Impacts of global, regional, and sectoral black carbon emission reductions on surface air quality and human mortality

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Response to reviewer comments

Response to Anonymous Referee #1:

We thank Referee 1 for suggesting useful changes that enhance this manuscript. Please see our responses to comments from Referee 1 below. Referee comments are in italics.

This is a most interesting paper that examines the health impact of halving anthropogenic black carbon emissions both globally and for different world regions as well as by sector. The authors calculate avoided cardiopulmonary and lung cancer deaths associated with chronic PM2.5 exposure using PM2.5 concentrations simulated by the MOZART-4 chemistry transport model. They present interesting findings in terms of mortality impacts per unit BC emissions, and SO4 concentration increases that result from BC as well as BC+OC emission reductions. They highlight the co-benefit of BC-related emission reductions that will also tend to reduce OC. They find very little direct health benefits relating to BC emission reduction outside of the source region. The concentrations-response factor sensitivity analyses are most useful providing insights into uncertainty associated with health impact estimation. The paper is very well-written although it contains some very dense text and most of my comments relate to improved clarity for the reader.

Thank you. We appreciate these comments and hope our responses to the comments below improve clarity.

S1) The abstract discusses East Asia (EA) and South Asia (SA) but the rest of the text discuss EA and IN which is inconsistent. Discuss either IN and CH or EA and SA and modify the text and Tables and Figures accordingly.

"IN" was chosen as an abbreviation for the South Asia region, which is mainly comprised of India, to distinguish it from South America. To improve consistency between the main text and the Abstract, where there are no abbreviations, we changed the region names in the Abstract to "South Asia (India)" and "East Asia (China)":

"Most of these avoided deaths can be achieved by halving emissions in East Asia (China; 54%), followed by South Asia (India; 31%), however South Asian emissions have 50% greater mortality impacts per unit BC emitted than East Asian emissions."

S2) The abstract should mention the results of the sensitivity analyses in section 4. It would also be very worthwhile to outline the SO4 changes here.

We have added the following text to the abstract:

"We find that reducing BC emissions increases regional SO₄ concentrations by up to 28% of the magnitude of the regional BC concentration reductions due to reduced absorption of radiation that drives photochemistry."

"The choice of concentration-response factor and health effect thresholds affects estimated global avoided deaths by as much as 56%, but does not strongly affect the regional distribution."

S3) Abstract line 20, section 3.2, page 10666, line 27.section 6, page 10670, line 28. "impacts of residential BC emissions are underestimated since indoor.. excluded". Can you be sure that this statement applies globally or is more likely to apply is developing world regions? In your main body text could you provide a reference or state for which regions this underestimate is likely to be most pronounced? In the abstract you could consider adding "likely" before "underestimated".

"Likely" has been added before "underestimated" in the abstract. In Section 6, the text now reads (2nd sentence added):

"Within each region, mortality per unit emission varies little by source sector; however, impacts of residential BC emissions are underestimates since impacts due to indoor PM_{2.5} exposure are excluded. The underestimation is likely most pronounced in areas of South Asia, East Asia, and Africa that rely on solid fuel combustion for cooking and heating (Smith et al. 2004)."

S4) section 2.1, page 10658 line 14, .and section 3. 2. page 10666 lines 10-14. It seems rather strange to discuss 8 major world regions and then "plus the US alone". Table 1 doesn't contain results from "US alone". Similarly the text in section 3.2 and Fig S19 seems out of context given the focus on world regions in the paper. I'm not sure of the added value of this short paragraph of text in section 3.2 (especially as the 2nd sentence "Compared with : : :" is rather complicated and confusing) and the additional figure. If kept, this sentence should be explained more clearly and simply if possible and line 13 "causes avoided deaths in NA to increase" should be re-phrased.

We agree that the US-only reduction is disconnected from the rest of the scenarios and results. However, we do believe this scenario is of value, particularly for US policy. We have therefore moved the discussion of this scenario and the comparison to the North American reduction to the Supplemental Material.

