Interactive comment on "The impact of residential combustion emissions on atmospheric aerosol, human health and climate" by E. W. Butt et al.

E. W. Butt et al.
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We would like to thank Jeffery Pierce and the anonymous reviewer for their helpful and constructive comments. Below we have responded to each comment in turn and made alterations to the manuscript where appropriate (shown enclosed in "speech marks and italic font"). The referee comments are shown first in grey shading and the author's response is shown below in normal font.

Response to Jeffery Pierce Referee #1

Comments:

1. Introduction
   The difference between 'residential emissions' and 'residential solid fuels' is not made until later in the paper. Might be useful to describe in paragraph 2.

The referee correctly states that we make the distinction between 'residential emissions' and 'residential solid fuels' in Section 2.2. We agree that making this distinction earlier in the paper is more helpful for the reader. We therefore have modified the original manuscript to include this distinction in paragraph 2 of the Introduction section.

The sentence on Page 20452, line 1 has been modified.
"Combustion of fuels within the household typically for cooking and heating needs, known as residential fuel combustion, is an important source of aerosol emissions with impacts on air quality and climate (Ramanathan and Carmichael, 2008;Lim et al., 2012)"

The sentence on Page 20452, line 26 has been modified.

"In China, residential combustion of both biomass (also referred to as biofuel) and coal is important, whereas across other parts of Asia and Africa, residential combustion of biofuel is dominant (Lu et al., 2011;Bond et al., 2013).

2. P20454: In review of previous work, please add Kodros et al. (2015) to this review (I would have liked us to include your paper too. . . I did not realize that you were going to submit around the same time we were submitting our revisions, sorry). Similarly it would be good to include comparisons to this paper when looking at number changes and climate changes. Emissions inventories are not exactly the same (biofuel vs. residential), but largely overlapping. It seems like globally the results are similar but regionally there can be some big differences.

We thank the referee for the pointing us to this resent publication in ACP. This publication was cited in an earlier version of our manuscript, but was removed because at the time it wasn't fully accepted in ACP. We therefore have modified the original manuscript and cited Kodros et al. (2015) twice on page 20454, line 17.

In addition, we have added a sentence summarising the findings from Kodros et al. (2015) on page 20455, line 3.
"However, a recent detailed global modelling study found that the climate effects of residential biofuel combustion aerosol is largely unconstrained because of uncertainties in emission mass flux, emitted size distribution, optical mixing state, and ratio of BC to POM (Kodros et al., 2015)."

3. P20455: In the health effects discussion, please discuss that with a coarse CTM that one only captures regional (~200 km mean values) air-quality effects on health, not indoor or even intra-village concentrations. If the models were run at higher resolution, health effects would likely be stronger (and this still does not even include indoor exposure).

We have added two additional sentences on page 20455, line 15 to highlight the referee's comment.
"These estimates rely on $PM_{2.5}$ concentrations from coarse global models with typical mean spatial resolutions of ~200km. At these resolutions, health impacts are likely underestimated at urban and semi-urban scales."

We also mention this later on in the manuscript in the Discussion and Conclusions section (Section 4), but have modified the sentence on Page 20482, line 28 to strengthen this.
"*Further simulations using higher resolution models and emission inventories will be required to accurately simulate $PM_{2.5}$ concentrations in urban and semi-urban areas and associated health effects using more recent CRFs that relate RR of disease to changes in $PM_{2.5}$ over a large range of concentration exposures (Burnett et al., 2014).*"

4. Methods
   What is meant by 'commercial sector'? Are solid fuels also used in the commercial sector?

We have modified the sentence on page 20458, line 6.
"We isolate the impact of residential fuel combustion through simulations where we switch off emissions from the "residential and commercial" sector.

We have added two additional sentences on page 20458, line 8 to address the referee's above comment.
"The term "residential" includes emissions from household activities, while "commercial" refers to emissions from commercial businesses activities (excluding agricultural and industrial activities). Both residential and commercial activities use similar fuels for similar purposes, but because emissions are dominated by residential activities, we refer to the "residential and commercial" sector collectively as the "residential sector."

5. Section 2.3: In-situ measurements
   What is the basis for the highlighted regions in Figure 2. Africa, Russian Federation and Southeast Asia in particular have huge and diverse regions without measurements. How do measurements in the country of South Africa represent the Congo or the Sahara regions?

