land carbon cycle model captures the meteorological (light, temperature, relative humidity, precipitation) responses of photosynthesis. The use of fixed canopy structures and phenology means that leaf mass is not driven by photosynthetic uptake of $CO_2$ and a closed carbon cycle is not simulated. Thus, the simulated GPP and isoprene emission responses may be underestimated because the LAI is insensitive to $CO_2$ uptake and climate. The objectives here are to examine the meteorological responses in detail and to offer a benchmark for future research that will incorporate additional feedbacks from dynamic LAI and phenology. For example, aerosol-induced effects on light and surface temperature may alter (i) the onset and shutdown dates of photosynthesis and growing season length (Yue et al., 2015a) (ii) the carbon allocation, LAI and tree height that provide a feedback to GPP (Yue et al., 2015b)."
We add to Section 4 (pag. 18, ll. 605–606):

"Thirdly, we did not include feedbacks from dynamic LAI and phenology that may lead to an underestimation of the effects of aerosol-induced effects on plant-productivity. Future research will address these three limitations."

**9. Table 1***: I would find this table quicker to understand if the changes were in percentages but it is not critical. Also one could expand the table to include the relevant numbers for regional changes so as to correspond to the discussions in sections 3.3.1 to 3.3.3 of the main text.*

**Authors**: To simplify the reading-understanding of Table 1, we provided percentage changes in ACB between the control and the sensitivity simulations. Moreover, we added two new tables (Table S1 and Table S2) to the Supplementary Material where we present changes in annual average sulfate, nitrate and SOA ACB (Table S1) and ERF (Table S2) over selected key regions.

**10. Comment***: There are a lot (!) of figures in the Supplementary Material but few if any of these figures are actually referenced in the main text. If its important than they should be mentioned in the main text.*

**Authors**: We agree with Referee #1, hence we kept only Figures that are discussed in the main text and we reduced the number of Figures in the Supplementary Material (from 14 figures in the previous version to 8 figures in the current one).

## Responses to Referee #2's Comments

We thank Referee #2 for their time and consideration. We closely considered the insightful and constructive comments from Referee #2. Referee #2's comments have helped us to improve the manuscript.

Referee #2 raises some pertinent points about (a) the assessment of model evaluation, (b) the complexity of Section 3.2 and (c) the unique attribution of changes in land carbon fluxes to prevailing mechanisms. Following Referee #1's recommendation, we completed model evaluation with computation of the linear correlation Pearsons coefficient (Pearsons $R$), the Pearsons R squared ($R^2$) and the root-mean-squared error ($RMSE$). In the revised manuscript, we re-organized Section 3.2 to discuss separately regional changes in aerosol pollution (now Sec. 3.2), surface solar radiation (now Sec. 3.3) and surface meteorology (now Sec. 3.4), and we inserted Table 5 that gathers changes in annual averages over target regions. Together with an attempt to clarify the discussion of results, we indicate the main limits of our approach, including the opportunities and challenges to linking aerosol-driven changes in surface meteorology to the changes in land carbon fluxes.

Referee #2's major comments are quoted in italics. Authors' answer follows referee's comment.

### Response to General Comments

**1. Section 3.2.1***: I found Sections 3.2.1 a bit difficult to follow. May I suggest treating the results for direct and diffuse radiation each alone in their own subsections/paragraphs, followed by a summary on the net impact on total radiation? Or alternatively, treat each region in their own separate subsections/paragraphs? In my opinion, this section could be refocused so that its more consistent with what is important for understanding the results presented in Section 3.3.1 and 3.3.2. I was not convinced how the effects of cooling and scattering were unequivocally separated later in the manuscript, and I suspect that could be laid out more clearly in the presentation of results here. I also had trouble being convinced of some of the regional comparisons that were being made.*

**Authors**: We agree with Referee #2 that the structure of Section 3.2 needed to be improved and clarified. For this reason, we re-structured the whole Section 3 and chose to discuss, in the order, contributions of aerosol pollution from the different sensitivity simulations (Sec. 3.2), aerosol-driven changes in surface solar radiation (Sec. 3.3) and in surface meteorology (Sec. 3.4), and, at last, changes in land carbon fluxes (plant productivity in Sec. 3.5, and isoprene emissions in Sec. 3.6). In each section, we firstly present changes at the global-scale, afterwards we focus on changes in five key regions: eastern North America, Eurasia, north-eastern China, the north-western Amazon Basin and central Africa. We agree with Referee #2 that comparisons among key regions were not clear from the figures showing the spatial distribution of annual/seasonal changes. Hence, we added a new table in the revised manuscript (Table 5) that presents, for each of the five key regions, absolute and percentage changes in annual average surface radiation, canopy temperature, GPP and isoprene emissions. For the five key regions, absolute and percentage changes in seasonal averages are reported in the Supplementary Material (Table S3 and Table S4). In the revised manuscript, the comparison between key regions is now mostly

based on results gathered in Table 5. The methodology to compute absolute and percentage differences in annual and seasonal averages over selected key regions is presented in Section 2.2 (pag. 7–8, ll. 231–237).

**2. p. 25446, lines 5–6***: Its unclear from Figure 4 that the eastern US shows much larger of an increase in diffuse radiation than over China for example (especially looking at panel (i)). This point seems important further on in the article, so I think it deserves further clarification.*

**Authors**: Table 5 should now help in clarifying this point. Table 5 shows a larger increase in annual average diffuse radiation over eastern North-America compared to Eurasia and north-eastern China due to all anthropogenic aerosols (pag. 13, ll. 415–420):
"The eastern North America shows the largest increase in annual diffuse radiation due to all anthropogenic aerosols (+8.6 W $m^{-2}$; +6.2 %), followed by north-eastern China and central Africa, which experience similar changes ($\sim$ +7.4–7.9 W $m^{-2}$; $\sim$ +5.7 %). Over the eastern North-America, the increase in diffuse radiation maximizes during boreal summer (+13.6 W $m^{-2}$; +8.9 %), with changes that are 1.6–5.7 W $m^{-2}$ (1.9–3.3 %) higher that those observed over north-eastern China and Eurasia (Table S3 in the Supplementary Material)."
However, in response to non-biomass burning aerosols, eastern North-America, Eurasia and north-eastern China show similar increases in diffuse radiation (pag. 17, ll. 568–570):
"In response to aerosol pollution from non-biomass burning sources Europe and China show a large decrease in annual average direct radiation (−24–26%), but a similar increase in diffuse radiation (+3–5 %) as eastern North America (Table 5).".

**3. p. 25446, lines 8–11***: The authors state that biomass burning aerosol drive the decrease in several regions (in the range of -6 to -28 W $m^{-2}$), but as I look at Figure 4 over the regions named, it seems to me that subtracting the industrial sources also result in decreases on the order of -6 to -12 W $m^{-2}$ and larger. This seems especially true when looking at the seasonal results in Figure S6. Am I misinterpreting the plots?*

**Authors**: Referee #2 is correct. In the revised manuscript, the insertion of Table 5 should make this point clearer and illuminate the comparison of impacts of biomass burning and industrial sources on surface radiation. In industrialized key regions (i.e., eastern North America, Eurasia and north-eastern China), industrial aerosols (non-biomass burning aerosols) mostly drive changes in surface radiation (pag. 13, ll. 415–425). On the contrary, in biomass burning key regions (i.e., the north-western Amazon Basin and central Africa), biomass and non-biomass burning aerosols share a similar contribution to changes in surface radiation (pag. 13, ll. 425–429).

**4. Section 3.3.1 and 3.2.1***: To pick up on this a little more, I also had some trouble with Section 3.3.1. Many of the conclusions here seemed to depend on contrasting the magnitude of certain effects over various regions. However, when I would try to corroborate the statements by consulting the Figures myself, in some cases the magnitudes didnt appear to be all that different. This might have to do with the Figures themselves, or maybe this could be improved by refocusing Section 3.2.1. In some cases, perhaps (re-?) stating some of the actual values would help.*

**Authors**: We agree with Referee #2 that figures showing the spatial distribution of annual/seasonal changes between the control and the sensitivity simulations do not provide a

proper support to compare the magnitude of aerosol-driven impacts in the five key regions. To answer to this point and underpin discussion of results, in the revised manuscript we added Table 5 (plus Table S3 and Table S4 in the Supplementary Material) to show changes (absolute and percentage changes) in annual (seasonal in the Supplementary Material) average surface radiation, canopy temperature, GPP and isoprene emissions. In the revised manuscript, the quantitative discussion of results is now mostly based on values summarized in these tables (i.e., Table 5 in the main text, Table S3 and Table S4 in the Supplementary Material).

**5. p. 25450, lines 4–8***: I dont see from Figure 4 how the increase in diffuse radiation over the eastern US is that much larger than over China and parts of Europe (as I mentioned above). Moreover, its not at all convincing from Figure 5 that SAT over the eastern US is "reduced". There is a very small isolated patch of blue, but there is no hatching anywhere to denote significance, and most of the region is blank. Im also confused as to what is "contrary" about Europe and China experiencing a strong reduction in total and direct radiation. Panel 4a and 4b show the US experiences comparable decreases in total and direct radiation as for parts of Europe, and maybe China. Maybe part of this confusion can be clarified by better summary of the results of Figure 4 in Section 3.2.1?*

**Authors**: We attempt to avoid confusion via Table 5, plus Table S3 and Table S4 in the Supplementary Material. As described in point 2, anthropogenic pollution aerosols drive a larger increase in annual average diffuse radiation over eastern North-America compared to Eurasia and north-eastern China (pag. 13, ll. 415–420). However, non-biomass burning aerosols (non-BBAs) drive similar increase in diffuse radiation in eastern North-America, Eurasia and north-eastern China (pag. 17, ll. 568–570).
In terms of total and direct radiation, due to non-BBAs, Eurasia and north-eastern China undergo the largest reduction in total and direct radiation. Over Eurasia and north-eastern China, decreases in total and direct radiation maximize during boreal summer, with changes that double those observed over eastern North-America (pag. 13, ll. 423–425).

**6. p. 25450, lines 22–23***: I can see from Figure 4 how it might be true that the increase in diffuse radiation over the Amazon is weaker than over central Africa but it doesnt seem that different, either. As a matter of fact, Section 3.2.1 places the two regions in the same sentence within the same range . . . So its not clear how the statement "the Amazon basin experiences a weaker increase in diffuse radiation" can be all that significant. Again, this might be helped by better structuring Section 3.2.1 to correspond to the conclusions being made here in Section 3.3.1 (and/or by referring to exact values over specific regions, for diffuse and direct radiation separately). Likewise, the "larger cooling" experienced by the Amazon basin compared to central Africa (Figure 5) doesnt appear notable to me either. In Panel 5a, they have roughly the same amount of area that is hatched as significant. This statement seems important to their conclusions about how "cooling dominates in the Amazon basin", but as is, I think the authors need to do a better job showing that this is true.*

**Authors**: In the revised manuscript, we reformulate our hypothesis regarding aerosol-driven effects on tropical regions (Sec. 3.4, pag. 13–15). In the model, photosynthesis and stomatal conductance are coupled through the Farquhar-Ball-Berry approach. Direct radiative forcing (DRF)-driven increases in photosynthesis and GPP are associated with increases in canopy con-

ductance and relative humidity via increased transpiration. Due to BBAs, the north-western Amazon Basin records the largest increase in transpiration efficiency and, as a corollary, the largest decrease in canopy temperature ($-0.31\,\mathrm{K}$; $-0.10\,\%$), which is $\sim 0.1\,\mathrm{K}$ larger than the decrease in canopy temperature over central Africa and north-eastern China. (pag. 13, ll. 460–467). We name this as "bio-meteorological effect" since reductions in the canopy temperature observed in the north-western Amazon Basin represents a positive feedback on plant productivity (further increases) in response to the DRF-driven increases. The same bio-meteorological effect (i.e., robust decrease in canopy temperature and corresponding GPP enhancement) seems to operate also in central Africa and north-eastern China; these regions undergo additional substantial robust reductions in direct radiation. In central Africa, the analysis of seasonal changes in GPP reveals that enhancement in GPP maximizes in boreal autumn, together with decrease in canopy temperature, while reductions in direct radiation maximizes in boreal summer (pag. 16, ll. 524–529).

**7. p. 25451, lines 12–16***: Again, given the results that have been presented, Im not yet convinced that the different mechanisms for each region (light scattering over Eastern US; reductions in direct radiation in Europe and China; cooling in the Amazon Basin) could have been established from the present model results alone. In my opinion, the arguments leading up to this based on the present model results alone have not been clearly developed.*

**Authors**: Please see Responses to points (1)–(6) above. We agree with Referee #2 that the way our original ideas were presented may not have been conclusively supported by the simulations results available to us. The new Table 5 makes the key drivers and processes across regions more quantitatively apparent and transparent. Of course, more than one mechanism operates in each region, and a confounding issue is that the mechanisms are not independent of each other. Therefore, given the quantitative data available to us from the completed global simulations e.g. as presented in Table 5, we identify the predominant mechanism in each region while fully recognizing the complexity of aerosol-meteorology-vegetation interactions.
As a final note, we expect the analyses to become increasingly complex when we turn on the dynamic carbon allocation and prognostic phenology. Therefore, in our on-going project work, we are developing a standalone version of YIBs that includes a fully coupled atmospheric radiative transfer scheme, which will be applied in our future studies.

## Response to Specific Comments

**1. Section 2.1 p. 25441, line 2***: Is there a particular reason that the Unger et al. 2013 ACPD article is being cited, when the ACP article is available?*

**Authors**: For Unger et al. (2013), the correct reference, which concerns the ACP article and not the ACPD, has been entered in the Reference list.

**2. Section 2.2 p. 25442, line 16***: Can you state/show some of the IPCC values that you are referring to for comparison, so the reader can see how consistent the results here are?*

**Authors**: Following Referee's #2 comment, we compare NASA ModelE2-YIBs to ERF and RF values from the IPCC AR5 values in Section 3.1 (pag. 8, ll. 241–255). Although the ERF and RF concepts differ, since our study only encompasses the direct aerosol effect and since the

IPCC AR5 report only presents RF by single component, we compare IPCC AR5 RF values to the corresponding ERF simulated by NASA ModelE2-YIBs, which are reported in Table 2 (i.e., SimCTRL−SimNOant values).