The previous reference to the US reduction in Section 2.1 has been deleted, and the following sentence has been added in its place to improve clarity:

"Finally, to isolate the impact of emissions in the United States (US), we examine a scenario in which BC emissions are halved in the US only and compare the results with the North American reduction in the Supplemental Material."

The following text has been deleted from the main text and added to the Supplemental Material (1st sentence is new), with the phrase "causes avoided deaths in NA to increase" revised:

"To isolate the impact of emissions in the United States (US), we compare the impact of halving BC emissions in the entire North America region (includes Canada and Mexico) versus halving emissions in the US only. Halving NA emissions reduces PM_{2.5} in that region by 151 ng/m³ and avoids 4,000 (95% CI, 3,000-5,000) annual premature deaths (12 per Gg BC reduced), 91% of which occur within the US. Compared with halving BC emissions in the US only, halving all NA emissions causes 12% more avoided deaths in NA and 1.6% more in the US, mostly in the Northeast and California near national borders (Fig. S21)."

S5) section 2.2 page 10659, lines 6-9. "Population.. generally larger, indicating co-locations compared with OM and SO4". This conclusion is unclear. The populated-weighted average values in Table S2 are in all cases larger than the simple average. A clearer justification of co-location is needed. Quoting the range of values for a factor that gives the ratio of the population-average to the simple average for the different chemical species would be more convincing.

We have revised the text to read:

"Population-weighted average concentrations of all species are 1.2-3.3 times larger than simple average concentrations, reflecting co-location of emissions and concentrations with population (Table S2)."

S6) section 2.2, having performed a detailed model evaluation, these results should be commented on, in terms of how they might influence the avoided mortality estimates in the "uncertainties" section (section 5) of the text.

The following text has been added to Section 5:

"We found that BC and OC concentrations are generally lower than observations in the US and Europe and for the few available observations in China and India, likely causing our calculated impacts of halving emissions to be underestimates."

S7) section 2.3, very briefly outline why you consider only chronic and not acute or short-term exposure.

A large body of epidemiology literature over the past several decades consistently finds that both shortterm and long-term exposure to PM_{2.5} is associated with premature mortality. Cohort studies in which populations are followed over many years generally find larger mortality effect estimates than are found by short-term time-series studies. Since cohort studies follow populations over time, both short-term and long-term PM_{2.5} mortality are captured in the effect estimates, whereas short-term studies are unable to capture a large portion of risk. Risk estimates from cohort studies are widely accepted and used by regulatory agencies and researchers around the world to quantify health impacts of changes in PM2.5. We briefly expand upon the discussion on this in Section 2.3:

"We calculate CRFs using estimates of RR of chronic mortality due to total $PM_{2.5}$. The impacts of chronic $PM_{2.5}$ exposure on mortality are established by a large body of epidemiology studies and include impacts of both short-term and long-term exposure. We use RR estimates from Krewski et al. (2009), the latest reanalysis of the American Cancer Society $PM_{2.5}$ studies (e.g. Pope et al., 2002) and the largest among long-term $PM_{2.5}$ mortality studies (e.g. Laden et al., 2006)."

S8) section 2.3, if the observed range of concentrations is up to 22.2 ugm-3, how can linearity be demonstrated to 30 ugm-3?

Linearity was demonstrated by Krewski et al. (2009) to 30 μ g/m³using different years of data (1979-1983 data vs. 1999-2000). To improve clarity, the sentence now reads:

"These RRs were determined for the observed range of concentrations, 5.8-22.2 μ g/m³, and the linearity of the concentration-response relationship was also demonstrated up to 30 μ g/m³ based on different years of PM_{2.5} data (1979-1983; Krewski et al., 2009)."

S9) section 3.1, the text concerning SO4 changes shown in Fig 5a and S14 are most interesting. NO3 concentrations seem also to be slightly affected by the BC concentrations, and again the sign of the change is different in the BC and BC+OC experiments. An explanatory sentence on the NO3 changes would be useful.

The following sentences were added to Section 3.1:

" NO_3 and SOA concentrations are also formed in the atmosphere by reaction with photochemical oxidants. We find changes in regional annual average NO_3 concentrations up to 20 ng/m³ (0.3%) as a result of the BC emission reduction, but no appreciable change in SOA."

