Response to review comments

acpd-15-20881-2015: Sensitivities of UK $PM_{2.5}$ concentrations to emissions reductions
Vieno et al.

We thank the reviewers for their time spent reviewing our manuscript and for highlighting the interesting and useful findings we report. Below we respond in turn to the additional individual comments made by the reviewers and indicate the revisions we have made to our paper. The original comments from the referees are in plain text and our responses are in *italic*.

Anonymous Referee #1

General: This study focuses on changes in $PM_{2.5}$ from emissions changes, but climate change itself may have an effect. Granted, the latter effect is usually found to be smaller, but it is probably worth mentioning the relevant lit on the topic

The focus of this work is an assessment of sensitivity of UK $PM_{2.5}$ surface concentrations to reductions of UK terrestrial emissions of relevant components/precursors. These reductions are with respect to a 'current' baseline and to a time-horizon relevant to developing and implementing additional policy on precursor emissions beyond those already in a 'current legislation' pipeline. 2030 was chosen as being not too soon for additional policy change to be implemented, but not too far that other factors might become overly influential (for example significant changes in global concentrations of methane, land-use change or climate). We agree that climate change may also have some effect on $PM_{2.5}$, but so might other factors (examples given above) which are not explicitly considered in this sensitivity study. In addition, as the reviewer notes, other work has shown that for the 2030 time horizon pollutant concentrations will be more strongly influenced by changes in pollutant emissions than by changes in climate (some citations given in the additional text below).

In response to the reviewer's comment, we have now added the following text to the revised paper: "It is recognised that climate change may also have some influence on future $PM_{2.5}$ concentrations in the UK; however the focus is here on UK precursor emission sensitivity and many studies have concluded that on the 2030 timescale air pollutant concentrations will be much more strongly influenced by changes in precursor emissions than by changes in climate (e.g. Langner et al. (2012);Colette et al. (2013);Coleman et al. (2013))."

20885.13: Please explain more the details of how observations are used to derive boundary conditions.
The import of $PM_{2.5}$ and its precursors to the UK from continental Europe is larger than for outside the European domain. The cross Atlantic transport of SIA has a small effect on the EU surface concentrations of SIA (as well as on deposition) as has been demonstrated by Simpson et al. (2012) and Sanderson et al. (2008). However, as European emissions are further reduced the relative cross Atlantic transport may play a larger role in the future

We have added the following text to the manuscript:
"The EMEP4UK model uses a nested approach, the European domain concentrations are used as boundary condition for the UK domain. The boundary condition at the edge of the European domain are prescribed concentrations in terms of latitude and adjusted for each year. For ozone, 3-D fields for the whole domain are specified from climatological ozone-sonde data-sets, modified monthly against clean-air surface observations as described in Simpson et al. (2012)"
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20885.20: What is the potential limitation? Can it be briefly described? At present this statement is too vague to be informative

The following text is now included in the revised paper in place of the original sentence that read "The potential limitation of the EMEP4UK approach is discussed in (Vieno et al., 2014)"

"Whilst fine nitrate production is modelled using a thermodynamic model (MARS), the formation of coarse nitrate from nitric acid uses a parameterised approach that seeks to capture the HNO_3 reaction with sea salt and crustal material. The conversion rate of HNO_3 to coarse nitrate depends on relative humidity, as described by Simpson et al. (2012), but is not explicitly linked to the surface area of the existing coarse aerosol. Both nitrate generation mechanisms compete for the same HNO_3, and whilst this constrains the total amount of nitrate produced, it is acknowledged that the resulting split into fine and coarse nitrate is somewhat uncertain as discussed in Aas et al. (2012). A more explicit aerosol scheme is under development for the model."

Moreover, we apologise that there was an error in the original text in that the version of EMEP4UK used in this study used the MARS rather than the EQSAM aerosol scheme. We have now corrected the text and amended the associated reference.

20887.1: How realistic is it to assume the spatial distribution is fixed? How might this bias the results of this work?