**3. p. 25442, line 23***: This is certainly on the low end of the global isoprene emissions estimate. Could you comment on why this might be?*

**Authors**: We inserted comments about the global isoprene estimation provided by NASA ModelE2-YIBs and changed the sentence to (pag. 8, ll. 260–266):
"The global isoprene source is 402.8 TgC $yr^{-1}$ , which is at the low end of the range of previous global estimates (e.g., 400–700 TgC $yr^{-1}$, Guenther et al., 2006). However, a recent study suggests a larger range of 250–600 TgC $yr^{-1}$ (Messina et al., 2015). The photosynthesis-based isoprene emission models tend to estimate a lower global isoprene source than empirical models because the scheme intrinsically accounts for the effects of plant water availability that reduce isoprene emission rates (Unger et al., 2013).".

**4. Sections 3.1.1 and 3.1.2***: Its not clear to the reader how "consistent" the AOD and GPP results are with observations. While the Figures do a good job showing that the model can broadly reproduce some of the spatial patterns, could some quantifiable statistics from the comparisons be shared?*

**Authors**: As suggested by Referee #2, we add a new table (Table 4) to present quantifiable statistics regarding model evaluation against observations: MODIS for coarse aerosol optical depth (AOD), and global FLUXNET-derived gross primary productivity. In the revised manuscript, for annual and seasonal (i.e., boreal summer and winter) average, Table 4 reports: the linear correlation Pearsons coefficient (Pearsons $R$), the Pearsons R squared ($R^2$) and the root-mean-squared error ($RMSE$). Comments concerning model evaluation and Table 4 are reported in Section 3.1 (for coarse-AOD: pag. 9–10, ll. 301–307; for GPP: pag. 10, ll. 316–319). Moreover, for GPP, we reported the main results of site-level evaluation of the model YIBs as performed in [**?** ] (pag. 10, ll. 320–323).

**5. Section 3.2.1: p. 25445, lines 14–16***: Should the authors clarify when they say "slightly affected" or "highly sensitive" that they are referring to the relative change (%)? The absolute magnitudes seem roughly equally considerable (∼ 2–8 $W m^{-2}$).*

**Authors**: We rephrased the mentioned sentence and referred to changes, precising the range of absolute and percentage changes in parentheses (pag. 12, ll. 392–397):
"Relative to the control simulation (SimCTRL), changes in global total and diffuse radiation are slightly affected by the pollution aerosol burden (absolute change for total radiation: from +1.6 W $m^{-2}$ to +5.1 W $m^{-2}$; absolute change for diffuse radiation: from −1.3 W $m^{-2}$ to −3.8 W $m^{-2}$; relative change: 1.7–2.5 %). On the contrary, changes in direct radiation shows a larger sensitivity range to the aerosol burden (absolute change: 2.9–8.9 W $m^{-2}$; relative change: 3.6–11.2 %)."
Following Referee's #1 suggestion, in Table 3 we provided relative changes between the control and the sensitivity simulations to help the readers interpreting results.

**6. p. 25445, line 26***: I think a word ("atmosphere"?) is missing between "aerosol laden" and*

*"due to".*

**Authors**: Since we modified the whole structure of Section 3.2.1 (now Section 3.3), the designated sentence is no more included in the revised manuscript.

**7. p. 25445, lines 25–27***: These lines seem to essentially repeat statements from the immediately preceding paragraph (lines* $\sim 12 - -14$*). Perhaps make it clearer that while the Table is global totals, Figure 4 shows the spatial distribution of the impacts.*

**Authors**: In the revised manuscript, we modified the whole structure of Section 3.2.1 (now Section 3.3), and we decided to comment, first, aerosol-driven changes in surface radiation at the global-scale via global totals gathered in Table 3 (Sec. 3.3.1). Afterwards, we present aerosol-driven changes in surface radiation in the five key regions by briefly commenting on Figure 4 and mainly comparing regions based on Table 5, which summarizes changes in annual average in the five key regions.
To clarify the content of Table 3, we inserted the following sentences at the beginning of Section 3.3.1 (pag. 12, 390–391):
"The global annual average shortwave visible solar radiation (total, direct and diffuse) for each simulations (control and sensitivity) are gathered in Table 3.".
To clarify the content of Figure 4, we inserted the following sentences at the beginning of Section 3.3.2 (pag. 13, ll. 409–411):
"Figure 4 shows the spatial distribution of aerosol-driven annual absolute changes in surface radiation (for annual percentage and seasonal absolute changes: Fig. S1 and S2 in the Supplementary Material)."

**8. p. 25446, line 17***: Correct "of" to "or".*

**Authors**: Since we substantially modified Section 3.2.1 (now Section 3.3), the designated and uncorrect sentence ("responsible of" instead of "responsible for") is no more included in the revised manuscript.

**9. Section 3.2.2 p. 25448, lines 14–15***: An explanation for how the changes will be linked to SSR and SAT uniquely might be useful here.*

**Authors**: Following Referee #2's suggestion, in the opening of Section 3.4 we remind that our experiments use fixed SSTs and do not consider aerosol indirect effects in clouds. These methodology limits the influence of pollution aerosols on the Earth System to direct changes in surface radiation that affect the atmosphere and land-surface only. For this reason, we mainly relate changes in land carbon fluxes to changes in surface radiation, surface meteorology (e.g., SAT) and plant conditions (e.g., transpiration, canopy temperature) (pag. 13, ll. 437–441).
In Section 3.4, we highlight as well that, by allowing rapid adjustments for the atmosphere and land-surface only, we do not observe significant changes (at 95 % confidence level) in SAT (on a global scale), nor in precipitation rate or cloud water content due to anthropogenic aerosol pollution (pag. 14, ll. 442–446).

**10. Section 3.3.1 p. 25449, lines 24–27***: The authors comment on how the impact is greatest for PFTs with complex canopy architectures. Maybe the evidence of this is found in the*

*Figure, but this its not explained clearly. Please elaborate.*

**Authors**: As suggested by Referee #2, we better explained how we assert that larger impact on GPP are observed in complex-forested canopy architectures with high tree heights and multiple layers (pag. 16, ll. 530–538).

**11. Section 3.3.3 p. 25453, line 19***: "not sensitive" Can you clarify how youve decided this? Do you mean that within 95% CI, there is no significant change?*

**Authors**: In the revised manuscript, the referred sentence has changed. Thank to comments of Referee #2, we corrected discussion of annual changes in isoprene emission in the north-western Amazon Basin, and we related changes in isoprene emission to changes in GPP. Actually, in the north-western Amazon Basin biomass burning aerosols drive a statistically significant (at 95 % confidence level) rise in isoprene emissions ($+0.4\,\mathrm{TgC\,yr^{-1}}$; +2.4 %), although the area of statistical significance is small (pag. 17–18, ll. 581–584):
"In the north-western Amazon Basin, annual average isoprene emission increases are simulated in response to BBAs ($+0.4\,\mathrm{TgC\,yr^{-1}}$; +2.4 %) (Table 5), although the area of statistical significance is small. In this region, the influence of increases in GPP on isoprene emission over-rides the influence of the cooler canopy temperatures (Table 5)."
Taking into account Referee #2's comments, at the end of Section 2 ("Methodology"), we defined the use of the adjective "significant" to refer to absolute/percentage changes that are statistically significant at 95 % confidence level (pag. 7–8, ll. 235–267).

**12. p. 25454, lines 2***: Insert a period between "US" and "This region . . . "*

**Authors**: Since we substantially modified Section 3, the designated sentences are no more included in the revised manuscript.

**13. Section 4 p. 25454, line 23–24***: I think the authors could include a brief comment about how aerosol pollution can drive plant phenology.*

**Authors**: Following Referee #2's suggestion, we commented about aerosol-driven changes in surface radiation and temperature that may affect plant phenology (pag. 6, ll. 194–197):
"For example, aerosol-induced effects on light and surface temperature may alter (i) the onset and shutdown dates of photosynthesis and growing season length (Yue et al., 2015a) (ii) the carbon allocation, LAI and tree height that provide a feedback to GPP (Yue et al., 2015b)."

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[1] Chen, M. and Zhuang, Q.: Evaluating aerosol direct radiative effects on global terrestrial ecosystem carbon dynamics from 2003 to 2010, Tellus B, 66, 21808, doi:10.3402/tellusb.v66.21808, 2014.

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# Potential sensitivity of photosynthesis and isoprene emission to direct radiative effects of atmospheric aerosol pollution

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**Abstract.** A global Earth system model is applied to quantify the impacts of direct anthropogenic aerosol effective radiative forcing on gross primary productivity (GPP) and isoprene emission. The impacts of different pollution aerosol sources (~~all~~ anthropogenic, biomass burning and non-biomass burning) are investigated by performing sensitivity experiments. ~~On the~~ The model framework includes all known light and meteorological responses of photosynthesis but uses fixed canopy structures and phenology. On a global scale, our results show that global land carbon fluxes (GPP and isoprene emission) are not sensitive to pollution aerosols, even under a global decline in surface solar radiation (direct~~+~~+diffuse) by ~~−9%. At the~~ −9%. At a regional scale, ~~plant productivity (GPP )~~ GPP and isoprene emission show a ~~-~~robust but opposite sensitivity to pollution aerosols, in regions where ~~complex~~ forested canopies dominate. In eastern North America and ~~Europe~~Eurasia, anthropogenic pollution aerosols (mainly from non-biomass burning sources) enhance GPP by ~~+8–12~~+5–8 ~~%~~ % on an annual average~~, with a stronger increase during the growing season (>12%)~~. In the ~~Amazon basin~~ north-western Amazon Basin and central Africa, biomass burning aerosols increase GPP by +2–5 ~~%~~ % on an annual average, with a peak in the ~~Amazon basin~~ north-western Amazon Basin during the dry-fire season (+5–8 ~~%). In~~ %). The prevailing mechanism varies across regions: light scattering dominates in the eastern North America while reduction in direct radiation dominates in Europe and China. Aerosol-induced GPP productivity increases in the Amazon and central Africa include an additional positive feedback from reduced canopy temperatures in response to increases in canopy conductance. In Eurasia and north-eastern China, anthropogenic pollution aerosols drive a decrease in isoprene emission of ~~−2 to −12% on the~~ −2% to −12% on an annual average. ~~Anthropogenic aerosols affect land carbon fluxes via different mechanisms and we suggest that the dominant mechanism varies across regions: (1) light scattering dominates in the eastern US; (2) cooling in the Amazon basin; and (3) reduction in direct radiation in Europe and China~~Future research needs to incorporate the indirect effects of aerosols and possible feedbacks from dynamic carbon allocation and phenology.

## 1 Introduction

Terrestrial gross primary productivity (GPP), the amount of carbon dioxide ($CO_2$) taken up every year from the atmosphere by plant photosynthesis, is the largest single flux in the carbon cycle and therefore plays a ~~-~~major role in global climate change. GPP is tightly connected to climatic variables (e.g., temperature, water, light) (Beer et al., 2010). In turn, terrestrial vegetation

provides the main source of isoprene to the atmosphere, which ~~influences~~ controls the loading of multiple short-lived climate pollutants and greenhouse gases (ozone, methane, secondary aerosols). Isoprene production is closely linked to plant photosynthesis (Pacifico et al., 2009; Unger et al., 2013). Hence, both GPP and isoprene emission may be influenced by a change in surface solar radiation (SSR, the sum of the direct and diffuse radiation incident on the surface) and surface atmospheric temperature (SAT). Anthropogenic aerosols affect directly the ~~Earth's~~ Earths radiation flux via: (a) scattering, which alters the partitioning between direct and diffuse radiation~~and~~, increases the diffuse fraction of SSR and affects SAT (Wild, 2009); and (b) absorption, which reduces SSR and SAT (Ramanathan et al., 2001). Furthermore, aerosols may attenuate indirectly SSR by acting as cloud condensation nuclei, thus perturbing cloud cover and cloud properties (Rosenfeld et al., 2008).

In 1991, Mount Pinatubo (Philippines) injected 20 megatons of sulfur dioxide ($SO_2$) into the stratosphere causing a massive production of sulfate aerosols, with substantial impacts on climate, and on the water and carbon cycles (Jones and Cox, 2001; Gu et al., 2003; Trenberth and Dai, 2007). In the aftermath of the eruption, a loss in net global radiation at the TOA (Top Of the Atmosphere) and a concomitant cooling were observed, and ultimately led to drying (Trenberth and Dai, 2007). By efficiently scattering light, the volcanic sulfate aerosol production caused a significant increase in diffuse solar radiation. In 1991 and 1992, at two northern mid-latitude sites, Molineaux and Ineichen (1996) recorded an increase in clear-sky diffuse radiation by +50 %, compensated by a concomitant decrease in direct radiation ~~by~~ of −30 %. Over the same period, in a deciduous forest in North-America, Gu et al. (2003) ascribed to increased diffuse radiation an enhancement in plant productivity of +23 and +8 % in the two years following the Pinatubo eruption. On the global scale, enhancement in the terrestrial carbon sink was proposed as one of the main drivers of the sharp and rapid decline in the rate of atmospheric $CO_2$ rise observed in the post-Pinatubo period, which resulted in a decrease of 3.5 ppmv by 1995 in atmospheric $CO_2$ (Keeling et al., 1995; Jones and Cox, 2001; Gu et al., 2003). The "Mount Pinatubo experiment" suggested a possible global response of terrestrial vegetation to the "diffuse fertilization effect" (DFE). Observational and theoretical studies show that plant productivity is more efficient under multi-directional diffuse rather than direct light because shaded non-light-saturated leaves increase their photosynthetic rate (Gu et al., 2002).

The DFE on plant photosynthesis has been extensively observed at ecosystem scale under cloudy skies (e.g., Gu et al., 2002; Niyogi et al., 2004; Cheng et al., 2015) and a chronic aerosol loading (e.g., Gu et al., 2003; Oliveira et al., 2007; Cirino et al., 2014) in diverse ecosystems (rainforest, deciduous and needleleaf forest, crop- and grasslands). The main conclusions of these studies are: (1) DFE prevails in complex and closed canopies, such as forests (Niyogi et al., 2004; Kanniah et al., 2012); (2) intermediate aerosol optical depth (AOD) enhances plant productivity, while high AOD (> 2–3) reduces carbon uptake rate because of a large reduction in direct radiation (Oliveira et al., 2007; Artaxo et al., 2013; Cirino et al., 2014). An ecosystem-scale measurement study in a European mixed needleleaf and deciduous forest reported increased isoprene emissions under conditions of higher diffuse light (Laffineur et al., 2013).