"We also find mixed directional changes in NO₃ (regional increase up to 200 ng/m³, 2.0%) and SOA (up to 47 ng/m³, 55%) that do not necessarily follow the directional change in SO₄ (Fig. 5b)."

S10) section 3.1, page 10664, line 19. It would be useful to add a further sentence describing the differences in magnitudes between the O3 and OH changes in the BC vs. BC+OC simulations.

We added percentage changes in global annual average concentration for O_3 , OH, H_2O_2 , and SO_4 to illustrate the differences in magnitude.

For the 50% global BC emission reduction:

"Through these reactions, increased concentrations of OH (global annual mean increase of 0.34%), O_3 (0.03%), and H_2O_2 (0.12%) in response to BC emission reductions lead to enhanced SO₄ production (0.13%)."

For the 50% global BC+OC emission reduction:

"We find increases in OH (global annual mean increase of 0.81%) and O_3 (0.44%) concentrations but decreases in H_2O_2 concentrations (-0.34%), resulting in mixed effects on SO_4 (-0.07% globally; Fig. S18)."

S11) section 3.1, page 10664, lines 19-26, this text is confusing and rather detailed. Describe the results of this paper, and then examine how they compare with a previous study rather than the other way around.

The mortality results for halving BC are described before the comparison with the previous study in the first paragraph of Section 3.1. The mortality results of halving BC+OC are also described before the previous study comparison as 8 times larger than the results of the BC-only emission reduction. We have now also added the value of BC+OC mortality results (1.2, 95% Cl, 0.9-1.5 million global premature deaths avoided):

"These $PM_{2.5}$ reductions are associated with ~8 times more (1.2, 95% CI, 0.9-1.5 million) global premature deaths than is estimated for halving BC alone (Table 1)."

S12) The scales on Figs 4 and 6 (also for Figs.S15-S18) are not the most informative; can these be revised?

The diverging scale is useful to show areas where BC reductions increased total $PM_{2.5}$ and $PM_{2.5}$ mortality. However, areas of red ($PM_{2.5}$ and mortality increases) were not pronounced using a symmetrical color scheme since their magnitude was much smaller than the magnitude of the decreases. We have thus retained the diverging color scale for Figs. 4 and 6 but compressed the negative portion of the scale to emphasize the areas where $PM_{2.5}$ and mortality are estimated to increase. We hope this results in a more informative scale.

We have elected not to change the scales for Figs. S17-S20 (previously Figs. S15-S18), since the diverging color scale is useful to show positive and negative changes across the different emission reduction experiments.

S13) Figs S4-S8 concerning PM2.5 = BC+OM+SO4+NO3; Fig S4 shows maximum PM2.5 concentrations of _50 ugm-3 over EA. But the addition of maximum values for the individual PM 2.5 components as given by the scales in Figs S5-S8 fall somewhat short of this (_10ugm-3). If there is no missing PM 2.5 component (?), then the scales in either Fig S4 or Figs S5-S8 are misleading. Can these be revised?

This issue is an artifact of the scales chosen to best display the information on each map. Since it may be misleading, we have changed the scale on Fig. S4 to have a maximum of 40 μ g/m³, the total of the maximum values on the color scales for Figs. S5-S8.

S14) Figs S9-S10, the text in section 2.2 discusses the results of these figures in terms of underestimates and overestimates model results. However the plots have a large number of green-coloured points. It is impossible to know if these points represent underestimates or overestimates due to the choice of scale which spans negative to positive values). Revise this colour scale for clarity and text in Section 2.2 if necessary.

The color scale for these figures was chosen to emphasize the points with the largest discrepancies (positive or negative) between the simulated and observed concentrations. Although that comes at the expense of differentiating the points in the middle of the spectrum, the points in the middle are those that have the greatest agreement between simulated concentrations and observations. Since we are comparing very large gridcells to point measurements, any close agreement between simulated and observed values indicates that the model produces reasonable concentrations, regardless if the slight discrepancy is positive or negative. We have elected to maintain the color scale since the values of greatest importance for understanding potential model biases are those at the extreme ends of the scale.