We have modified the original manuscript to explain the distinction between the measurement locations and the coloured geographical regions more clearly.

We have added an additional sentence on page 20460, line 11

“Note that the coloured geographical regions also in Fig. 2 are only used to distinguish differences in mortality across different regions in Section 3.3.”

We have modified the sentence on page 20460, line 19
“For measurement sites located in Eastern Europe, we used BC and OC mass concentrations from Czech Republic and Slovenia (Table 2).”

We have modified the sentence on page 20460, line 20
“For measurement sites located in South Africa, we used PM2.5 and BC mass and aerosol number size distribution (Vakkari et al., 2013).”

We have modified the sentence on page 20460, line 24
“For measurement sites located in South Asia, we also used PM2.5, EC and OC mass, and aerosol number size distribution from the island of Hanimaadhoo in the Maldives (Stone et al., 2007), and EC and OC measurements from Godavari in Nepal (Stone et al., 2010).”

We have modified the sentence on page 20460, line 26
“For measurement sites located in East Asia, we used EC and OC mass data compiled by Fu et al. (2012) for 2 background (Qu et al., 2008) and 7 rural sites (Zhang et al., 2008;Han et al., 2008) in China, measurements from Gosan, South Korea were taken from Stone et al. (2011).”

We have added an additional sentence on page 20507, Figure 2 caption.
“Note that geographical regions are only used to distinguish differences in mortality across different regions (see Section 3.3).”

We have replaced Table 2 first column title on page 20502.
“Region and measurement location name”

We have also replaced Figure 3 on page 20508, so that “Africa” is replaced with “SAfrica” (e.g., South Africa).

We have made a modification to the sentence on page 20466, line 9.
“In contrast, over Eastern Europe the model is unbiased against BC (NMBF= 0.01) but underestimates OC (NMBF= -2.63).”

We have made a modification to the sentence on page 20467, line 17.

“Model simulations where residential emissions have been switched off show that residential combustion contributes about two thirds of simulated BC and OC at these locations.”

We have made a modification to the sentence on page 20468, line 20.
“Figure 6 compares simulated and observed aerosol at South African and Eastern European locations.”

6. What is assumed about C14 for RSF (how to distinguish coal vs. modern carbon?)? Section 2.4:

We explain our assumptions about 14C relating to residential emissions in Section 3.2, but this could be made earlier in the manuscript. We have modified our original manuscript and mention our assumptions earlier.

We have added an additional sentence on page 20461, line 14.
“As previously mentioned, residential emissions consist of a mixture of both fossil and non-fossil sources, with a greater proportion coming from the former. To make distinctions on the fossil vs. non-fossil fraction of residential BC emissions, we make assumptions based on information from other emission inventories and models over the South Asia region (see Sect. 3.2 for more details).”

We have also modified the sentence on page 20472, line 18
“To estimate non-fossil values from the model, we assume that 90% of residential BC transported to Hanimaadhoo originates from residential biofuel sources (consistent with ≥ 90% estimates from the GAINS model), while the remaining non-fossil BC originates from open biomass burning (including agricultural waste and open waste/rubbish burning).”

We have also spotted a mistake in Section 3.2 comparing our model analysis with 14C observations. On page 20472 and in Figure 8 (and caption), we mention that Sheesley et al. (2012) used observations of optically derived BC. This is incorrect as Sheesley et al. (2012) used observations of thermo-optically derived EC. We have modified the original manuscript accordingly.

We have replaced Figure 8 and caption on page 20513, so that “She_BC” is replaced with “She_EC”. We have modified the manuscript to reflect this change.

7. Briefly mention why using population over 30.

Mortality is only calculated for persons over the age of 30 years because this fraction of the population is more susceptible to cardiopulmonary disease and lung cancer . We have changed the original manuscript to explain this.

We have included an additional sentence on page 20462, line 22
"We only calculate premature mortality for persons over the age of 30 years because this fraction of the population is more susceptible to cardiopulmonary disease and lung cancer. "

8. Volume weighting (homogeneous internal mixture) will lead to a more positive effect (and is unrealistic since it would require the BC to spread itself through the scattering material). This should be stated and discussed as a limitation, and please mention the wide range of DRE uncertainty from biofuel due to optical mixing assumptions as shown in Kodros et al. (2015).