A few modeling studies have investigated aerosol-induced effects on plant productivity. Regional- and daily-scale assessments have been performed over: the Yellow River region (China), selecting a period of five days (Steiner and Chameides, 2005); and over the eastern ~~US~~United States, selecting two growing seasons (Matsui et al., 2008). Results in both studies are consistent with the main conclusions of the local observational studies. Steiner and Chameides (2005) demonstrated the im-

portance of both aerosol-induced radiative (i.e., change in light amount and its partitioning) and thermal (i.e., change in surface temperature) effects on plant transpiration and productivity. However, these studies focus on short time periods and a ~~-~~limited number of ecosystems using offline models with single-layer canopy schemes.

By applying a ~~-multy-layer-~~ multi-layer canopy scheme in ~~the framework of~~ an offline modeling ~~setup~~ framework (i.e., aerosol, radiative transfer and ~~land use~~ land-surface models are coupled offline), Rap et al. (2015) performed a ~~-~~regional- and ~~decade-scale~~ decadal-scale assessment of aerosol-induced effects on plant productivity in the Amazon basin from 1998 to 2007. The authors specifically focused on biomass burning aerosols (BBAs) and quantified that BBAs increase the annual mean diffuse light and net primary production (NPP) by, respectively, ~~~ 5~~ ~ 5 % and ~ 2.5 %. Deforestation fires play a ~~-~~key role and drive ~ 40 % of the estimated changes in light and photosynthesis. Moreover, Rap et al. (2015) assessed that in the Amazon basin during ~~1998–2007~~ 1998–2007 the DFE (a) was larger than the $CO_2$ fertilization effect, and (b) it could counteract the negative effect of droughts on land carbon fluxes.

A ~~-~~global-scale assessment of the aerosol-induced effects on the carbon cycle was performed by Mercado et al. (2009) using an offline land-surface model ~~(with a~~ with a multi-layer canopy scheme~~)~~. The authors concluded that DFE enhanced the global land carbon sink by +23.7 % over the 20th century, under an overall radiation (direct+diffuse) change of +9.3 %. Mercado et al. (2009) reconstructed historical SSR using radiative transfer calculations and a ~~-~~global climate dataset for the "global dimming" (period 1950–1980) and the "global brightening" period (after 1990s) (Wild, 2009, 2012; Streets et al., 2009). Recently, Chen and Zhuang (2014) applied an atmospheric radiative transfer module coupled with a ~~terrestrial ecosystem module~~ terrestrial ecosystem model to quantify aerosol direct radiative effects on global terrestrial carbon dynamics during 2003–2010. Using transient atmospheric $CO_2$ and prognostic leaf area index (LAI, one-sided green leaf area per unit ground area), the authors evaluated aerosol impacts on plant phenology, thermal and hydrological conditions as well as solar radiation. Chen and Zhuang (2014) estimated that, on a ~~-~~global scale, aerosols enhance GPP by 4.9 $Pg\,C\,yr^{-1}$ and slightly affect respiration. Chen and Zhuang (2014) accounted for all atmospheric aerosols and they did not target anthropogenic pollution aerosols.

Understanding all anthropogenic factors that influence the land carbon cycle is crucial to better manage terrestrial vegetation and to any effort to mitigate climate change by stabilizing atmospheric $CO_2$ concentrations. In the present study, we quantify the sensitivity of GPP and isoprene emission to the direct radiative effects of a ~~-~~realistic present-day pollution aerosol loading. Using a ~~-~~global Earth system model that represents vegetation–oxidant–aerosol–climate coupling, we perform sensitivity simulations to isolate the impact of the present-day pollution aerosols on GPP and isoprene emission. We tackle the direct aerosol effect only (absorption + scattering) and its impact on SSR and SAT that affects land carbon fluxes. Aerosol indirect effects on cloud properties are not addressed in this study due to the large uncertainties (Boucher et al., 2013; Myhre et al., 2013a). This study focuses on GPP because it is the first step in the long-term storage of atmospheric $CO_2$ in the living tissues of plants and is directly affected by solar radiation. We do not address aerosol effects on other land carbon cycle fluxes (e.g., respiration, net ecosystem exchange). We employ the effective radiative forcing (ERF) concept metric introduced in the IPCC AR5 in which all physical variables are allowed to respond to the direct aerosol–radiation perturbations except for those concerning the ocean and sea ice (Myhre et al., 2013b). The inclusion of these rapid adjustments in the ERF metric allows us to investigate the multiple ~~short-term~~ aerosol-induced concomitant meteorological impacts on the biosphere.

Section 2 describes the global Earth system model tool (NASA ModelE2-YIBs) and the experimental design. In Sect. 3, we evaluate simulated present-day atmospheric aerosols and GPP against global observational datasets including AOD from the Moderate Resolution Imaging Spectroradiometer (MODIS) and global gridded GPP that was generated using data orientated diagnostic upscaling of site-derived GPP from FLUXNET (Beer et al., 2010; Bonan et al., 2011; Jung et al., 2011). In addition, we present the analysis of results from the sensitivity simulations. In Sect. 4, we discuss the results and summarize conclusions.

## 2 Methodology

### 2.1 Global Earth system model: NASA ModelE2-YIBs

We apply the NASA GISS ModelE2 global chemistry-climate model at $2° \times 2.5°$ latitude by longitude horizontal resolution with 40-vertical layers extending to 0.1 hPa (Schmidt et al., 2014). The Yale Interactive Terrestrial Biosphere Model (YIBs) is embedded inside NASA ModelE2 in a ~~-~~framework known as NASA ModelE2-YIBs (Unger et al., 2013). The global climate model provides the meteorological drivers for the vegetation physiology. The land-surface hydrology submodel provides the grid cell level soil characteristics to the vegetation physiology. The model framework fully integrates the land biosphere–oxidant–aerosol system such that these components interact with each other and with the physics of the climate model. On-line oxidants affect aerosol production and on-line aerosols provide surfaces for chemical reactions and influence photolysis rates. The chemistry and aerosol schemes and their coupling have been well documented and extensively compared with observations and other global models (e.g., Bell et al., 2005; Bauer et al., 2007; Koch et al., 2006; Koch and Del Genio, 2010; Unger, 2011; Myhre et al., 2013a; Shindell et al., 2006, 2013a, b; Stevenson et al., 2013).

The aerosol package includes mass-based simulation of sulfate, nitrate and sea salt (e.g., Koch et al., 2006), carbonaceous aerosols (black carbon, BC, and primary organic matter, OC) (Koch and Hansen, 2005), mineral dust (Miller et al., 2006), and biogenic secondary organic aerosol (BSOA) (Tsigaridis and Kanakidou, 2007). The model assumes log-normal size distributions with effective radii: 0.2 µm (sulfate); 0.3 µm (nitrate); 0.1 µm (BC); 0.3 µm (OC). Sea salt aerosols are represented by two size bins with effective radii of 0.44 and 5 µm. Mineral dust aerosols are tracked in four size bins, ranging from 0.1 to 10 µm, and can be coated by sulfate and nitrate aerosols. Hygroscopic aerosols (sulfates, nitrates, sea salt and organic carbon) increase in size ~~as the relative humidityincrease~~with increasing relative humidity, which increases the aerosol scattering efficiency and radiative forcing (Schmidt et al., 2006).

The direct effect interaction between aerosols and radiation is reproduced by the on-line (two-way coupled) mode: aerosol fields are simulated at each model time step (30 min) and influence the simulated short and longwave radiation through scattering and absorption in the radiation submodel, which in turn influences the climate dynamics. Thus, aerosols induce (a) changes in simulated diffuse and direct photosynthetically active radiation (PAR, spectral range of surface visible solar radiation, 400–700 nm, used by plants to photosynthesize) that are passed from the radiation submodel to the vegetation model; and (b) ~~fast feedback~~ rapid adjustment changes in meteorology (temperature, precipitation, circulation) that are passed from the ~~model's atmosphere~~ model atmosphere and land surface to the vegetation model.

**The Yale Interactive Terrestrial Biosphere model (YIBs)**

The vegetation structure describes eight plant functional types (PFTs): tundra, grassland, shrubland, deciduous broadleaf forest, savannah, tropical rainforest, evergreen needleleaf forest, and cropland. The PFT-specific vegetation cover fraction and LAI are the standard atlas-based distribution in NASA GISS ModelE2 (Schmidt et al., 2014). ~~Leaf area index (LAI )~~ LAI for each PFT is prescribed according to regular seasonal sinusoidal variation between PFT-specific minimum and maximum seasonal LAI values that is insensitive to climate drivers or carbon balances (Rosenzweig and Abramopoulos, 1997; Friend and Kiang, 2005). Each model PFT fraction in the vegetated part of each grid cell represents a ~~-~~single canopy. The canopy radiative transfer scheme assumes a ~~-~~closed canopy and distinguishes vertically canopy layers into sunlit and shaded leaves, as well as the different ~~contribution~~ contributions from direct and diffuse PAR (from the climate model's radiation scheme) at the leaf level (Spitters et al., 1986). The leaf-level carbon and water fluxes are scaled up to the canopy level by integrating over each canopy layer, using an adaptive number of layers (typically ~~1~~62–16) (Friend and Kiang, 2005). After upscaling from leaf to canopy, the carbon and water fluxes are exchanged with the atmosphere on the ~~30~~

The vegetation biophysical fluxes are calculated using the well-established ~~Michealis–Menten enzyme-kinetics~~ leaf model of photosynthesis (Farquhar et al., 1980; Von Caemmerer and Farquhar, 1981) and the stomatal conductance model of Ball and Berry ~~(?)~~ (Ball et al., 1987) . In the leaf model, the rate of net $CO_2$ uptake in the leaves of $C_3$ plants is the result of three competing processes: $J_c$, the carboxylation-limited rate; $J_e$, the electron transport-limited photosynthesis rate; and $J_s$, the export-limited rate to use photosynthesis products. The coupled photosynthesis~~and stomatal conductance~~ , stomatal conductance and diffusive $CO_2$ flux transport equations are solved analytically at the leaf level using a ~~-~~cubic function in ~~net photosynthetic~~ the net carbon assimilation rate. Isoprene emission is calculated as a ~~-~~function of $J_e$, intercellular and atmospheric $CO_2$ and canopy temperature (Unger et al., 2013).

As theoretical and observational studies have demonstrated, the aerosol effect on plant photosynthesis strongly depends on the canopy separation into sunlit and shaded leaves. These two parts of the canopy have different responses to the change in light partitioning driven by aerosols (Knohl and Baldocchi, 2008) . Under low PAR, both shaded and sunlit leaves are in a light-limited environment ($J_e$ controls the photosynthesis rate). Under high PAR, sunlit leaves are light-saturated and in a Rubisco-limited environment ($J_c$ controls the photosynthesis rate), while shaded leaves are in a light-limited environment ($J_e$). Hence, sunlit canopy photosynthesis depends on both direct and diffuse light, and on both $J_c$ and $J_e$ photosynthesis rates; while shaded canopy photosynthesis is directly influenced by diffuse light and mainly depends on the $J_e$ photosynthesis rate. The aerosol light-scattering directly influences $J_e$, hence it mainly affects shaded leaves (Matsui et al., 2008; Chen and Zhuang, 2014) .

Linkages between vegetation and atmospheric aerosols are extremely complex. This version of the land carbon cycle model captures the meteorological (light, temperature, relative humidity, precipitation) responses of photosynthesis. The use of fixed canopy structures and phenology means that leaf mass is not driven by photosynthetic uptake of $CO_2$ and a ~~-~~closed carbon cycle is not simulated. Thus, the simulated GPP and isoprene emission responses may be underestimated because the LAI is insensitive to $CO_2$ uptake and climate. The objectives here are to examine the meteorological responses in detail and to offer

a benchmark for future research that will incorporate additional feedbacks from dynamic LAI and phenology. For example, aerosol-induced effects on light and surface temperature may alter (i) the onset and shutdown dates of photosynthesis and growing season length (Yue et al., 2015a) , (ii) the carbon allocation, LAI and tree height that provide a feedback to GPP (Yue et al., 2015b) .

## 2.2 Simulations

The atmosphere-only configuration of NASA ModelE2-YIBs is used to perform a ~~-~~control simulation ("SimCTRL") representative of the present-day (~ 2000s). Prescribed decadal average monthly-varying sea surface temperature (SST) and sea ice observations for 1996–2005 from the HadSST dataset (Rayner et al., 2006) provide the lower boundary conditions for the global climate model. The present day trace gas and aerosol emissions are prescribed to year 2000 values from the historical inventory developed for IPCC AR5 ~~(RCP4.5; Lamarque et al., 2010)~~ (Lamarque et al., 2010) . Atmospheric levels of long-lived greenhouse gases are prescribed to: $CO_2 = 370$ ppmv; $CH_4 = 1733$ ppbv in Southern Hemisphere and 1814 ppbv in Northern Hemisphere; $N_2O = 316$ ppbv. A ~~-~~set of three sensitivity perturbation simulations ~~is~~ are performed that selectively remove anthropogenic short-lived gas-phase precursor and primary aerosol emissions: ~~(a)~~

(a) all anthropogenic emissions, including biomass burning~~(~~, are removed in "SimNOant"~~), (b) ~~;

(b) only biomass burning emissions ~~only (~~are removed in "SimNObb"~~), and (c) industrial emissions("SimNOind", ~~;

(c) all industrial emissions, which means all anthropogenic emissions ~~are removed~~ except biomass burning emissions ~~).~~ (e.g., industry, power generation, road vehicles; hereafter, we refer to these emissions as "non-biomass burning emissions"), are removed in "SimNOind".