S15) Fig S17 doesn't look much different from Fig S15, is it necessary?

Figures S19 (previously Fig. S17) and S20 (previously S18) are included to demonstrate that BC affects oxidant and sulfate concentrations not just for the global emission perturbation scenarios, but also for the regional scenarios. Figure S17 (previously S15) looks similar to Figure S19 because global emissions are so dominated by emissions from East Asia; therefore the pattern of concentration changes for the East Asian emission reduction scenario is similar to the concentration changes from the global emission reduction scenario. We do believe that both figures are necessary to support our explanation of the oxidant and sulfate changes resulting from BC emission changes.

Technical corrections:

T1) Abstract, line 6 insert "individually" or some other phrase before "from eight world regions" for clarity.

Added "individually"

T2) Abstract, line 16: "Globally : : :1.3, 1.2". This sentence is difficult to follow. The fuller explanation given in section 3.3 would be useful here.

Added sentence in abstract from Section 3.3 (1st sentence below):

"Globally, halving residential, industrial, and transportation emissions contributes 47%, 35%, and 15% to the avoided deaths from halving all anthropogenic BC emissions. These contributions are 1.2, 1.2, and 0.6 times each sector's portion of global BC emissions, owing to the degree of co-location with population globally."

T3) section 2.1 page 10658, line 18 "Because each source" – add at least "emissions" and be more specific if possible.

Added "emissions"

T4) Section 2.2 page 10659, lines 18-20. Give the years and number of sites for IMPROVE and EMEP to be consistent with the information provided for the China and Indian sites. The text "surface observations ...outside : : :Europe are limited". However, the number of EMEP sites used (for BC at least in Fig 3) is about the same or less as for India or China. Re-phrase the text to reflect this.

Added the number of sites for IMPROVE and EMEP, and moved the sentences pertaining to years of observations to directly follow:

"We therefore compare simulated concentrations to surface observations mainly in remote locations from the Interagency Monitoring of Protected Visual Environments (IMPROVE; http://vista.cira.colostate.edu/improve/) network for the US (134 monitors) and the European Monitoring and Evaluation Programme (EMEP; http://www.emep.int/) network for Europe (13 monitors for BC and OC, 75 for SO₄). Although we simulate 2002 for our base case, for the model evaluation only, we ran the base case through 2003 to leverage additional observations from IMPROVE (available for both 2002 and 2003) and EMEP (available for July 2002-June 2003)." Removed the sentence on surface observations outside the US and Europe – it was unnecessary.

T5) Section 2.2 page 10659, line 23. "Each of these: ::", clarify if "these" includes also IMPROVE and EMEP measurements or not.

Changed "Each" to "All" to make clearer that the statement does include IMPROVE and EMEP.

T6) Section 2.2 page 10660 lines 5-16. Add "USA" after "after "Northeast" etc and "Europe" after "in the West", to avoid confusion with World regions.

Clarified text by adding "US" and "Europe" after geographic references.

T7) section 2.2 page 10660, lines 15-25, clarify text concerning "lower and "higher" simulated values as compared to observations. Are simulated BC concentrations for EMEP locations not always lower in Fig 3? Add generally" before higher" when referring to SO4 for EMEP locations in Fig. S10. In Fig S11 simulated values are not always lower for regional and urban locations. Relate the sentence "Measurement methods : : : potentially higher EC than : : :EMEP" to the results in Fig 3.

Changed "generally lower" to "lower" for the EMEP comparison of BC.

Added "generally" before "higher" for EMEP comparison of SO₄.

Changed "lower" to "often lower" for regional and urban locations in Fig S13.

Added reference to Fig. 3 for the "Measurement methods..." sentence.

T8) section 3.1, page 10663, line 7, add "by" before "25-49".

Added "by"

T9) Section 3.1 page 10663, line 11. Fig 6 is not described where it is referenced and is not that informative with its current scale. (see comment S12).