We thank the reviewer for this comment. Our use of volume weighting (e.g., homogenous internal mixture) for BC will indeed lead to greater BC positive effect. We note that this, and the fact that we not do explore other optical mixing states for residential emissions, is a limitation of our study. We have changed the original manuscript to specifically state this, as well as to reflect on its limitation.

We have included a new sentence on page 20463, line 18.
"The above assumption that BC is internally or homogeneously mixed with scattering species is unrealistic, providing an upper bound for DRE(Jacobson, 2001;Kodros et al., 2015)."

We have removed the sentence on page 20483, line 29 and replaced it with the following three sentences:
"Furthermore, our DRE analysis is limited because we do not explore the full range of optical mixing states for residential emissions. We assume that BC is mixed homogeneously with scattering species, which provides an upper limit for BC DRE compared to other optical mixing states (Jacobson, 2001). A full investigation of the different optical mixing states commonly used in global models such as in Kodros et al. (2015) would yield a better understanding of DRE from residential emissions."

9. Equation 4 – units don't work out. Did you mean to the 1/3 power not 1 2 ? Typo?

We thank the reviewer for spotting this typo. We have corrected this in the original manuscript.

10. Section 2.6: Model Simulations
    The use of 'emission ratio' does not seem to be the best descriptor here. The total mass of emissions are also changing in the 'emission ratio' simulations (as opposed to holding the mass of emissions fixed while changing the ratio).

The reviewer is correct in that the description for res_BC×2 and res_POM×2 simulations is misleading. We have made altered the original manuscript to clarify this.

We have made a modified the sentence on page 20465, line 18
"We also perform experiments where only residential BC and OC emissions are doubled separately relative to the baseline simulation (res_BC×2 and res_POM×2) to explore uncertainties in both emission mass flux and emission ratio."

We have made a modified the sentence on page 20483, line 6
"The simulated global mean DRE is sensitive to the ratio of BC, POM and SO2 in emissions."

11. Please put the assumptions about the emitted size distribution in the small and large simulations in the main text. It took me a bit to realize they were in the footnotes of Table 3 (it said this when Table 3 was introduced but not when it was talked about in depth in Section 2.6).

We have modified the original manuscript and placed our assumptions about emitted particle size distributions in the main text.

We have removed three sentences on page 20465, line 7 and replaced them with the following three sentences:
"For the majority of our simulations, we use D and σ recommended by Stier et al. (2005) (D = 150nm, σ = 1.59). To account for the uncertainty in the size of emitted residential carbonaceous combustion aerosol and uncertainty of sub-grid ageing of the size distribution, we conduct simulations spanning the range of observed size distributions for primary BC and OC residential combustion particles, while keeping emission mass fixed. We use AeroCom (Dentener et al., 2006) recommended

particle size settings (res_aero) (D = 80nm, σ = 1.8), and following a similar approach to Bauer et al. (2010), we use the range identified by Bond et al. (2006) for lower (res_small) (D = 20nm, σ = 1.8) and upper (res_large) (D = 500nm, σ = 1.8) estimates."

We have also modified footnote b on Table 3
"AeroCom (Dentener et al., 2006) recommended residential (biomass/biofuel) primary carbonaceous particle sizes, D = 80nm σ = 1.8."

12.3.1 Model Evaluation

Figure 3: What does each datapoint correspond to? Are these each sites at multiple times or just averaged over all times available? Were the times of the model co-sampled with the times of the measurements or a comparison with the overall averaged values of the model with the overall average values from the measurements (that may have been for different times)? Are you averaging over the entire regions defined in Figure 2 or using the grid box of the site? Also would be good to include the number of datapoints.

We thank the reviewer for these comments. In Figure 3 a-c, each data point corresponds to an observed and corresponding simulated monthly mean at each measurement location as depicted in Table 2. Simulated meanly means where taken from the model during the same period the measurements were collected and from the same model grid box (e,g., weights the relevant gridbox value to account for the relative location of the measured observation location) corresponding to the same latitude, longitude, and altitude for each measurement location as defined in Table 2. In Figure 3 d, we have calculated the NMBF for each of the three simulations. We have modified the original manuscript to make this clearer.