We agree that the spatial distribution of emissions is likely to change with time; for example, point source emissions can change between years (e.g. power plant closing or new build). Land-use changes may also affect ammonia emissions. However, these changes are not easily predicted for a future scenario and it might be anticipated that changes in spatial patterns of emissions will likely be smaller than the changes in absolute amounts of emissions. We have added the following sentence to acknowledge this point in the text:
"Whilst there will likely be some changes in the spatial distribution of emissions, such changes are not easily predicted for a future scenario, and are anticipated to be smaller than the changes in absolute amounts of emissions."

In addition, the focus of this study is on quantifying the contribution of different reduction options, so while changes in distribution patterns will affect comparisons between modelled and observed surface concentrations in the future, the quantification of the relative effect of emission reductions as a UK average should be relatively less affected.

20887.5: As mentioned above, some discussion of how large such impacts might be would be worth including

We already mention in the Discussion that inter-annual variability in meteorology will mean that the sensitivities of UK $PM_{2.5}$ to additional reductions in a variety of precursor emissions will also have some inter-annual variability. However, as emphasised above and in the paper, the focus of this work was on understanding the effect of these emissions sensitivities which was why meteorology was deliberately kept constant to isolate these. Whilst it is possible that climate change by 2030 may also change the nature of the UK meteorology (to a small extent), as indicated in one of our responses above, any such impact on this time horizon is likely to be much smaller than the impact of precursor emissions changes.

20887.23: Can the authors be more rigorous here and provide statistical analysis such as correlation coefficients, bias and error.

We have now added the linear regression between observation and model at the top of each panel, along with the correlation coefficient, bias, and mean square error, and also a summary of the comparison reported in Vieno et al. (2014). The four sites included in Vieno et al. 2014 showed good agreement between EMEP4UK simulation and the observed NO_3^- and SO_4^{2-} as shown in the Table 1 of this paper. This is reproduced below for these responses but not in the main paper since this provides the citation to the original source.

*Table 1: Mean concentrations, and correlation and regression statistics, for monthly-averaged modelled and measured NO_3^- and SO_4^{2-} in particulate matter for the period 2001-2010 at four sites of the AGANet network: Strathvaich Dam (north-west Scotland), Bush 1 (central Scotland), Rothamsted (south-east England), and Yarner Wood (south-west England). The comparison is based on a linear fit where measurement = slope * model + intercept.*

	Particulate NO_3^-			
	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
Measurement mean	0.49 µg m^{-3}	1.37 µg m^{-3}	3.35 µg m^{-3}	1.98 µg m^{-3}
Model mean	0.77 µg m^{-3}	1.42 µg m^{-3}	2.73 µg m^{-3}	2.23 µg m^{-3}
R	0.49	0.91	0.81	0.86
Slope	0.59	0.96	0.68	0.95
Intercept	0.48 µg m^{-3}	0.10 µg m^{-3}	0.44 µg m^{-3}	0.34 µg m^{-3}
	Particulate SO_4^{2-}			
	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
Measurement mean	0.57 µg m^{-3}	0.94 µg m^{-3}	1.75 µg m^{-3}	1.20 µg m^{-3}
Model mean	0.61 µg m^{-3}	0.95 µg m^{-3}	1.48 µg m^{-3}	1.28 µg m^{-3}
r	0.72	0.79	0.65	0.69
Slope	0.86	0.76	0.56	0.65
Intercept	0.12 µg m^{-3}	0.24 µg m^{-3}	0.50 µg m^{-3}	0.36 µg m^{-3}

20887.24: Can the authors summarize the species specific evaluation of Conolloy 2011? Does the model do better at estimating concentrations of any particular component of SIA? What were the biases, quantitatively?

The citation to Conolly et al. (2011) is for the description of the monitoring network; the model-observation evaluations against the monitor data were presented in Vieno et al. (2014), but as requested in the previous comment we have now added the essential aspects of the evaluation data to this paper too. We have also added a reference to another study in which the EMEP4UK model was evaluated against observations and other model (Carslaw, 2011a, b).

20881.1: This seems rather unsubstantiated. How did the authors rule out the role of SOA? Why was particle-bound water not included in the model PM2.5 calculations (it is easily done using hydroscopic growth curves â˘A˘T textbook undergraduate level calculation)?