Rearranged the text to reference Fig. 6 where it is stated that >80% of deaths occur in EA and IN:

"We estimate that these PM_{2.5} reductions would avoid ~157,000 (95% CI, 120,000-194,000) annual premature deaths worldwide (Table 1), over 80% of which occur in EA (81,000, 95% CI, 61,000-100,000) and IN (48,000, 95% CI, 37,000-59,000; Fig. 6)."

We addressed the scale issue in our response to S12.

T10). Check % or ratio results in sections 3.2, 3.3 and 6 as compared to the values entered in Table 1, Table 3 or given earlier in the text.

Page 10665 line 7-33% 32% is correct.

Page 10667 line 4-1.3 The 46% and 1.3 ratio for residential should be 47% and 1.2

Page 10667 line 9- 15%, 19%, 59% Should be 15%, 20%, and 60%

Page 10670, line 21- 54% 54% is correct. The 53% given in Section 3.2 is also correct. The difference is due to the fact that when BC emissions are reduced in EA only, there is a PM_{2.5} mortality increase in NA,

EU, and AF/ME (negative values), causing the estimated deaths in the EA receptor region to be a larger percentage of the total (54% vs. 53% occurring in the EA receptor region when all global BC emissions are halved).

T11) section 3.2, page 10665, lines 25-26, add a reference to Table 1 here; values in Table 1 are slightly different from the values given in the text.

The correct reference is Table 2, which we have now added. Values in Table 2 (and here in the text) are slightly different from those in Table 1 because of the increase in $PM_{2.5}$ mortality in some regions following regional BC emission reductions. For example, when all global BC emissions are halved, SO4 increases slightly in EA causing the total $PM_{2.5}$ mortality reduction to be 81,000, compared with 85,000 when only EA emissions are reduced.

T12) section 3.2 page 10666, line 2), refer to Table S3 here.

Added reference to Table S3.

T13) section 3.2, page 10666, lines 7-9 "This is likely : : :smaller –per- unit: : :", this sentence is confusing; is Table 2 the right Table to refer to?

Revised text to read: "Reducing BC emissions in IN is more effective at reducing within-region $PM_{2.5}$ (2.1 ng/m³ $PM_{2.5}$ reduction per unit BC emission reduced in IN versus 1.3 in EA; Fig. 1 and Table 2). IN also has higher baseline cardiopulmonary mortality rates (Table S3)."

T14) section 3.3, page 10666, line 19. Fig 1 does not have a global category- this would be useful.

Added global category to Fig. 1.

T15) section 3.3 page 10667, line12, this text is a repeat of text at the beginning of that paragraph.

Removed this sentence.

T16) section 3.3 page 10667, line16, "the same pattern"- be more specific do you mean the same ranking? Re-phrase "The only exception: : :"

Changed "pattern" to "ranking." Revised "The only exception..." to read: "However, reducing BC emissions from the transportation sector in EA has a smaller per unit impact on mortality relative to the residential and industrial sectors."

T17) section 4, page 10667, line 26, is "much higher" more appropriate?

Added "much"

T18) section 4, page 10668, line 4, "marginal" is confusing.

Changed "marginal" to "per-unit"

T19) section 4, page 10668, line 11, add "as compared to the standard 50% global BC reduction experiment" for clarity.

Added "compared with no threshold."

T20) section 4, page 10668, line 16, it is difficult to see any reduction for SE/AU- is it worth mentioning?

We believe it is worth mentioning to demonstrate that the regions where estimated deaths are affected by the low-concentration thresholds are the least polluted, even if the difference is only slightly visible on the figure.

T21) section 5, page 10669, line 10, re-phrase "and a component of OM".

Rephrased to "and the primary component of OM"

T22) Fig 2- it is difficult to see any BC contribution (3-5% is stated in section 2.2) in most regions in Fig 2. Is it possible to enlarge the figure or expand the low values for clarity?

We considered artificially increasing the BC concentration (e.g. x2 or x10) so that fraction is more visible on the graph, but since concentrations of each component are given in Table S2, we felt doing so might add confusion without adding too much value.

T23) In the caption for Fig 3, re-iterate what the dashed lines represent.

Added "Dashed lines represent the 1:2 and 2:1 lines indicating agreement within a factor of 2."