The control and sensitivity simulations are run for 32 model years recycling the year 2000 boundary conditions every year but allowing the changes in atmospheric aerosol composition to influence meteorology and the land biosphere. By prescribing SSTs and sea ice cover at climatological values, while letting all other physical components of the Earth system to respond until reaching steady state, we capture short-term response of the land surface climate to the aerosol radiation perturbation. This fixed-SST technique allows us to compute ERF, the forcing metric that accounts for rapid tropospheric adjustments and better characterizes drivers in the troposphere (e.g., aerosols) (Myhre et al., 2013b). Hence, the fixed-SST technique enables us to analyze multiple meteorological effects of the direct aerosol–radiation interactions. The long run-time is necessary to allow the fast land and atmosphere climatic feedbacks to respond to the aerosol perturbations and the TOA radiation fluxes to equilibrate. ~~The~~ Integrations of 32 model years are completed for all simulations (control and sensitivity runs). The global atmospheric oxidant–aerosol composition usually takes about 2 years to spin-up, while the atmospheric dynamics and land–surface climate takes about 10 years to reach steady–state due to an imposed aerosol radiative forcing. Therefore, we discard the first 12 model ~~years are discarded~~ run years as spin-up. The ~~last~~ remaining 20 ~~of each simulation are used~~ model run years are averaged for analysis. Twenty model years of data are necessary such that any aerosol-driven variable differences between the control and sensitivity simulations are statistically significant relative to internal climate model variability. Our goal is to isolate the

effects of aerosol pollution on the land biospheric fluxes. Therefore, we compute the absolute differences in $X$ variable as: $\Delta X = X_{\text{ctrl}} - X_{\text{sens}}$. Percentage changes in $X$ are calculated relative to the control experiment (i.e., $\Delta_{\%} X = \Delta X / X_{\text{ctrl}} \times 100$) and~~are illustrated in the Supplement.~~ , for selected variables, are gathered in the Supplementary Material. Applying the same methodology, we compute absolute and percentage differences in annual and seasonal averages over selected regions. Hereafter, we define "significant" all absolute/percentage changes that are statistically significant at the 95% confidence level.

## 3 Results

### 3.1 Evaluation of present-day control simulation

Present-day values of global mean aerosol column burden (ACB) and ERF for aerosol–radiation ~~interaction by component shown~~ interactions (i.e., aerosol direct effect) are presented by component in Table ~~??~~ 1 and 2. The IPCC AR5 provides RF (not ERF) by single aerosol species (Boucher et al., 2013; Myhre et al., 2013b) . NASA ModelE2-YIBs ERF values for single aerosol species are consistent with ~~ranges presented in~~ the AR5 RF ranges. Nitrate ERF is on the lower bound of the AR5 RF range ($-0.30$ to $-0.03\,\mathrm{W\,m^{-2}}$). ERFs of sulfate, BC from industrial sources and SOAs fall into the AR5 RF ranges (respectively: $-0.60$ to $-0.20\,\mathrm{W\,m^{-2}}$, $+0.05$ to $+0.80\,\mathrm{W\,m^{-2}}$, and $-0.27$ to $-0.20\,\mathrm{W\,m^{-2}}$). OC from industrial sources and BBAs show ERFs consistent with the AR5 RF values ($\mathrm{OC_{ind}}$: $-0.09\,\mathrm{W\,m^{-2}}$, BBAs: $0.00\,\mathrm{W\,m^{-2}}$). Based on a combination of methods (i.e., global aerosol models and observation-based methods), the AR5 report estimates the total ERF due to aerosol-radiation interactions: $-0.45$ ($-0.95$ to $+0.05$) $\mathrm{W\,m^{-2}}$; in AR5, the best total RF estimate of the aerosol–radiation interaction is: $-0.35$ ($-0.85$ to $+0.15$) $\mathrm{W\,m^{-2}}$ (Myhre et al., 2013b) . The total ERF is computed in NASA ModelE2-YIBs as the arithmetic mean of all anthropogenic aerosol components (i.e., sulfate, nitrate, OC and BC from both industrial and biomass burning sources, SOA and dust). The NASA ModelE2-YIBs estimates a total ERF due to aerosol-radiation interactions of $-0.34$ ($-0.76$ to $+0.18$) $\mathrm{W\,m^{-2}}$, at the low end of the IPCC AR5 ~~report (Boucher et al., 2013; Myhre et al., 2013b) . Similarly the~~ range.

Similarly to the aerosols, the present-day land carbon fluxes are in good agreement with previous estimates (Table 3). Simulated global annual GPP ($116.0\,\mathrm{Pg\,C\,yr^{-1}}$) is in reasonable agreement with current understanding of the present-day carbon cycle budget (based on FLUXNET: $123 \pm 8\,\mathrm{Pg\,C\,yr^{-1}}$, Beer et al., 2010; based on MODIS: $109.29\,\mathrm{Pg\,C\,yr^{-1}}$, Zhao et al., 2005; based on the Eddy Covariance-Light Use Efficiency model: $110.5 \pm 21.3\,\mathrm{Pg\,C\,yr^{-1}}$, Yuan et al., 2010). The global isoprene source is ~~402.8and agrees with~~ $402.8\,\mathrm{Tg\,C\,yr^{-1}}$, which is at the low end of the range of previous global estimates (e.g., 400–$700\,\mathrm{Tg\,C\,yr^{-1}}$, Guenther et al., 2006~~; 412~~. However, a recent study suggests a larger range of 250–~~601, Arneth et al., 2008 ).~~ $600\,\mathrm{Tg\,C\,yr^{-1}}$ (Messina et al., 2015) . The photosynthesis-based isoprene emission models tend to estimate a lower global isoprene source than empirical models because the scheme intrinsically accounts for the effects of plant water availability that reduce isoprene emission rates (Unger et al., 2013) .

### 3.1.1 Aerosol Optical Depth (AOD)

We use the quality assured Terra MODIS Collection 5 (C5.1) monthly mean product (Level 3), a ~~-~~globally gridded dataset at $1^\circ \times 1^\circ$ resolution ~~regridded to at~~ re-gridded to $2^\circ \times 2.5^\circ$ resolution for comparison with the global model. To infer clear-sky (non cloudy) aerosol properties in part of the visible and shortwave infrared spectrum, MODIS C5.1 relies on two algorithms depending on surface reflectance: (1) the Dark Target (DT) algorithm, under conditions of low surface reflectance (e.g., over ocean, vegetation) (Levy et al., 2010); (2) the Deep Blue (DB) algorithm, designed to work under high surface reflectance, such as over desert regions (Hsu et al., 2004; Shi et al., 2014). To cover both dark and bright surfaces, we merge the DT and DB AOD products (i.e., DT missing data are filled in with DB values). We use MODIS TERRA C5.1 AOD data from 2000 to 2007 because DB AOD data are only available for this period due to calibration issues (Shi et al., 2014). The MODIS instrument also measures the fine model weighting (ETA) at 550 nm, consequently the fine-mode AOD can be computed as: fine-AOD = AOD × ETA, where fine-AOD is ~~a~~ the fraction of the AOD contributed by fine mode sized particles (i.e., effective radius $\ll 1.0\,\mu$m) (Levy et al., 2010; Bian et al., 2010). Quantitative use of MODIS fine-AOD is not appropriate because fine-mode aerosols play a ~~-~~main role in the scattering process (Levy et al., 2010).

NASA ModelE2-YIBs provides separately all-sky and clear-sky AOD diagnostics; we focus on clear-sky output since that is more comparable to the spaceborne observations. The model coarse-mode ~~AOD (~~($PM_{10}$, atmospheric particulate matter with diameter $< 10\,\mu$m) AOD includes all simulated aerosol species (sulfate, nitrate, organic and black carbon, SOA, sea salt and mineral dust); the model fine-mode ~~AOD~~ ($PM_{2.5}$, atmospheric PM with diameter $< 2.5\,\mu$m) AOD accounts for all simulated aerosol species except sea salt and dust.

Figure **??** compares the spatial distribution of annual and seasonal (boreal summer and winter) mean coarse-mode AOD in NASA ModelE2-YIBs (control present-day simulation) with observations from the MODIS satellite instrument (averaged over 2000–2007). Model global mean coarse-mode AODs are consistent with MODIS AOD global means. NASA ModelE2-YIBs reproduces strong biomass burning and dust episodes over Africa. In contrast, on both annual and seasonal averages the model underestimates the optical thickness of the aerosol layer over China and India, which is likely related to dust. The ~~model'~~models underestimate of Asian dust should not influence the focus of this study, to assess the impacts of anthropogenic pollution aerosols on the land carbon fluxes. The spatial and temporal distribution of fine-mode aerosols in NASA ModelE2-YIBs is consistent with MODIS observations (Fig. **??**). In general, the model shows a ~~-~~slightly higher fine-aerosol layer compared to MODIS ~~,~~ (e.g., over Europe, India and South America). Over China, model fine-AOD distribution is consistent with MODIS on the annual average; however, the model does not show the seasonal variability that MODIS observes. To quantify the model evaluation, on an annual average the NASA ModelE2-YIBs coarse-mode AOD global means present an acceptable correlation with the MODIS AOD global means ($R = 0.7$, $R^2 = 0.5$ and $RMSE = 0.05$, Table 4). Between boreal summer and winter, boreal summer shows the best model performance ($R = 0.8$, $R^2 = 0.6$ and $RMSE = 0.06$, Table 4). During boreal winter, outside the growing season, the NASA ModelE2-YIBs overestimates coarse-mode AODs. Since quantitative use of MODIS fine-AOD is not recommended, we do not quantify model performance for fine-mode AODs.

### 3.1.2 Gross Primary Productivity (GPP)

In Fig. **??**, we compare the spatial distribution of annual and seasonal (boreal summer and winter) mean GPP in NASA ModelE2-YIBs model (control present-day simulation) with a ~~-~~global FLUXNET-derived GPP product (averaged over 2000–2011). The model is consistent with the broad spatio-temporal variability in FLUXNET-derived GPP. We find a ~~-~~weaker annual and seasonal signal in the model GPP over the cerrado area in central South-America. However, since the FLUXNET-derived GPP product mainly relies on the availability of FLUXNET sites, which are densely distributed in temperate zone not in the tropics, FLUXNET-derived GPP may be biased over central South-America. On an annual average the NASA ModelE2-YIBs GPP highly correlates with the FLUXNET-derived GPP ($R = 0.9$, $R^2 = 0.7$, $RMSE = 1.0$, Table 4). Between boreal summer and winter, boreal winter presents the best model performance ($R = 0.9$, $R^2 = 0.9$ and $RMSE = 1.1$, Table 4). Recently, Yue and Unger (2015) performed a site-level evaluation of the YIBs model over 145 sites for different PFTs. Depending on PFT, GPP simulation biases range from $-19\,\%$ to $+7\,\%$. For monthly-average GPP, among the 145 sites, 121 have correlations higher than 0.8. High correlations ($> 0.8$) are mainly achieved at deciduous broadleaf and evergreen needle leaf sites; crop sites show medium correlation ($\sim 0.7$).

## 3.2 ~~Aerosol-induced~~ Aerosol pollution changes ~~to surface meteorology~~in sensitivity simulations

~~Table ?? shows the changes in~~

### 3.2.1 Global-scale

Table 1 shows the aerosol column burden (ACB) by component in the control and the three sensitivity simulations. Anthropogenic pollution emissions (SimCTRL−SimNOant) contribute $0.85\,mg\,m^{-2}$ to sulfate ACB (36 % of the total sulfate burden due to both anthropogenic and natural emissions), $4.47\,mg\,m^{-2}$ to nitrate ACB (87 %) and ~~ERF for each sensitivity simulation. Removal of all anthropogenic emissions (SimNOant): (a) reduces by ~ 10 % sulfate column burden and ERF; (b) decreases by ~ 70–80 % nitrate and SOA column burden and reduces respectively by ~ 80 and ~ 60 % sulfate and SOA ERF; and~~ $0.99\,mg\,m^{-2}$ to SOA ACB (72 %). Biomass burning emissions (SimCTRL−SimNObb) contribute $1.62\,mg\,m^{-2}$ to nitrate ACB (31 %) and $0.23\,mg\,m^{-2}$ to SOA ACB (17 %), while they do not significantly contribute to sulfate ACB. Non-biomass burning emissions (SimCTRL−SimNOind) contribute $0.89\,mg\,m^{-2}$ to sulfate ACB (37 %), $3.69\,mg\,m^{-2}$ to nitrate ACB (72 %) and $0.47\,mg\,m^{-2}$ to SOA ACB (34 %). For carbonaceous aerosols, anthropogenic pollution emissions contribute $1.45\,mg\,m^{-2}$ to the total OC ACB ($0.48\,mg\,m^{-2}$ from non-biomass burning, $OC_{ind}$, and $0.97\,mg\,m^{-2}$ from biomass burning, $OC_{bb}$) and $0.26\,mg\,m^{-2}$ to the total BC ACB ($0.17\,mg\,m^{-2}$ from non-biomass burning, $BC_{ind}$, and $0.09\,mg\,m^{-2}$ from biomass burning, $BC_{bb}$). Non-biomass burning emissions contribute $0.15\,mg\,m^{-2}$ to $OC_{bb}$ ACB (15 %) and $0.01\,mg\,m^{-2}$ to $BC_{bb}$ ACB (15 %).

Table 2 presents, by aerosol component, the ERF for aerosol-radiation interactions due to anthropogenic pollution, biomass burning and non-biomass burning emissions. Anthropogenic pollution emissions contribute $-0.31\,W\,m^{-2}$ to sulfate ERF (40 % of the total sulfate ERF due to both anthropogenic and natural emissions), $-0.38\,W\,m^{-2}$ to nitrate ERF (85 %) and $+0.10\,W\,m^{-2}$ to SOA ERF (c)~~reduces nearly to zero both column burden and ERF of OC and BC. Removal of biomass~~

~~burningemissions~~ 63 %). Biomass burning emissions contribute $-0.14\,W\,m^{-2}$ to nitrate ERF (30 %) and $-0.03\,W\,m^{-2}$ to SOA ERF (16 %), while they do not significantly contribute to sulfate ERF. Non-biomass burning emissions contribute $-0.30\,W\,m^{-2}$ to sulfate ERF (40 %), $-0.31\,W\,m^{-2}$ to nitrate ERF (70 %) and $-0.05\,W\,m^{-2}$ to SOA ERF (~~SimNObb) results in (a) negligible column burden and ERF from OCand BC, (b) reduction by ~ 30 % on both nitrate ACB~~ 29 %). For carbonaceous aerosols, anthropogenic pollution emissions contribute $-0.17\,W\,m^{-2}$ to the total OC ERF ($-0.06\,W\,m^{-2}$ from non-biomass burning, $OC_{ind}$, and $-0.11\,W\,m^{-2}$ from biomass burning, $OC_{bb}$) and $+0.30\,W\,m^{-2}$ to the total BC ERF ($+0.18\,W\,m^{-2}$ from non-biomass burning, $BC_{ind}$, and $+0.12\,W\,m^{-2}$ from biomass burning, $BC_{bb}$). Non-biomass burning emissions contribute $-0.01\,W\,m^{-2}$ to $OC_{bb}$ ERF (9 %) and ~~ERF; and (c) no impacts on the other aerosol components. Removal of anthropogenic emissionsexcept~~ $+0.02\,W\,m^{-2}$ to $BC_{bb}$ ERF (11 %).