We have replaced the Figure 3 caption on page 20508.
"Observed and simulated monthly mean BC ***(a)****, OC* ***(b)*** *and $PM_{2.5}$* ***(c)*** *concentrations for the baseline simulation (res_base) using ACCMIP emissions at each measurement location depicted in Table 2, and normalised mean bias factor (NMBF) for each region defined in Table 2.* ***(d)*** *NMBF where square shows the baseline simulation, bottom error bar shows the range for removed residential emissions (res_base_off) and top error bar shows residential carbonaceous emissions doubled (res_×2) for each region defined in Table 2. Colours represent observed, simulated and NMBF for measurement location regions defined in Table 2: all measurement locations (All: black), South Asian locations (SAsia: blue), East Asian locations (EAsia: green), Eastern European locations (EEurope: red) and South African locations (SAfrica: orange)."*

13. Figures 4-6: I can’t read the yellow writing in the legend. The ’res_off’ simulations are not included. It would be beneficial to see how including res emissions changes comparisons

We thank the reviewer for the spotting that the yellow writing in the legend is difficult to read.

We have replaced Figs 4-6 on pages 20509, 20510 and 20511 and have used a different colour

Figs 4-6 do include the simulations where residential emissions have been removed; please see Figs 4-6 captions: “*Experiments where residential emissions have been removed are represented by the blue (res_base_off) and green (res_monthly_off) dotted lines.”* This does give the reader an idea of the model comparison to observations without residential emissions. Figure 3 d also highlights NMBF when residential emissions have been removed, switched on and doubled. This is discussed further in Sect. 3.1 page 20466.

14. Figures 4-6 are used to suggest smaller res emissions are unrealistic. It would be good to explicitly acknowledge that other errors in the model limit the ability determine this for sure.

We have also included an additional sentence on Page 20480, line 14
“Uncertainty in aerosol removal processes and transport, and missing anthropogenic SOA and nitrate formation may all contribute to underestimation of aerosol mass.”

We have also included an additional sentence on Page 20480, line 14
“Nevertheless, previous modelling studies have also suggested that residential emission datasets underestimate emissions….”

15. Figures 4-6: It would be helpful to explain why the number of simulations differs between plots (I assume because some simulations have little effect on the masses, only number).

The reviewer is correct. Figs 4 and 6 comparisons to number concentrations contain a greater number of model simulations because these simulations (experiments where particle size and nucleation scheme has been perturbed) have little effect of aerosol mass, but will have more of an effect on number concentrations. We have modified the original manuscript to explain this to the reader.

We have added an additional sentence to Figure 4 caption on page 20509.
"Note that additional experiments (res_BHN, res_aero, res_small and res_large) are included in k-l because these experiments have little impact on aerosol mass (a-j)."

We have added an additional sentence to Figure 6 captions on page 20511.
"Note that additional experiments (res_BHN, res_aero, res_small and res_large) are included in a-f because these experiments have little impact on aerosol mass (g-j)."

16. Section 3.2: PM Changes
    When discussing which species contribute the largest change to PM2.5, absolute changes in BC, POM, and SO4 mass are given. Is this the mass for the species with Dp less than 2.5 microns? This should probably be stated.

We have modified the Figure 7 caption on page 20512 to state this.
"Percentage contribution of residential emissions to annual surface mean $PM_{2.5}$ concentrations ***(a)****, BC* ***(b)****, POM* ***(c)*** *and sulfate (SO4)* ***(d)*** *concentrations (in size fraction $PM_{2.5}$) for the baseline simulation (res_base), relative to an equivalent simulation where residential emissions have been removed (res_base_off)."*

17. Uncertainties in the assumed modern/fossil carbon ratio of residential burning. How much does this impact the comparison?

We agree with the reviewer that our assumption about non-fossil/fossil fraction of residential BC at the Indian Ocean location depicted in Figure 8 is uncertain. Here, we assume that ≥90% of residential BC at that location is from non-fossil (i.e. biomass sources) during the time period the 14C observations were collected. The ≥90% estimate comes from an analysis using the GAINS model, which provides estimates for the non-fossil/fossil fraction of fuel used in the residential sector for India. During the time period that the observations were collected (e.g., pre and post monsoon seasons) the Indian Ocean location in question experiences aerosol transport from the Indian sub-continent (Gustafsson et al., 2009;Sheesley et al., 2012;Bosch et al., 2014) (simulated by our model too), so applying this faction is a reasonable assumption, although we acknowledge there is some uncertainty involved. Nevertheless, the spread in the observed non-fossil EC contribution (46-

73%) is large enough to make it difficult to constrain the residential BC contribution, even in light of uncertainties associated with the ≥90% estimate of residential BC being non-fossil.