T24) In the captions for Figs. 5 and S15, add that negative values represent increases for clarity.

Added "Negative values indicate increases." to the captions for Figs. 5, S16 (previously S14), and S17 (previously S15).

T25) In the caption for Table S2, it would be helpful to remind the reader than the total PM2.5 values are given in Table 1.

Added "Total PM_{2.5} values are given in Table 1." to the caption for Table S2.

T26) What do the black colours in Fig S15. represent?

The color scale in Figs. S17-S20 (previously S15-18) has been corrected so that the black areas are now dark red as they should be.

Response to Anonymous Referee #2:

We thank Referee 2 for suggesting useful changes that enhance this manuscript. Please see our responses to comments from Referee 2 below. Referee comments are in italics.

This manuscript evaluates the global/regional health impacts of halving black carbon emissions based on variety of sensitivity tests. The methodology is scientifically sound and the results are policy relevant. It supports additional motivations to mitigate black carbon emission which may have a large positive effect on global warming. I recommend it being published in Atmospheric Chemistry and Physics after the following major and minor issues have been addressed:

Thank you – we hope our responses below adequately address your comments and believe the paper has improved as a result.

1. The key assumption of this study is the toxicity of black carbon is equal to that of PM2.5, which could result in substantial uncertainties of the results. Usually epidemiological studies derive concentration-response relationships based on temporal/spatial changes of total PM2.5 mass. In section 2.3, the authors show some evidence that PM2.5 mixtures with high BC fractions have stronger associations with mortality than other mixtures, but comment on "evidence for differential toxicity of BC and BC-containing mixtures remains inconclusive", and therefore "assume all mixtures of PM2.5 are equally toxic". The discussion here is insufficient and needs additional analysis on whether or not the assumption is appropriate based on existing epidemiological/toxicological studies.

We have now expanded the discussion of differential toxicity of PM_{2.5} mixtures, and although we believe an extensive review of the literature is not appropriate here, we now reference a draft report by Industrial Economics Incorporated, written for the US EPA, on uncertainty analyses to support the second section 812 Benefit-Cost Analysis of the Clean Air Act, where this issue is discussed in detail. The revised text now reads:

"Some evidence suggests that air pollution mixtures with high BC fractions, "black smoke," "diesel $PM_{2.5}$," and "traffic $PM_{2.5}$," have stronger associations with mortality than other mixtures (Cooke et al., 2007; Brunekreef et al., 2009). Some studies that use ambient BC concentrations as a marker for air pollution mixtures also find stronger associations with mortality than those using total $PM_{2.5}$ (Ostro et al., 2007, 2008; Bell et al., 2009; Peng et al., 2009; Smith et al., 2009). However, these studies are subject to large measurement and exposure error since BC is very spatially heterogeneous (Bell et al., 2010). Furthermore, many $PM_{2.5}$ constituents are correlated, subjecting single-pollutant risk estimates to confounding by co-pollutants and often preventing definitive conclusions about their relative importance to risk (Smith et al., 2009). The body of evidence for differential risk of $PM_{2.5}$ components, including BC, is not as robust as for long-term $PM_{2.5}$ (Smith et al., 2009; US EPA, 2009). We therefore assume that all mixtures of $PM_{2.5}$ are equally potent in causing premature mortality, and use the change in total $PM_{2.5}$ in Eq. (2)."

2. It is nice to have a discussion on the feedbacks of changing BC emissions on sulfate concentrations. However, BC aerosols absorb/scatter radiation, which not only influences photolysis rates, but also changes the lapse rate of atmosphere, properties of CCN, and therefore influences cloud and precipitation. The former may influence atmospheric circulation and the latter will increase/decrease aerosols' wet deposition. Currently, it is not clear whether or not these feedbacks are being included in this study and how they may influence the results.

This is a good point. We use a chemical transport model (CTM) with meteorology as input to the CTM, rather than a coupled chemistry-meteorology model that would also account for feedbacks of chemistry on meteorology. However, our CTM does include a module that allows for feedback of aerosols on photolysis rates, but not on atmospheric dynamics. To clarify this, we added discussion to the Methodology and Discussion sections.