### 3.2.2 Five key regions

Beyond the global results, our simulations reveal five strongly sensitive regions that correspond to important sources of aerosol pollution: eastern North-America, Eurasia, north-eastern China, the north-western Amazon Basin and central Africa (green boxes on Fig. **??**). Besides a substantial contribution to primary aerosol (PA) sources (i.e., BC and OC), all selected regions considerably contribute to secondary aerosol (SA) sources such as sulfate, nitrate and SOA (Table S1 for ACB and Table S2 for ERF in the Supplementary Material). We focus on SAs since, being finer than PAs, they play a key role in scattering and may trigger DFE.

In terms of aerosol burden, in the five key regions, nitrate is the dominant aerosol source, with a larger contribution from non-biomass burning compared to biomass burning emissions. Sulfate source is mainly governed by non-biomass burning emissions, except in central Africa where biomass burning emissions ~~(SimNOind) : (a) has the same effect of SimNOant on sulfate burden and ERF~~importantly contribute to sulfate ACB. For SOA source, both biomass and non-biomass burning emissions feed SOA ACB, with a larger contribution from biomass burning in central Africa.

Eastern North America and Eurasia share a similar contribution to nitrate ACB ($\sim 14$–$15\,mg\,m^{-2}$; $\sim 93$ %) and ERF ($-1.2$–$1.3\,mg\,m^{-2}$; $\sim 94$ %) due to anthropogenic emissions, with the largest input from non-biomass burning emissions (ACB: $12.7\,mg\,m^{-2}$; ERF: $-1.1\,mg\,m^{-2}$, $\sim 80$ %) compared to biomass burning emissions (ACB: $3.4\,mg\,m^{-2}$; ERF: $-0.3\,mg\,m^{-2}$, $\sim 20$ %). Eastern North America and Eurasia also show a similar contribution to SOA source due to anthropogenic emissions (ACB: $2.1\,mg\,m^{-2}$, $\sim 78$ %; ERF: $-0.2\,mg\,m^{-2}$, $\sim 72$ %). In both regions, non-biomass burning emissions provide a larger input to SOA source compared to biomass burning emissions, with a larger contribution in Eurasia compared to eastern North America (ACB: $1.4\,mg\,m^{-2}$ vs. $0.9\,mg\,m^{-2}$, 52 % vs. 32 %) and even a different sign in ERF (~~b)decreases by ~ 70 % nitrate column burden and ERF; (c) reduces nearly to zero both column burden and ERF of industrial OC and BC; and~~ $-0.2\,mg\,m^{-2}$ vs. $+0.08\,mg\,m^{-2}$, 45 % vs. 25 %). Compared to eastern North America and Eurasia, north-eastern China presents nearly a half nitrate source, while contributions to sulfate ACB due to anthropogenic emissions are about 0.5–$1\,W\,m^{-2}$ (~~d) decreases by ~ 30 % both ACB and ERFof SOA. Above-listed changes in the aerosol burden ultimately affect solar radiation, temperature and relative humidity (RH)at the Earth's surface, as we explore below.~~ 5–10 %) larger, and lead to more intense negative ERF (by 0.4–$0.6\,W\,m^{-2}$, 5–10 %). In the north-eastern China, anthropogenic emissions largely contribute as well to SOA

source with a share between biomass and non-biomass burning similar to Eurasia. The north-western Amazon Basin shows the smallest contributions to SA sources. However, compared to the other key regions, biomass burning and non-biomass burning emissions contribute by the same amount to SOA source (ACB: 0.5 W m$^{-2}$, 24–29 %; ERF: −0.06 W m$^{-2}$, 24–29 %). As previously commented, central Africa substantially contributes to sulfate source via both biomass (ACB: 0.6 W m$^{-2}$, 30 %; ERF: −0.2 W m$^{-2}$, 30 %) and non-biomass burning emissions (ACB: 0.7 W m$^{-2}$, 40 %; ERF: −0.3 W m$^{-2}$, 45 %). In this region, biomass burning emissions substantially feed SOA source, with contributions that nearly double those from non-biomass burning emissions (ACB: 2.1 W m$^{-2}$ vs. 1.1 W m$^{-2}$, 44 % vs. 22 %; ERF: −0.16 W m$^{-2}$ vs. −0.08 W m$^{-2}$, 48 % vs. 23 %).

#### 3.2.3 Surface solar radiation

## 3.3 Aerosol pollution changes to surface solar radiation

### 3.3.1 Global-scale

The global annual average shortwave visible solar radiation (total, direct and diffuse) for each ~~simulation are reported~~ simulations (control and sensitivity) are gathered in Table 3. Hereafter, we shorten "shortwave visible solar radiation" to "radiation". ~~Global~~ Relative to the control simulation (SimCTRL), changes in global total and diffuse radiation are slightly affected by the pollution aerosol burden ~~, and their changes have opposite sign but similar value (percentage changes range from 1.7 to~~ (absolute change for total radiation: from +1.6 W m$^{-2}$ to +5.1 W m$^{-2}$; absolute change for diffuse radiation: from −1.3 W m$^{-2}$ to −3.8 W m$^{-2}$; relative change: 1.7–2.5 %)~~; on~~ . On the contrary, ~~direct radiation is highly sensitive to change in~~ changes in direct radiation shows a larger sensitivity range to the aerosol burden (~~percentage changespans from~~ absolute change: 2.9–8.9 W m$^{-2}$; relative change: 3.6~~to −~~11.2 %)~~(Table 3). Referred to~~ . In the present-day ~~conditions, anthropogenic emissions drive a~~ world, anthropogenic pollution aerosols drive a decrease in global total and direct radiation by ~~, respectively,~~ −2.3 % (~~−5.2) and to −~~5.2 W m$^{-2}$) and −11.2 % (~~−9.0),~~ −9.0 W m$^{-2}$), respectively, while global diffuse radiation increases by +2.5 % (~~+3.7~~+3.7 W m$^{-2}$). Biomass burning ~~emissions~~ aerosols have almost zero effects on global total and diffuse radiation, while they reduce direct radiation by −3.6 % (~~−2.9~~−2.9 W m$^{-2}$). Non-biomass burning ~~emissions (industry, power generation, road vehicles~~aerosols (non-BBAs) decrease global total radiation by −1.7 % (~~−3.8~~−3.8 W m$^{-2}$) and increase global diffuse radiation by the same percentage (absolute change: ~~+2.6~~+2.6 W m$^{-2}$), while global direct radiation reduces by ~~−8 % (−6.4).~~

~~Anthropogenic aerosol burden affect globally annual average radiation (total, direct and diffuse) at the Earth's surface (Fig. **??**). Under the aerosol laden due to anthropogenic pollution, total and direct radiation decrease, while diffuse radiation rises. Via light absorption and scattering, anthropogenic aerosols drive a significant decrease in total and direct radiation by, respectively, −12 to −20(−10 to −20 %) and by −20 to −36(−20 to −40 %) over industrialized (eastern North America, Europe and Asia) and biomass burning (the Amazon basin and central Africa) regions (Fig. **??**a–b). Over these regions, via light scattering, anthropogenic aerosols increase diffuse radiation by +6–20(+6–12 %). Among industrialized regions, the eastern US shows the largest increase in diffuse radiation (+8–20; +8–12 %), most likely due to sulfate and BSOA production that~~

characterize this region (e.g., Carlton et al., 2010; Ford and Heald, 2013) (Fig. **??**c). Biomass burning aerosols drive a decrease in total radiation of −12 to −28(−10 to −20%) in the Amazon basin and central Africa, and a weaker decrease of −6 to −12(−5 to −10%) over boreal regions (Canada and Eurasia) (Fig. **??**d). Diffuse radiation increases with a larger signal over central Africa (+8–12; +8–10%) compared to the Amazon basin (+6–8; +8–10%) (Fig. **??**f). Changes in total and diffuse radiation are localized over the main biomass burning regions (the Amazon basin, Africa, South-East Asia and boreal regions), while decrease in direct radiation also affect robustly minor biomass burning regions such as North America and Australia (Fig. **??**e). Pollution aerosols from non-biomass burning sources are responsible of a strong reduction in total and direct radiation over Europe and China (Fig. **??**g and h).

During boreal summer, aerosol-induced impacts on solar radiation amplify over aerosol-source regions, such as the eastern US, Europe, China, the Amazon basin and central Africa (Figs. S6 and S7 in the Supplement). Driven by anthropogenic pollution aerosols, total radiation decreases by −15 to −30(−3 to −15%), direct radiation decreases by −20 to −50(−25 to −80%) and diffuse radiation increases by +10–25(+6–22%), with largest signals over the eastern US (+15–20; +10–14%) and central Africa (> 25; > 14%).

In summation −8.0% (−6.4 W m$^{-2}$). In summary, anthropogenic pollution aerosols drive an overall SSR (direct+diffuse) global decline of ~5~ 5 W m$^{-2}$. In the literature, estimates for the overall SSR decline during the "global dimming" (period 1950–1980) range from 3 to 93 to 9 W m$^{-2}$ (Wild, 2012). In percentage, anthropogenic pollution aerosols drive an overall SSR global decline of 8.7%.

### 3.3.2 Five key regions

Figure **??** shows the spatial distribution of aerosol-driven annual absolute changes in surface radiation (for annual percentage and seasonal absolute changes: Fig. S1 and S2 in the Supplementary Material). Regionally, on both annual and seasonal average, eastern North America, Europe, East Asia, the Eurasia, north-eastern China, the north-western Amazon basin and central Africa are highly affected by aerosol-induced changes in surface solar radiation. The eastern US For these five key regions, Table 5 presents absolute and percent changes in annual average radiation (total, direct and diffuse) between the control and sensitivity simulations. The eastern North America shows the largest increase in diffuse radiation among industrialized regions. Europe and China undergo a strong reduction in total and direct radiation mainly due to non-biomass burning sources. Both Mercado et al. (2009) and Chen and Zhuang (2014) simulated a consistent annual diffuse radiation due to all anthropogenic aerosols (+8.6 W m$^{-2}$; +6.2%), followed by north-eastern China and central Africa, which experience similar changes (~ +7.4–7.9 W m$^{-2}$; ~ +5.7%). Over the eastern North-America, the increase in diffuse solar radiation in East Asia; Mercado et al. (2013)estimated a increase in diffuse fraction by 25radiation maximizes during boreal summer (+13.6 W m$^{-2}$; +8.9%), with changes that are 1.6–30% over East Asia and Europe during the "global dimming" period. Due to biomass burning aerosols, the Amazon basin and central Africa record comparable decrease 5.7 W m$^{-2}$ (1.9–3.3%) higher that those observed over north-eastern China and Eurasia (Table S3 in the Supplementary Material). Driven by non-BBAs, Eurasia and north-eastern China undergo the largest reduction in total and direct radiation ; however, the Amazon basin experiences a weaker increase in diffuse radiation compared to central Africa. with a larger increase over north-eastern China

(total: $-12.3\,W\,m^{-2}$, $-6\%$; direct: $-19.4\,W\,m^{-2}$, $-26.1\%$) than Eurasia (total: $-9.5\,W\,m^{-2}$, $-4.8\%$; direct: $-14\,W\,m^{-2}$, $-23.8\%$). Over Eurasia and north-eastern China, decreases in total and direct radiation maximize during boreal summer, with changes that double those observed over eastern North-America (Table S3 in the Supplementary Material). In Central Africa and the north-eastern Amazon, on an annual average basis, BBAs drive changes in surface radiation that are similar in magnitude to those driven by non-BBAs. Yet, in these tropical ecosystems, the BBA effects on surface radiation exhibit a strong seasonal cycle with the maximum signal in the dry-fire season (boreal summer–boreal autumn, JJA–SON).

For these five key regions, our results are broadly consistent with Mercado et al. (2009) and Chen and Zhuang (2014) , with one exception. Over the Amazon basin, Chen and Zhuang (2014) simulated an aerosol-driven decrease in diffuse radiation; the authors ascribed this behavior to ~~both (a)~~ the combination of an aerosol-driven decrease in total radiation (less solar radiation to be scattered above, and subsequently under, clouds) ~~and (b)~~ with the high cloud fractions over the Amazon basin (cloud scattering effectively limits aerosol light scattering).

### 3.3.3 ~~Surface temperature and relative humidity~~

## 3.4 Aerosol pollution changes to surface meteorology

~~Compared to the global effect~~ Accounting for only the direct aerosol effect and using the fixed-SST technique, we limit the influence of pollution aerosols on ~~surface solar radiation~~ the Earth System to direct changes in surface radiation that affect the atmosphere and land-surface only. For this reason, in the ~~short-term pollution aerosols affect surface atmospheric temperature over a few regions: Europe, part of Middle-East, central Africa, the Amazon Basin (Fig. **??**a–c). Biomass burning aerosols reduce~~ following we mainly relate changes in land carbon fluxes to changes in surface radiation, surface meteorology (e.g., SAT and relative humidity) and plant conditions (e.g., transpiration, canopy temperature).

The radiation changes caused by anthropogenic aerosol pollution do not exert a statistically significant change in global and annual average SAT ~~by −0.6 to −1(−0.2 to −0.3%)in the Amazon basin; a weaker, but similar signal is observed over~~ because our experiments use fixed SSTs and do not consider aerosol indirect effects on clouds. The rapid adjustments are allowed for the atmosphere and land-surface only. For the same reasons, we do not find statistically significant changes in precipitation or in cloud water content due to anthropogenic aerosol pollution (not shown). The model does simulate statistically robust changes in annual average SAT in the two tropical key regions: the north-western Amazon Basin and central Africa (Fig. ~~**??**b). Concomitant to a cooling in the Amazon basin, surface RH increases by +1.5–3% (Fig. **??**e), with a maximum rise at the peak of the dry-fire season (+4–6% during boreal summer, Figs. S11 and S12 in the Supplement). On annual average, anthropogenic pollution aerosols drive a rise in surface RH by +1–1.5% in the eastern North-America experiences.In this region, during the growing season (boreal summer), SAT decreases by −1 to −1.5(−0.4 to −0.6%) and surface RH increases by +4–6% (Figs. S11 and S12 in the Supplement).~~ S3 in the Supplementary Material). From the sensitivity experiments, we ascertain that the SAT changes are associated with the BBAs in the tropical regions (Fig. S3 and Table S4 in the Supplementary Material). The mechanism occurs through a bio-meteorological feedback described below.