18. Should discuss that running at higher resolution would likely lead to a higher number of deaths since emissions likely correlate with population density (so coarser grids smear this effect).

We thank the reviewer for this comment – this is an important point. We have changed the original manuscript to highlight this point more roughly in the Discussion and conclusions section (Sect. 4).

We have included an additional sentence on page 20482, line 28
"We also note that the coarse resolution of our global model likely provides a conservative estimate of premature mortality due to residential emissions because it cannot simulate higher concentrations associated with highly populated urban and semi-urban areas."

19. It is stated that the health results are most sensitive to changes in emitted POM mass. But is this just because the POM is the largest emitted species, so doubling this causes the greatest change PM change? The C-R response function is not determined by species. Or on the other hand, does this have something to do with spatial OC:BC ratio (perhaps caused by fuel type correlated with population)? This is not discussed.

The reviewer is correct. Health impacts are most sensitive to changes in POM because POM dominates the mass of residential emissions as stated on page 20474, line 3:

"Factorial simulations where residential emissions of POM, BC and SO_2 are increased individually shows that health effects are most sensitive to uncertainty in POM emissions which dominates the total emission mass."

However we have modified this sentence to make this clearer.
"The CRF function treats all aerosol components as equally harmful, so simulations where residential emissions of POM, BC and SO_2 are increased individually shows that health effects are most sensitive to uncertainty in POM emissions because this component dominates the total emission mass."

20. Figure 9 should really say res_base - res_off, right? Similar comment for Figure 10 (the “off” simulations are required as a health baseline).

The reviewer is absolutely right here. We have changed the original manuscript to reflect this.

We have modified Figure 9 caption on page 201514.
“Simulated annual premature mortality (cardiopulmonary diseases and lung cancer) due to ambient exposure to ambient $PM_{2.5}$ from residential emissions (res_base - res_off).”

We have modified Figure 10 caption on page 201514
“Simulated global annual premature mortality (cardiopulmonary diseases and lung cancer for persons over the age of 30 years) due to exposure to ambient $PM_{2.5}$ from residential emissions, relative to an equivalent simulation where residential emissions have been removed. Results are shown for standard emissions (res_base and res_monthly) and where residential emissions have been doubled (res_×2 and res_monthly_×2). Mortality is shown for Eastern Europe and Russian Federation (EEurope), Africa (Africa), South Asia (SAsia), South East Asia (SEAsia), East Asia (EAsia) and rest of the world (as defined by the coloured regions in Fig. 2).”

21. “To our knowledge, this is the first study of the global excess mortality due to ambient PM2.5 from residential cooking and heating emissions. A recent study by Chafe et al. (2014) concluded that ambient PM2.5 from RSF cooking emissions resulted in 420 000 annual excess deaths in 2005 and 370 000 annual excess deaths in 2010.” Please stress that Chafe removed heating, or maybe add “both” to the first sentence. It took me reading these sentences a couple times to realize how they were not contradictory.

We thank the reviewer for this comment. We have changed the original manuscript to make this clearer.

We have modified the sentence on page 20474, line 22.
“To our knowledge, this is the first study of the global excess mortality due to ambient PM2.5 from both residential cooking and heating emissions.”

22. Section 3.4: Number
    Can you include brief comparisons to Kodros et al. (2015) to this and the following sections when comparisons are relevant?

Yes, we can certainly provide brief comparisons to (Kodros et al., 2015), however direct comparison is problematic because ‘residential’ emissions are not the same as ‘biofuel’ emissions.

We have modified the sentence on page 20475, line 12.
“This reduction is caused by primary particles acting as a coagulation sink for nucleated particles and a condensation sink for nucleating and condensing vapours, suppressing new particle formation (Spracklen et al., 2006), which is broadly consistent with the findings of Kodros et al. (2015) for particle number concentrations due to the effect of biofuel emissions.”

We have modified the sentence on page 20478, line 17.
“These estimates differ somewhat to Kodros et al. (2015) that found a homogenous optical mixing state produced a positive DRE of +15 mWm^{-2} for biofuel emissions, however, because residential emissions differ to biofuel emissions, comparisons become problematic. We therefore, assume differences in radiative effect compared to Kodros et al. (2015) are likely dominated by differences in the emissions used and differences in the optical calculation.”