The following sentence in Section 2 on Methods was revised:

"An online photolysis scheme accounts for the impact of aerosols on photolysis rates, affecting production of photochemical oxidants (Tie et al., 2005; Emmons et al., 2010b), however, aerosol feedback on meteorology (including atmospheric circulation and interactions with clouds) is excluded."

The following sentence was added to Section 5 on Uncertainties:

"While we include feedback of aerosols on photolysis rates, we exclude the impacts of aerosols on meteorology, including on the atmospheric lapse rate, which would affect circulation, and cloud condensation nuclei, which would affect aerosol wet deposition rates."

3. Only annual mean surface observations are used to evaluate model results. How about the model performance on seasonal variability and vertical profile of BC concentrations? More model evaluations on BC are needed.

We focus here on evaluating annual average concentrations at the surface (as mentioned in the title of our paper), the quantities we report as concentrations and use for the health impact assessment in the results section. While evaluation of the vertical profile of concentrations is outside the scope of this paper, we plan to write a follow-on paper focusing on column concentrations and radiative forcing impacts, where we will consider evaluating vertical profiles as they are pertinent to those results. While monthly concentrations are not reported in this paper nor are they used for the health impact assessment, we agree on the value of including such an evaluation for BC and have added monthly comparisons with observations in the Supplemental Material.

The following sentence was added to the main text in Section 2.2:

"See the Supplemental Material for comparisons of simulated monthly concentrations with IMPROVE and EMEP observations (Figs. S9 and S10)."

Minor comments:

1. Page 10655, "but all PM2.5 components are thought to be damaging to health". Need a reference here.

Added reference to Krewski et al. (2009).

2. Pages 10656-10657, need to describe how SOA is simulated in this study. Are there any mechanisms that the change of BC will influence the production of SOA?

In MOZART-4, SOA is linked to gas-phase chemistry through the oxidation of monoterpenes and toluene (Emmons et al., 2010). We would therefore expect SOA mass to be affected by the BC and BC+OC reduction scenarios, through the associated changes in oxidant concentrations.

The following sentences were added to Section 3.1:

" NO_3 and SOA concentrations are also formed in the atmosphere by reaction with photochemical oxidants. We find changes in regional annual average NO_3 concentrations up to 20 ng/m³ (0.3%) as a result of the BC emission reduction, but no appreciable change in SOA."

"We also find mixed directional changes in NO₃ (regional increase up to 200 ng/m³, 2.0%) and SOA (up to 47 ng/m³, 55%) that do not necessarily follow the directional change in SO₄ (Fig. 5b)."

3. Middle of Page 10657, dry deposition is set to 0.1 cm s-1. However, a number of previous studies use surface resistant method to simulate the dry deposition velocities of aerosols, and find dry deposition velocities could range from 0.02 to 0.8. The authors may comment on the effects of changing tuning variables (e.g., dry deposition velocity) on the final results.

Added "and dry" to the sentence on aging and wet deposition uncertainties in Section 5 and the 2nd sentence below:

"Simulated BC concentrations vary widely among global CTMs due to differing assumptions for emissions and parameterization of aerosol processes, such as aging and wet and dry deposition rates (Koch et al., 2009; Vignati et al., 2009). Underestimation of deposition fluxes would cause overestimation of $PM_{2.5}$ concentration and mortality impacts, and vice versa."

4. Page 10658, second paragraph, sensitivity tests are made based on 50% reduction of anthropogenic BC emissions, including residential, industrial and transportation sectors. Given the fact that BC aerosols disturb photochemistry in the model, many other chemical fields will change as well. Therefore, there would be some non-linearity involved in the system, such as sulfate and SOA. The authors should briefly explain the reasons to halve BC emissions from science/policy perspectives (e.g., why not 20% or 80%?)

Added the sentence:

"The 50% reductions are chosen to simulate realistic but ambitious policy targets, while producing changes in PM_{2.5} that are sufficiently large to be analyzed in all reduction scenarios."

5. Section 4 "Sensitivity analysis": may change to "Sensitivity analysis on CRFs".

Changed to "Sensitivity of results to concentration-response factors"