~~The evolution of surface temperature and relative humidity are tightly connected through vegetation. Cooler surface temperatures reduce canopy temperatures and favor an increase~~ Figure 5 shows changes in annual transpiration efficiency (i.e., a proxy of canopy conductance), relative humidity and canopy temperature driven by anthropogenic pollution aerosols in the three sensitivity cases (Fig. S4 for the corresponding annual percentage changes and Fig. S5 for the seasonal absolute changes, both in the Supplementary Material). In the model, photosynthesis and stomatal conductance are coupled through the Farquhar–Ball–Berry approach. Direct radiative forcing-driven (DRF-driven) increases in photosynthesis and GPP are associated with increases in canopy conductance and ~~RH, via evapotranspiration; hence, aerosol-driven cooling may lastly induce a change in the water cycle. However,~~ relative humidity (RH) via increased transpiration. Under anthropogenic aerosol pollution, transpiration efficiency shows significant modifications in all five key regions (Fig. 5 and Table 5). The north-western Amazon Basin records the largest increase in transpiration efficiency due to BBAs ($\sim 0.51\,\%$; $\sim 5\,\%$). Among industrialized regions, the largest increases in transpiration efficiency are observed in Eurasia due to all anthropogenic aerosols ($0.16\,\%$; $\sim 5\,\%$), one-third of the increases in transpiration efficiency observed in the north-western Amazon Basin. Among the five key regions, changes in canopy temperature are statistically robust only in the north-western Amazon Basin, central Africa and north-eastern China. The north-western Amazon Basin experiences the largest decrease in canopy temperature driven by BBAs ($-0.31\,\mathrm{K}$; $-0.10\,\%$), which is $\sim 0.1\,\mathrm{K}$ larger than the decrease in canopy temperature over central Africa and north-eastern China. Due to anthropogenic pollution aerosols, central Africa and north-eastern China experience similar decrease in canopy temperature ($-0.23\,\mathrm{K}$; $-0.08\,\%$), and, compared to the north-western Amazon Basin, they undergo substantial decreases in direct radiation ($-35\,\%$ in central Africa and $-29\,\%$ in ~~our experiment we do not observe robust changes in precipitation nor in total cloud cover. Reduction in surface temperature may favor plant productivity, if temperatures are above the temperature optimum for photosynthesis (25). The role of surface temperature for plant photosynthesis might be important in tropical regions where carbonaceous aerosols from biomass burning efficiently absorb incoming solar radiation and induce a cooling at the surface. Elsewhere, change~~ north-eastern China vs. $-8\,\%$ in the north-western Amazon Basin).

To summarize, in the model, reductions in the canopy temperature observed in the north-western Amazon Basin represents a positive feedback on plant productivity (further increases) in response to the DRF-driven increases. In industrial key regions such as eastern North America and Eurasia, changes in the quantity and quality of surface solar radiation ~~may~~ play the main role in affecting plant photosynthesis. In ~~the following section, we analyze~~ north-eastern China and central Africa multiple aerosol-driven ~~changes in land carbon fluxes and we link them to changes in SSR and SAT~~effects may combine to affect plant photosynthesis: changes in the quantity and quality of surface solar radiation (as in eastern North America and Eurasia) and reductions in the canopy temperature (as in the north-western Amazon Basin).

## 3.5 ~~Global sensitivity~~ Sensitivity of GPP ~~and isoprene emission~~ to aerosol pollution

~~The~~

### 3.5.1 Global-scale

Changes in the global annual average GPP flux ~~and isoprene emissions for each simulation~~ between the control and the sensitivity simulations are reported in Table 3. ~~Across all simulations global annual GPP and isoprene emission are consistent with actual estimates of the present-day carbon cycle budget (GPP: 123 ± 8, Beer et al., 2010 ; isoprene: 400–700, Guenther et al., 2006 ).~~

~~Global GPP and isoprene emission are not sensitive~~ Global GPP shows a weak sensitivity to pollution aerosols (~~Table 3~~~ 1–2 %). Global GPP is ~~reduced~~ increased by up to ~~−2.0 % (−2.4~~2.0 % (2.4 PgC $yr^{-1}$) at most ~~for SimNOant. Global isoprene emission increases by up to +2.0 % (+6.9) for SimNOant. Removal of biomass burning emissions has almost zero effects on global GPP and isoprene emission.~~

~~Under removal of all anthropogenic pollution aerosols, we observe a change in global GPPthat is half the value obtained by Chen and Zhuang (2014) (4.9). However, Chen and Zhuang (2014) removed all atmospheric aerosols and simulated a decrease in total radiation of −21.9, which is four times the reduction we simulated in total radiation (−5.2). Furthermore, they applied transient atmospheric and prognostic LAI; hence, aerosol-induced changes in environmental parameters (e.g., light, temperature, concentration) affect plant productivity as well plant phenology. In contrast with Mercado et al. (2009) , we do not ascertain a significant~~ due to all anthropogenic aerosol pollution. Biomass burning and non-biomass burning aerosols have a comparable effect on global GPP. In contrast to Mercado et al. (2009) , our model results do not suggest a substantial change in global GPP due to ~~removal of~~ pollution aerosols.

### 3.5.2 ~~Regional sensitivity of GPP to aerosol pollution~~Five key regions

Anthropogenic aerosol pollution drives regional increases in annual average plant productivity (GPP) that affect the five key regions (Fig. **??** and, for percentage changes, Fig. S6 in the Supplementary Material). The strongest increases in GPP occur in eastern North America and ~~Europe~~ Eurasia (+0.2~~–0.4; +8~~0.3 gC $m^{-2}$ $day^{-1}$; +5~~–12 %, 8 %~~) (Fig. **??**a ~~). Biomass burning aerosols drive~~ and Fig. S7a). In the north-western Amazon Basin, BBAs drive similar absolute increases in GPP ~~of~~ (+0.2–~~0.4~~(0.3 gC $m^{-2}$ $day^{-1}$; +2–5 %) ~~in the Amazon basin, central Africa and eastern Europe~~ (Fig. **??**b ~~). Industrial pollution aerosols increase GPP by +0.05~~and Fig. S7b).

Anthropogenic aerosols drive the strongest absolute enhancement in GPP in Eurasia (+0.62 PgC $yr^{-1}$; ∼ 5 %), followed by eastern North America, which experiences a third of the absolute increase in GPP but similar relative increases (+0.21 PgC $yr^{-1}$; ∼ 5 %) (Table 5). In north-eastern China, anthropogenic aerosols drive the lowest enhancement in GPP, which is one tenth of the absolute increases in GPP observed in Eurasia (+0.06 PgC $yr^{-1}$; 1.2 %; Table 5). The north-western Amazon basin and central Africa records increases in GPP that are slightly stronger than those observed in north-eastern China (+0.07~~–0.2(+2~~0.10 PgC $yr^{-1}$; 1.6~~–5 %) in the eastern US (Great Lakes region), Europe and Asia (China and South-Eastern Asia)~~2.4 %; Table 5).

In each key region, increases in GPP are governed by different aerosol types. In the industrial key regions, non-BBAs play a key role in GPP enhancement; while, in biomass burning regions (i.e., the north-western Amazon Basin and central Africa), BBAs govern GPP enhancement. In north-eastern China, BBAs do not drive any robust change in GPP; in Eurasia, BBAs drive increases in GPP that is two-third of the increases due to non-BBAs (+0.2 PgC $yr^{-1}$ vs. +0.3 PgC $yr^{-1}$; 1.5 %

vs. 2.4%) (Table 5). In eastern North-America, BBAs and non-BBAs contribute by a similar amount to GPP enhancement ($+0.1\,\mathrm{PgC\,yr^{-1}}$, ∼2%; Table 5). In central Africa, BBAs entirely control increases in GPP; whereas, in the north-western Amazon Basin, BBAs drive increases in GPP larger than the increase due to all anthropogenic aerosols and non-BBAs (~~Fig. ??c~~). ~~+~~$0.1\,\mathrm{PgC\,yr^{-1}}$, 3.4%; Table 5)

During boreal summer, anthropogenic aerosol pollution increases GPP in eastern North America and ~~Europe~~ Eurasia by up to ~~+12% (> 0.6~~+5~~–0.8,~~ 8%, 0.6–0.8 $\mathrm{gC\,m^{-2}\,day^{-1}}$ (Fig. **??**~~). Comparison between SimCTRL-SimNOant and SimCTRL-SimNOind indicates that pollution emissions from non-biomass burning sources (industry, power generation, road vehicles) drive these increases~~ a and Fig. S7c in the Supplementary Material); particularly, in Eurasia aerosol pollution from non-BBAs drive the increase in GPP (Fig. ~~**??**a vs. Fig. **??**c ). In~~ **??**c and Fig S7f in the Supplementary Material). Driven by BBAs in the dry-fire season~~(boreal summer and fall; here, only summer is shown), biomass burning aerosols increase GPP~~ , GPP increases by +0.05~~–0.4~~0.4 $\mathrm{gC\,m^{-2}\,day^{-1}}$ (+2–5%) in eastern Europe (boreal evergreen and mixed forests), and by +0.4~~–0.6~~0.6 $\mathrm{gC\,m^{-2}\,day^{-1}}$ (+5–8%) in the north-western Amazon basin (Fig. **??**b and Fig. S7e in the Supplementary Material).

~~Pollution aerosols have largest impacts on GPP for PFTs with complex canopy architectures such as deciduous broadleaf and evergreen needleleaf forests and rainforests, in agreement with observational studies (e.g., Niyogi et al., 2004; Alton et al., 2007; Cirino et al., 2014) .~~

~~Anthropogenic pollution aerosols, mainly from non-biomass burning sources, enhance plant productivity in industrialized regions such as~~ During boreal summer, Eurasia shows the largest absolute enhancement in GPP ($+1.8\,\mathrm{PgC\,yr^{-1}}$; +6%), mainly driven by non-BBAs ($+1.1\,\mathrm{PgC\,yr^{-1}}$; +3.4%) compared to BBAs ($+0.5\,\mathrm{PgC\,yr^{-1}}$; +1.5%). The absolute GPP increase in eastern North America ~~, Europe and China. These industrialized regions undergo considerable changes in surface solar radiation due to anthropogenic pollution aerosols (Fig. **??**) . The eastern US records the largest increase in diffuse radiation that likely induces a reduction in SAT and, consequently,~~ is one-third of that observed in Eurasia ($+0.5\,\mathrm{PgC\,yr^{-1}}$; +6%) (Table S3 in the Supplementary Material). In the north-western Amazon Basin, the largest enhancement in GPP occurs during boreal autumn driven by BBAs ($+0.2\,\mathrm{PgC\,yr^{-1}}$; +6%), when the largest decrease in canopy temperature ~~. On~~ is observed as well; on the contrary, ~~Europe and China experience a strong reduction in total and direct radiation. Simulated enhancement in plant productivity in the cited regions agrees with both observational and modeling studies (e.g., Niyogi et al., 2004; Steiner and Chameides, 2005; Knohl and Baldocchi, 2008; Matsui et al., 2008) . In eastern North America, Europe and China, Mercado et al. (2009) simulated a substantial land carbon uptake due to diffuse-fraction contribution between 1950 and 1980 ("global dimming" for SSR, Wild et al., 2009 ). However, the authors observed changes in land carbon uptake due to diffuse-fraction contribution that are one order of magnitude smaller than our results (+0.03–0.07; see Fig. 2d in Mercado et al., 2009 ) ; moreover, diffuse fraction seems to be unchanged over the eastern North America (see Fig. 2c in Mercado et al., 2009 ). Chen and Zhuang (2014) simulated positive aerosol effects on GPP in North America, Europe, central Africa and South and East Asia; however, they recorded the largest increase in~~ changes in surface radiations maximize during boreal summer (Table S3 in the Supplementary Material). Likewise, in central Africa, changes in surface radiations peak during boreal summer, while the largest enhancement in GPP (and decrease in canopy temperature) occur

during boreal winter (Table S4 in the Supplementary Material). The area selected to represent central Africa mostly stretches toward southern of the equator, where boreal winter corresponds to the growing season. The seasonal behavior of GPP in central Africa ~~and Asia (+0.8–1; see Fig. 4a in Chen and Zhuang, 2014 )~~suggests that the bio-meteorological feedback to canopy temperature has a larger influence on plant productivity than reduction in direct radiation.

~~Biomass burning aerosols enhance plant productivity in the Amazon basin and central Africa. These biomass burning regions observe a comparable decrease in total and direct radiation; however, the Amazon basin experiences a weaker increase in diffuse radiation, but a larger cooling compared to central Africa (Fig. **??**).~~ The GPP sensitivities to aerosol pollution in the five key regions presented in this work agree well with values from previous measurement-based and modeling studies (e.g., Niyogi et al., 2004; Steiner and Chameides, 2005; Knohl and Baldocchi, 2008; Matsui et al., 2008) . Consistent with previous measurement-based studies, pollution aerosols have largest impacts on GPP for these PFTs with complex canopy architectures (e.g., Niyogi et al., 2004; Alton et al., 2007; Cirino et al., 2014) . For instance, the five key regions are all populated by PFTs with multi-layer canopies, large canopy heights and LAIs, such as deciduous broadleaf forests, evergreen needleleaf forests, mixed forests, and tropical rainforests, which happen to be co-located with high sources of anthropogenic aerosol pollution. In the Amazon ~~basin~~Basin, previous studies ~~observed~~ measured enhancement in $CO_2$ uptake at ecosystem scale during biomass burning season; these ~~observational studies mainly attributed~~ observationally-based studies attributed the rise in $CO_2$ uptake to the increase in diffuse light, although ~~significant~~ substantial changes in surface temperature and humidity were also measured (e.g., Oliveira et al., 2007; Doughty et al., 2010; Cirino et al., 2014). ~~Notably, aerosol-induced reduction in surface temperature directly affects leaf temperature and might be important for sunlit leaves (Doughty et al., 2010) . Based on a modeling study~~Using a modeling framework, Rap et al. (2015) estimated that BBAs enhance GPP by 0.7–1.6%, ~~under~~ for an increase in diffuse radiation ~~by~~ 3.4–6.8%. ~~These perturbations are weaker than our results;~~ Rap et al. ~~(~~2015~~) stated that their results might be conservative because they do not accounted~~ Their estimated GPP sensitivity for this region is lower than values presented here because Rap et al. (2015) did not account for aerosol-induced ~~reduction~~ reductions in leaf temperature. ~~In contrast with our results, Chen and Zhuang (2014) simulated a negative aerosol effects on GPP in the Amazon basin. The authors ascribed this reduction in GPP to the high cloud fraction and water vapor concentration over the region that both reduce incoming solar radiation and, consequently, aerosol light-scattering. Due to the large cooling and the role of high cloud fraction and water vapor concentration in limiting diffuse radiation in the Amazon basin, we hypothesize that the aerosol-induced cooling is the main driver of GPP enhancement.~~

Anthropogenic aerosol pollution ~~significantly~~ substantially enhances plant productivity at a regional scale. ~~The~~ This analysis suggests that aerosol-driven ~~enhancement in GPP seems to result from three different mechanisms : (1) light scattering , which partly reduces canopy temperature (eastern US), (2)~~ enhancements in GPP result from different mechanisms that depend on region. In the model, light scattering and DRF dominates in eastern North America, reduction in direct radiation ~~(Europe and China), and (3) cooling (the Amazon basin)~~ dominates in Eurasia and north-eastern China, and tropical ecosystems (i.e., the north-western Amazon Basin and central Africa) benefit from a bio-meteorological feedback to canopy temperature.