23. Why say ’CCN’ instead of N50? It’s probably more precise to just call it N50 and say in the text that this is a proxy for CCN. Again changes to N50 most sensitive to changes in POM. That is just a mass thing though right? This has nothing inherently to do with OM vs BC other than the emissions mass of OM is higher, right?

We agree with the reviewer that it make more sense to use N50 instead of CCN. We have changed the original manuscript accordingly. The reviewer is also correct that changes in N50 are most sensitive to POM because POM consist of most of the mass.

For section 3.4 (pages 20475-20476), we have change the text so that N_{50} is used instead of CCN.

We have also replaced Figure 12 and caption so that N_{50} is used instead of CCN.

24. Figure 12 colorscheme. . . Blues for both positive and negative numbers. . . this is very misleading. Please make all blues negative and red positive (even

if it means the plot has very little of one color). This will allow the reader to instantly recognize which regions have increases vs. decreases.

25. Section 3.5 - Figure 13: Same comment as Figure 12.

We agree with the reviewer that colour scheme is both Figure 12 and 13 is misleading. We have changed the original manuscript and used a different colour scheme making blues negative and red positive for both Figure’s 12 (see above) and 13.

26. Discussion and Conclusions
    Should reiterate that running at higher resolution would likely lead to a higher number of deaths since emissions likely correlate with population density (so coarser grids smear this effect).

We have changed the original manuscript to address this comment sufficiently. Please refer to comment 18 and the additions and modifications made on page 20482, line 28.

27. “Furthermore, BC particles coated in a non-absorbing shell produce stronger absorption than the BC core alone (Jacobson, 2001), which we do not treat here.” Note, that coating BC yields more absorption than assuming BC and scattering species are *externally mixed*. However, you treat all species as being volume internally mixed (the BC is homogeneously mixed throughout the particle), which give *more* absorption than core-shell. This is discussed in the Jacobson, 2001 article.

We have changed the original manuscript to address this comment sufficiently. Please refer to comment 8 and the additions and modifications made on page 20483, line 29.

Response to anonymous Referee #2

1. Abstract:
   Please reiterate that the results are presented for the year 2000. As the authors mentioned in the conclusion, the use of solid fuels changes rapidly with time because of the population growth and technology innovation, especially in the developing countries.

   Please also specify that the reported excess mortality is only for the cardiopulmonary diseases and lung cancer, not all-cause mortality.

We thank the reviewer for these comments. We have changed the original manuscript according.

We have modified the sentence on page 20451, line 6.
"We use a global aerosol microphysics model to simulate the uncertainties in the impact of residential fuel combustion on atmospheric aerosol for the year 2000."

We have modified the sentence on page 20451, line 13.
"We use a concentration response function to estimate the health impact due to long-term exposure to ambient $PM_{2.5}$ *from residential emissions for the year 2000."*

We have modified the sentence on page 20485, line 4.
"We have reported human health and climate impacts for year 2000, but in China, emissions from the residential sector have increased 34% during the period 2000–2012 due to the growth of coal consumption (Cui et al., 2015)."

We have modified the sentence on page 20451, line 14
"We estimate global annual excess adult (> 30 years of age) premature mortality (due to both cardiopulmonary disease and lung cancer) of 308 000 (113 300–497 000, 5th *to 95*th *percentile uncertainty range) for monthly varying residential emissions and 517 000 (192 000–827 000) when residential carbonaceous emissions are doubled."*

2. Introduction, paragraph 2:

Besides BC, gas-phase SO2 and primary OC, does the combustion of biofuels also emit volatile/semi-volatile organic compounds that can produce secondary organic aerosols (SOA) via atmospheric oxidation? The missing SOA mechanisms in the model may in part explain the gap between the simulation and the measurements. It would be good to acknowledge this limitation.

The reviewer is correct that that combustion of residential fuels does emitted volatile organic compounds, and that the non-treatment of this process in our model may an impact on model evaluation. We have changed the original manuscript accordingly to highlight this limitation.

We have included an additional sentence on page 20452, line 23
"The combustion of residential fuels also emits volatile and semi-volatile organic compounds that can lead to the production of secondary organic aerosols via atmospheric oxidation, however we do not treat this process in the present study."

We have modified the sentence on page 20479, line 27.
"The model typically had a larger underestimation of OC compared to BC concentrations, possibly due to uncertainty in emission factors or potentially due to an underestimation of anthropogenic SOA (Spracklen et al., 2011b)."