### 3.5.3 ~~Regional sensitivity of plant isoprene emission to aerosol pollution~~

## 3.6 Sensitivity of isoprene emission to aerosol pollution

~~Compared~~

### 3.6.1 Global-scale

Changes in the global annual average isoprene emission between the control and the sensitivity simulations are reported in Table 3. Similar to GPP, ~~isoprene emission has an opposite and uneven~~ global isoprene emission shows a weak sensitivity to pollution aerosols (~~Fig. ??) .~~ ~ 1–2 %). Global isoprene emission decreases by up to 1.7 % (6.9 $\mathrm{PgC\,yr^{-1}}$) for SimNOant. Global isoprene emissions is sensitive to industrial emissions but not to biomass burning emissions.

### 3.6.2 Five key regions

Anthropogenic aerosol pollution drives a ~~–~~decrease in annual average isoprene emission of ~~–0.5 to –1(–2~~ ~~–0.5 $\mathrm{mgC\,m^{-2}\,day^{-1}}$ to –1 $\mathrm{mgC\,m^{-2}\,day^{-1}}$ (–2 %~~ to −12 %) over Europe and China (Fig. **??** ~~a). Pollution emissions from non-biomass~~ and Fig. S7 in the Supplementary Material). Non-biomass burning sources are ~~the main drivers, as comparison between SimCTRL-SimNObb and SimCTRL-SimNOind indicates (Fig. **??**b vs. Fig. **??**c). During boreal summer~~mainly responsible for the observed regional decrease in annual average isoprene emission. In peak growing season in the temperate and tropical zone, pollution aerosols do not affect isoprene emission (Fig. ~~**??**).~~

~~On both annual and seasonal average , Europe and China have a lower isoprene flux compared to the main isoprene source regions (the Amazon basin, central Africa and the eastern US) . Under aerosol pollution ,~~ S8 in the Supplementary Material). On an annual average basis, anthropogenic aerosols mainly from non-biomass burning sources (i.e., BBAs have no robust effect) drive the largest decreases in isoprene source over north-eastern China (−1.04 $\mathrm{TgC\,yr^{-1}}$; −5.6 %) and Eurasia (−0.86 $\mathrm{TgC\,yr^{-1}}$; −2.7 %) (Table 5).

In response to aerosol pollution from non-biomass burning sources Europe and China ~~record a larger decrease in~~ show a large decrease in annual average direct radiation (~~< −40 %), and a weaker~~ −24–26 %), but a similar increase in diffuse radiation (~~+8~~+3~~–10 %) , compared to high-isoprene regions such as the eastern US and central Africa ($\Delta_{\%}$ Direct Radiation from −40 to −30 %; $\Delta_{\%}$ Diffuse Radiation > +10 %) (Fig. **??**a–c). Over~~ 5 %) as eastern North America (Table 5). Hence, over Europe and China aerosol-driven reduction in direct light is not adequately sustained by an increase in diffuse radiation~~and may considerably limit~~ , which limits isoprene emission, due to ~~a~~ the reduced light supply (reduced $J_e$). ~~Recently,~~ Stavrakou et al. ~~(~~2014~~) analyzed the interannual variability in isoprene source in Asia for the period 1979–2012 by applying sensitivity simulations, in-situ measurements and formaldehyde (used as VOC proxy) spaceborne observations. Compared to a well-established inventory of biogenic emissions, the authors found that isoprene source in Asia is a factor of 2 lower. The decreasing trend in isoprene source is confirmed by satellite-derived isoprene emission estimates: inferred isoprene fluxes decrease by 2.1over Chinabetween 2007 and 2012 (~~Stavrakou et al., 2014~~) . In China~~Thus, in Europe and China, we find that

lower SSR caused by the aerosol loading drives a decrease of ~ 10% in the isoprene source, on an annual average. Over Europe and China, the sensitivity of isoprene emission to aerosol pollution diverges from GPP sensitivity. We discuss below this divergent regional response of GPP and isoprene to aerosol pollution.

### 3.6.3 Divergent regional sensitivity of GPP and isoprene emission to aerosol pollution

The divergent response over Europe and China (i.e., aerosol-driven GPP increase and concomitant isoprene decrease) suggests a "decoupling" between the modeled processes of photosynthesis and isoprene emission over these regions. In Europe and China, we posit that aerosol-induced reduction in direct radiation drives isoprene decreases and concomitant GPP increases. In the model, the rate of net uptake results from the minimum of three competing processes: supply of the Rubisco enzyme ($J_c$); supply of light ($J_e$) and supply of nutrients ($J_s$). As theoretical and observational studies have demonstrated, the aerosol effect on plant photosynthesis strongly depends on the canopy separation into sunlit and shaded leaves. These two parts of the canopy have different responses to the change in light partitioning driven by aerosols (Knohl and Baldocchi, 2008) . Under low PAR, both shaded and sunlit leaves are in a light-limited environment ($J_e$ controls the photosynthetic rate). Under high PAR, sunlit leaves are Even when photosynthesis is light-saturated and in a (in a Rubisco-limited environment( $J_c$ controls the photosynthetic rate), while shaded leaves are in a light-limited environment ($J_e$). Hence, sunlit canopy photosynthesis depends on both direct and diffuse light, and on both the $J_c$ and $J_e$ photosynthesis rate; while shaded canopy photosynthesis is directly influenced by diffuse light and depends on the $J_e$ photosynthesis rate. The aerosol light-scattering directly influences $J_e$, hence it mainly affects shaded leaves (Matsui et al., 2008; Chen and Zhuang, 2014) .

At the same time, in the model, isoprene emission depends on light supply ($J_e$), hence isoprene emission ), isoprene emission continues to rise under increasing PAR , even when photosynthesis is light-saturated (in a Rubisco-limited environment) (Morfopoulos et al., 2013). This response was divergent response has been observed at the ecosystem scale and showed an important dependence on both light quantity and temperature (Sharkey and Loreto, 1993). At 20 °C 20° C and at any photon flux, the authors recorded nearly no isoprene emission; at 30 °C 30° C isoprene emission increased with photon flux up to 16001600 $\mu mol\ m^{-2}\ s^{-1}$, while photosynthesis was already saturated; at 40 °C40° C, isoprene emission maximized at 10001000 $\mu mol\ m^{-2}\ s^{-1}$, afterwards it decreased when the photon flux raised to 1600was raised to 1600 $\mu mol\ m^{-2}\ s^{-1}$.

In the model, isoprene emission in the Amazon basin is not sensitive to pollution aerosols. Over this region, absorbing and scattering aerosols from biomass burning significantly reduce direct radiation and surface temperature, with a smaller increase in diffuse radiation compared to central Africa. Isoprene synthase has a larger temperature optimum (35 °C) compared to photosynthesis (25 °C), hence isoprene emission will decrease under cooling conditions. Since in the Amazon basin isoprene emission does not respond to the aerosol-driven decrease in direct radiation, implying offsetting thermal and radiative responses, we deduce that aerosol-driven cooling in the Amazon basin plays a role in increasing the plant productivity there on an annual and seasonal scale.

We posit that the aerosol-induced rise in diffuse radiation (the diffuse fertilization effect) drives enhancement in plant productivity in the eastern US This regionexperiences one of the largest increase in diffuse light that may enhance GPP via

the increase of $J_e$ photosynthesis rate in shaded leaves. The concomitant cooling observed in the eastern US may limit the sensitivity of isoprene emission to an increase in the supply of light.

To conclude, anthropogenic aerosols affect GPP and isoprene emissions via three mechanisms: (1) light scattering, (2) cooling, and (3) reduction in direct radiation. We suggest that the dominant aerosol-driven mechanism that influences land carbon fluxes varies across regions: (1)light scattering, and concomitant cooling, dominates in the eastern US; (2) cooling dominates in the Amazon basin; and (3) reduction in direct radiation dominates in Europe and Chinanorth-western Amazon Basin, annual average isoprene emission increases are simulated in response to BBAs ($+0.4\,\mathrm{TgC\,yr^{-1}}$; +2.4%) (Table 5), although the area of statistical significance is small. In this region, the influence of increases in GPP on isoprene emission over-rides the influence of the cooler canopy temperatures (Table 5).

## 4 Discussion and conclusions

Aerosol-induced effects on land carbon fluxes (GPP and isoprene emission) were investigated using a coupled vegetation–chemistry–climate coupled global vegetation-chemistry-climate model. By performing sensitivity experiments, we isolated the role of pollution aerosol sources (all anthropogenic, biomass burning and non-biomass burning).

We acknowledge three main limitations in our study. Firstly, we tackled the direct aerosol effects only and did not consider 1st and 2nd indirect effects of aerosols. Hence, we are partly missing the impact of aerosol–cloud interactions on the land carbon fluxes. Secondly, we used the fixed SST-technique, hence we accounted only for rapid adjustments of land surface climate to aerosol radiation perturbation. Thirdly, we prescribed LAI, hence plant phenology does not respond to the changes in aerosol pollution. We are likely underestimating the magnitude of aerosol-induced effects on plant productivity by not including these feedbacks.

Despite these limitations, our Our results suggest that global-scale land carbon fluxes (GPP and isoprene emission) are not sensitive to the direct effects of pollution aerosols, even under a robust overall SSR (direct+diffuse) global change ($\sim 9\,\%$). We found a significant, but divergent , sensitivity substantial but divergent sensitivities of GPP and isoprene emission to pollution aerosols at the regional scale, in locations where complex canopies dominatea regional scale. In eastern North America and EuropeEurasia, anthropogenic pollution aerosols (mainly from non-biomass burning sources) enhance GPP by +8+5–12% 8% on an annual average, with a stronger increase during the growing season (> 12%). In the north-western Amazon basin and central Africa, biomass burning aerosols increase GPP by +2–5% on an annual average (+5–8% at the peak of the dry-fire season in the north-western Amazon basin). In Europe and Eurasia and north-eastern China, anthropogenic pollution aerosols (mainly from non-biomass burning sources) drive a decrease in isoprene emission of −2 −2% to −12% on annual average. This study highlights the importance of accounting for both aerosol-induced radiative and thermal effects on plant productivity (Steiner and Chameides, 2005) . Our model results imply that a further reduction reductions of anthropogenic pollution aerosols over Europe and China below the present day loadings may trigger an enhancement in isoprene emission, with consequences on ozone production/destruction (depending on $_x$ levels), methane lifetime and secondary aerosols (through BSOA production). In future research, we will (1) assess co-impacts of aerosol indirect effects, (2) apply a fully coupled

ocean-atmosphere global climate model (GCM) to quantify the long-term aerosol climate effects (e.g., Koch et al., 2009) ; (3) apply a full for ozone and aerosol air quality.

We acknowledge three main limitations. Firstly, we tackled the direct aerosol effects only and did not consider 1st and 2nd indirect effects of aerosols such that we are partly missing the impact of aerosol-cloud interactions on land carbon fluxes. Secondly, we used the fixed SST-technique, hence we accounted only for rapid adjustments of land surface climate to aerosol radiation perturbation. Thirdly, we did not include feedbacks from dynamic LAI and phenology that may lead to an underestimation of the effects of aerosol-induced effects on plant-productivity. Future research will address these three limitations. Future changes in regional atmospheric aerosol loadings will have substantial implications for the regional land carbon cyclemodel with dynamic LAI and tree growth, and respiration responses (Yue and Unger, 2015) .

*Author contributions.* S. Strada and N. Unger designed the experiments. S. Strada performed the simulations. S. Strada and N. Unger prepared the manuscript.