We have also included an additional sentence on Page 20480, line 14
"Uncertainty in aerosol removal processes and transport, and missing anthropogenic SOA and nitrate formation may all contribute to underestimation of aerosol mass."

3. In the calculations of DRE, the authors used the volume-weighted mean of refractive indices for each log-normal mode. It is not quite clear to me what is the mixing state of black carbon assumed in these calculations. Are the POM and BC emitted as an internal mixture? Are the hydrophilic modes and hydrophobic modes externally mixed in the optical calculations? Please clarify.

In our study we assume that BC within individual modes is internally or homogeneously mixed with scattering species, but because we calculate modes separately hydrophilic and hydrophobic modes are externally mixed in the optical calculation. We have changed the original manuscript to clarify this.

We have modified the sentence on page 20463, line 12.
"A refractive index is calculated for each individual mode separately, as the volume-weighted mean of the refractive indices for the individual components (including water) present (given at 550nm in Table A1 of Bellouin et al. (2011))."

We have included a new sentence on page 20463, line 18.

"The above assumption that BC is internally or homogeneously mixed with scattering species is unrealistic, but this assumed optical mixing state does provide an upper bound for DRE (Jacobson, 2001;Kodros et al., 2015)."

4. In addition, POM emitted from the residential combustions is assumed to be nonabsorbing. This assumption seems to be unrealistic, although the authors have discussed this limitation in the paper. I would recommend adding one more simulation in the revised paper, using a small but non-zero value of imaginary refractive index for the POM to test the sensitivity of DRE to brown carbon.

The reviewer is correct that we highlight the limitation of not including positive DRE induced by absorbing POM in our study. Unfortunately, due the way we calculate DRE, it makes it very difficult us to calculate the effect of absorbing POM from our model simulations. This important point needs to be left for future work.

5. For the benefit of the readers, it would be good to specify how the NMBF values were calculated.

We agree with the reviewer that this will be helpful for the readier. We have changed the original manuscript to include the NMBF equation.

We have modified the sentence on page 20466, line 1.

"*Figure 3 compares observed and simulated monthly mean BC, OC and PM2.5 concentrations, and normalised mean bias factor (NMBF) (Yu et al., 2006) where M_i are the simulated concentrations by the model and O_i are the observed concentrations at each measurement location, i.*

$$NMBF = \frac{\sum(Mi - Oi)}{\sum Oi} \quad if \ \bar{M} \geq \bar{O}, and \quad NMBF = \frac{\sum(Mi - Oi)}{\sum Mi} \quad if \ \bar{M} < \bar{O}$$

"

6. How does the excess mortality due to residential emissions compare to the baseline mortality? For example, what is percentage increase compared to the excess mortality of the total PM2.5?

We agree with the reviewer that it would be good to calculate baseline mortality in order to calculate fraction of total mortality directly responsible from residential emissions. It is difficult however to do this using the function we use because it would mean we would have to make assumptions about the counterfactual concentrations below which no increase in relative risk is seen. Instead, we compare our mortality estimate from the residential sector against other estimates of excess mortality from all emissions (see Sect. 3.3 and Sect. 4)

7. The health calculation assumes that the effect is identical for different PM species. Some epidemiological studies show that this is not the case. This should be discussed.

The reviewer is correct that the concentration response function that we use treats all PM species as equally harmful, which may not be realistic. We have included a short discussion to our study.

We have included an additional sentence on page 20483, line 2.
"In addition, exposure response functions, such as the one used in this study, treat all PM species as equally toxic, however carbonaceous aerosol, which make up a large fraction of residential emissions, may be more toxic compared to inorganic or crustal PM (Tuomisto et al., 2008). New exposure response functions will therefore need to account for the different toxicity of chemical components present in atmospheric aerosols."

8. Technical corrections

Thanks for spotting these corrections. They have been corrected:

Page 20465 line 4: re_base_off -> res_base_off

This has been done.

Page 20470 line 25: Define "NH" in the acronym table

This has been done.

Page 20471 line 6: Wang et al. (2015b) is not provided in the reference list.

This has been placed in the reference list.

Page 20504 Table 3: “%” are missing in several places.

This has been done.

Table 3, footnote d: Lower -> upper

This has been done.

Figure 2 caption: Southeast Aisa -> Southeast Asia

This has been done.

References

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