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**Table 1.** Global annual average of aerosol column burden (ACB, mg $m^{-2}$) ~~and of the effective radiative forcing (ERF) for aerosol–radiation interactions ()~~ as simulated by NASA ModelE2-YIBs in the control and sensitivity present-day ~~simulation (20 run years)~~ simulations for, in the order: sulfates, nitrates, organic (OC) and black carbon (BC) from industrial (ind) and biomass burning (bb), and secondary organic aerosols (SOA). ~~Empty cases~~ Cases filled with "–" refer to negligible values of ACB (i.e., order of magnitude~~is~~: pg $m^{-2}$ ~~for ACB; for ERF~~). For sensitivity simulations, percentage values in parentheses indicate contribution of target emissions (i.e., anthropogenic, biomass burning and non-biomass burning) to each aerosol component.

| Simulation | ACB (mg $m^{-2}$) | | | | | | |
|---|---|---|---|---|---|---|---|
| | $SO_4$ | $NO_3$ | $OC_{ind}$ | $OC_{bb}$ | $BC_{ind}$ | $BC_{bb}$ | SOA |
| SimCTRL | 2.41 | 5.16 | 0.48 | 0.97 | 0.17 | 0.09 | 1.37 |
| SimNOant | 1.55 (35.68 %) | 0.69 (86.63 %) | – | – | – | – | 0.39 (71.53 %) |
| SimNObb | 2.42 | 3.54 (31.40 %) | 0.48 | – | 0.17 | – | 1.14 (16.79 %) |
| SimNOind | 1.51 (37.34 %) | 1.47 (71.51 %) | – | 0.82 (15.46 %) | | 0.08 (11.11 %) | 0.90 (34.31 %) |


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**Table 2.** Global annual average of effective radiative forcing (ERF) for aerosol–radiation interactions ($W\,m^{-2}$) as simulated by NASA ModelE2-YIBs in present-day simulations for, in the order: sulfates, nitrates, organic (OC) and black carbon (BC) from industrial (ind) and biomass burning (bb), and secondary organic aerosols (SOA). Global annual average ERF is calculated as the difference between the control experiment (SimCTRL) and sensitivity experiments: SimNOant, without all anthropogenic emissions; SimNObb, without biomass burning emissions; and SimNOind, without anthropogenic emissions except biomass burning. Percentage values in parentheses specify the contribution of target emissions (i.e., anthropogenic, biomass burning and non-biomass burning) to the ERF of selected aerosol component. The acronym "ns" indicates differences that are not statistically significant at the 95 % confidence level (based on a Student's t-test).

| Simulation Species | ERF ($W\,m^{-2}$) | | |
|---|---|---|---|
| | SimCTRL − SimNOant | SimCTRL − SimNObb | $OC_{ind}$ $OC_{bb}$ $BC_{ind}$ $BC_{bb}$ SOA SimCTRL − |
| $SO_4$ | ACB ( −0.31 (40.17 %) | 2.41 ns | 5.16 0.48 0.97 0.17 0.09 1.37 −0.30 (39 |
| $NO_3$ | ERF ( −0.38 (85.09 %) | −0.76 0.14 (30.38 %) | −0.45 0.31 (69.80 %) |
| $OC_{ind}$ | −0.06 (100.00 %) | −0.11 ns | 0.18 0.12 −0.16 0.06 (100.00 %) |
| SimNOant$OC_{bb}$ | ACB () 1.55 0.69 0.39 ERF ( −0.11 (100.00 %) | −0.45 0.11 (100.00 %) | −0.07 −0.06 0.01 (9.43 %) |
| SimNObb$BC_{ind}$ | ACB ()2.42 3.54 0.48 0.17 1.14 ERF (0.18 (100.00 %) | −0.76 ns | −0.31 −0.06 0.18 −0.13 (100.00 % |
| SimNOind$BC_{bb}$ | ACB (0.12 (100.00 %) | 1.51 0.12 (100.00 %) | 1.47 0.82 0.08 0.90 0.02 (11.42 % |
| SOA | ERF (0.10 (63.48 %) | −0.46 0.03 (15.86 %) | −0.14 −0.10 0.10 −0.11 0.05 (29.27 |

**Table 3.** Global annual average gross primary productivity (GPP), isoprene emission and shortwave visible (SW VIS) total, direct and diffuse solar radiation as simulated by NASA ModelE2-YIBs in the control and sensitivity present-day simulations(20 run years). For sensitivity simulations, percentage changes compared to the control simulation are indicated in parentheses and reported only if changes are statistically significant at the 95 % confidence level.

| Simulation | GPP | Isoprene | SW VIS Solar Radiation | | |
|---|---|---|---|---|---|
| | | | Total | Direct | Diffuse |
| | ($Pg\,C\,yr^{-1}$) | ($Tg\,C\,yr^{-1}$) | | ($W\,m^{-2}$) | |
| SimCTRL | 116.0 | 402.8 | 230.9 | 80.3 | 150.6 |
| SimNOant | 113.6 (−2.1 %) | 409.7 (+1.7 %) | 236.1 (+2.3 %) | 89.3 (+11.2 %) | 146.8 (−2.5 %) |
| SimNObb | 114.8 (−1.0 %) | 402.9 | 232.6 (+0.8 %) | 83.2 (+3.6 %) | 149.4 (−0.8 %) |
| SimNOind | 114.7 (−1.1 %) | 407.8 (+1.2 %) | 234.7 (+1.7 %) | 86.7 (+8.0 %) | 148.0 (−1.7 %) |

**Table 4.** Linear correlation Pearson's coefficient (Pearson's $R$), Pearson's $R$ squared ($R^2$) and root-mean-squared error (RMSE) as computed for model evaluation for annual and seasonal average coarse aerosol optical depth (AOD) and gross primary productivity (GPP). Performances of the NASA ModelE2-YIBs in the control present-day simulation ($\sim$ 2000s) are compared to: (1) MODIS AOD (at 550 nm; averaged over 2000–2007) for NASA ModelE2-YIBs $PM_{10}$ optical depth, and (2) global FLUXNET-derived GPP product (averaged over 2000–2011). Only boreal summer (JJA) and winter (DJF) seasonal averages are reported.

| ~~Simulation~~Variable | ~~GPP~~Average | ~~Isoprene~~Pearson's $R$ | ~~Total~~$R^2$ | ~~Direct~~$RMSE$ |
|---|---|---|---|---|
| ~~Diffuse~~AOD | ~~()~~ Annual | ~~()~~ 0.679 | 0.461 | 0.054 |
| ~~SimCTRL~~ | ~~116.0~~ JJA | ~~402.8~~ 0.769 | ~~230.9~~ 0.591 | ~~80.3~~ ~~150.6~~ 0.064 |
| ~~SimNOant~~ | ~~113.6~~ DJF | ~~409.7~~ 0.591 | ~~236.1~~ 0.349 | ~~89.3~~ ~~146.8~~ 0.065 |
| ~~SimNObb~~ GPP | ~~114.8~~ Annual | ~~402.9~~ 0.863 | ~~232.6~~ 0.745 | ~~83.2~~ ~~149.4~~ 1.025 |
| ~~SimNOind~~ | ~~114.7~~ JJA | ~~407.8~~ 0.782 | ~~234.7~~ 0.611 | ~~86.7~~ 1.796 |
| | ~~148.0~~ DJF | 0.899 | 0.808 | 1.137 |

**Table 5.** Absolute and percent changes in annual average shortwave visible (SW VIS) solar radiation, surface atmospheric temperature (SAT), transpiration efficieny, canopy temperature, gross primary productivity (GPP) and isoprene emission in: eastern North America, Eurasia, north-eastern China, north-western Amazon Basin and central Africa (green boxes on Fig. **??**(a)). Changes are computed between the control experiment (SimCTRL) and sensitivity experiments: SimNOant, without all anthropogenic emissions; SimNObb, without biomass burning emissions; and SimNOind, without anthropogenic emissions except biomass burning. The acronym "ns" indicates differences that are not statistically significant at the 95 % confidence level (based on a Student's t-test).

| Region | Variable | | SimCTRL − SimNOant | SimCTRL − SimNObb | SimCTRL − SimNOind |
|---|---|---|---|---|---|
| Eastern | SW VIS Solar Radiation | Total | $-12.16\,W\,m^{-2}$ | $-2.81\,W\,m^{-2}$ | $-7.85\,W\,m^{-2}$ |
| North America | | | −5.68 % | −1.31 % | −3.66 % |
| (70°–100°W; 36°–52°N) | | Direct | $-20.76\,W\,m^{-2}$ | $-3.53\,W\,m^{-2}$ | $-12.95\,W\,m^{-2}$ |
| | | | −27.73 % | −4.71 % | −17.30 % |
| | | Diffuse | $8.60\,W\,m^{-2}$ | ns | $5.10\,W\,m^{-2}$ |
| | | | 6.17 % | ns | 3.65 % |
| | SAT | | ns | ns | ns |
| | | | ns | ns | ns |
| | Transpiration Efficiency | | 0.12 % | ns | ns |
| | | | 3.72 % | ns | ns |
| | Canopy Temperature | | ns | ns | ns |
| | | | ns | ns | ns |
| | GPP | | $0.21\,Pg\,C\,yr^{-1}$ | $0.09\,Pg\,C\,yr^{-1}$ | $0.10\,Pg\,C\,yr^{-1}$ |
| | | | 4.96 % | 2.17 % | 2.45 % |
| | Isoprene | | $-0.28\,Tg\,C\,yr^{-1}$ | ns | ns |
| | | | −1.91 % | ns | ns |
| Eurasia | SW VIS Solar Radiation | Total | $-11.08\,W\,m^{-2}$ | $-1.67\,W\,m^{-2}$ | $-9.46\,W\,m^{-2}$ |
| (−10°W–80°E; 40°–65°N) | | | −5.59 % | −0.84 % | −4.77 % |
| | | Direct | $-16.88\,W\,m^{-2}$ | $-2.33\,W\,m^{-2}$ | $-13.95\,W\,m^{-2}$ |
| | | | −28.78 % | −3.97 % | −23.79 % |
| | | Diffuse | $5.80\,W\,m^{-2}$ | $0.67\,W\,m^{-2}$ | $4.49\,W\,m^{-2}$ |
| | | | 4.15 % | 0.48 % | 3.22 % |
| | SAT | | ns | ns | ns |
| | | | ns | ns | ns |
| | Transpiration Efficiency | | 0.16 % | ns | 0.10 % |
| | | | 5.09 % | ns | 3.27 % |
| | Canopy Temperature | | ns | ns | ns |
| | | | ns | ns | ns |
| | GPP | | $0.63\,Pg\,C\,yr^{-1}$ | $0.19\,Pg\,C\,yr^{-1}$ | $0.31\,Pg\,C\,yr^{-1}$ |
| | | | 4.85 % | 1.47 % | 2.41 % |
| | Isoprene | | $-0.86\,Tg\,C\,yr^{-1}$ | ns | $-0.13\,Tg\,C\,yr^{-1}$ |
| | | | −2.71 % | ns | −3.92 % |

**Table 5.** Continued.

| Region | Variable | | SimCTRL − SimNOant | SimCTRL − SimNObb | SimCTRL − SimNOind |
|---|---|---|---|---|---|
| North-eastern China | SW VIS Solar Radiation | Total | $-13.56\,W\,m^{-2}$ | ns | $-12.59\,W\,m^{-2}$ |
| (100°–120°E; 10°–35°N) | | | −6.41 % | ns | −5.95 % |
| | | Direct | $-21.45\,W\,m^{-2}$ | $-2.35\,W\,m^{-2}$ | $-19.42\,W\,m^{-2}$ |
| | | | −28.87 % | −3.16 % | −26.14 % |
| | | Diffuse | $7.89\,W\,m^{-2}$ | $1.08\,W\,m^{-2}$ | $6.83\,W\,m^{-2}$ |
| | | | 5.74 % | 0.79 % | 4.97 % |
| | SAT | | ns | ns | ns |
| | | | ns | ns | ns |
| | Transpiration Efficiency | | 0.12 % | ns | 0.10 % |
| | | | 3.18 % | ns | 2.70 % |
| | Canopy Temperature | | −0.23 K | ns | ns |
| | | | −0.08 % | ns | ns |
| | GPP | | $0.06\,Pg\,C\,yr^{-1}$ | ns | $0.06\,Pg\,C\,yr^{-1}$ |
| | | | 1.18 % | ns | 1.15 % |
| | Isoprene | | $-1.04\,Tg\,C\,yr^{-1}$ | ns | $-0.86\,Tg\,C\,yr^{-1}$ |
| | | | −5.60 % | ns | −4.64 % |
| North-western | SW VIS Solar Radiation | Total | $-4.09\,W\,m^{-2}$ | $-2.18\,W\,m^{-2}$ | $-2.52\,W\,m^{-2}$ |
| Amazon Basin | | | −1.84 % | −0.98 % | −1.14 % |
| (73°–65°W; 5°S–5°N) | | Direct | $-7.67\,W\,m^{-2}$ | ,$-4.08\,W\,m^{-2}$ | $-4.80\,W\,m^{-2}$ |
| | | | −7.86 % | −4.17 % | −4.92 % |
| | | Diffuse | $3.58\,W\,m^{-2}$ | $1.90\,W\,m^{-2}$ | $2.28\,W\,m^{-2}$ |
| | | | 2.88 % | 1.52 % | 1.83 % |
| | SAT | | −0.15 K | −0.28 K | −0.11 K |
| | | | −0.05 % | −0.09 % | −0.04 % |
| | Transpiration Efficiency | | 0.31 % | 0.51 | 0.23 % |
| | | | 3.20 % | 5.19 | 2.31 % |
| | Canopy Temperature | | −0.17 K | −0.31 K | −0.13 K |
| | | | −0.06 % | −0.10 % | −0.04 % |
| | GPP | | $0.07\,Pg\,C\,yr^{-1}$ | $0.10\,Pg\,C\,yr^{-1}$ | $0.05\,Pg\,C\,yr^{-1}$ |
| | | | 2.42 % | 3.42 % | 1.78 % |
| | Isoprene | | ns | $0.37\,Tg\,C\,yr^{-1}$ | ns |
| | | | ns | 2.39 % | ns |

**Table 5.** Continued.

| Region | Variable | | SimCTRL − SimNOant | SimCTRL − SimNObb | SimCTRL − SimNOind |
|---|---|---|---|---|---|
| Central Africa | SW VIS Solar Radiation | Total | $-17.40\,W\,m^{-2}$ | $-14.41\,W\,m^{-2}$ | $-8.53\,W\,m^{-2}$ |
| (10°–25° E; 10° S–5° N) | | | −8.68 % | −7.19 % | −4.25 % |
| | | Direct | $-24.80\,W\,m^{-2}$ | $-18.72\,W\,m^{-2}$ | $-10.75\,W\,m^{-2}$ |
| | | | −34.85 % | −26.32 % | −15.12 % |
| | | Diffuse | $7.40\,W\,m^{-2}$ | $4.31\,W\,m^{-2}$ | $2.22\,W\,m^{-2}$ |
| | | | 5.72 % | 3.33 % | 1.72 % |
| | SAT | | −0.19 K | −0.16 K | −0.08 K |
| | | | −0.06 % | −0.05 % | −0.03 % |
| | Transpiration Efficiency | | 0.28 % | 0.31 | 0.14 % |
| | | | 3.60 % | 4.06 | 1.79 % |
| | Canopy Temperature | | −0.23 K | −0.21 K | −0.11 K |
| | | | −0.08 % | −0.07 % | −0.04 % |
| | GPP | | $0.10\,Pg\,C\,yr^{-1}$ | $0.08\,Pg\,C\,yr^{-1}$ | ns |
| | | | 1.61 % | 1.27 % | ns |
| | Isoprene | | $-0.55\,Tg\,C\,yr^{-1}$ | ns | $-0.30\,Tg\,C\,yr^{-1}$ |
| | | | −1.89 % | ns | −1.02 % |