We apologise in not being explicit in the model description section in stating that the EMEP4UK model does include both primary organic aerosols and secondary organic aerosols. We have now added the following description of this to the revised paper:

"In the model version used here, $PM_{2.5}$ is the sum of the fine ($PM_{2.5}$) fraction of: ammonium (NH_4^+), sulphate (SO_4^{2-}), nitrate (NO_3^-), elemental carbon (EC), organic matter (OM), sea salt (SS), mineral dust, and 27% of the coarse nitrate. PM_{10} is the sum of $PM_{2.5}$ plus the coarse ($PM_{2.5-10}$) fraction of EC, OM, NO_3^-, SS, and dust."
The reason why the paper does not discuss SOA is because the impact of reductions in emissions of UK VOC has very little impact (via formation of anthropogenic SOA) on the UK $PM_{2.5}$ as shown in Figures 4e (or 2010 scenario) and 8e (for the 2030 scenario), as compared with the impact on $PM_{2.5}$ for emissions reductions in primary $PM_{2.5}$, NH_3, SO_2 and NO_x. (Note that the impact of primary $PM_{2.5}$ emissions reductions on UK $PM_{2.5}$ will include the contribution from reductions in primary organic aerosol.)

The issue of whether or not to include an estimate of particle-bound water is not straightforward. Different measurement techniques and conditions will incorporate different proportions of the ambient $PM_{2.5}$ water content. Because of this uncertainty in what measurements measure (against which legislation for PM is based), we focus here on changes to the dry mass of surface $PM_{2.5}$ derived from changes in the emissions of primary $PM_{2.5}$ and in secondary $PM_{2.5}$ precursor gases. However, we acknowledge that changes in mass of secondary inorganic components will be accompanied by changes in mass of particle-bound water and now incorporate caveats to this effect in the both the Results and Discussion section .

20888.2: What evidence do the authors provide that such missing mechanism don't affect the sensitivities calculated here? It's not entirely implausible. For example, if they have neglected uptake of HNO_3 on dust, then they are overestimating their response of nitrate to changes in NO_x emissions…Or if they considered the role of NO_x on SOA, which can be quite significant.

The RH-dependent coarse nitrate formation parameterises the effect of HNO_3 onto mineral dust, however the details are not reproduced mechanistically (see response 20885.20). A new version of the EMEP4UK model is currently under development that will explicitly calculate this but is not available at this time. We confirm again, however, that the model does represent the changes in SOA due to emissions reduction in NO_x (and other relevant chemistry). Description stating that SOA is included in the model has been added to the Methods section.

20888.8: Likewise, some overestimation would occur for the background site. This affect may somewhat cancel the low-biases discussed above.

For rural background sites without significant emissions the grid average should be representative, rather than overestimated. However, the two rural background AURN sites which have enough data to compare the monthly values are Harwell and Auchencorth. At this two sites the bias is -1.1 and 2.5 ($\mu g\ m^{-3}$) for Harwell and Auchencorth Moss, respectively.

20888.10 & 26: It seems like one additional model run with boundary conditions set to zero could easily be performed to quantify this aspect more completely.

This has undertaken and presented previously in an report of the Air Quality Expert Group (AQEG, 2015) The analysis showed that UK emissions contribute around 50-55% of total annual average $PM_{2.5}$ in the UK. We add the following text and reference in the manuscript:
"An analysis presented in AQEG (2015) also using the EMEP4UK model showed that UK emissions contribute around 55% of the total $PM_{2.5}$ in the UK. This limits the extent to which long-term average concentrations can be reduced by UK action alone"

20889.13: This results is “key” to what, exactly? Also, why does this occur, from a standpoint of atmospheric chemistry and aerosol partitioning? If the authors wish to draw attention to this finding, it should be better explained.

The word ‘key’ here has now been replaced by ‘important’. The result to which this comment refers is the observation of different spatial patterns for reductions in $PM_{2.5}$ across the UK for different precursor reductions. Rationalisation for these different patterns is discussed at a number of places in the Results and Discussions sections (e.g. the different locations of the sources of different precursors and the different timescales, and hence spatial scales, over which chemistry and transport interact).

20889.22: why?

This query is referring to our statement that reductions in $PM_{2.5}$ arising from reductions in SO_x emissions are not generally associated with urban areas. This is because the major sources of SO_x emissions in the UK are large power-plants and large industrial plants (e.g. steelworks) that are not located within urban areas but outside urban areas. The couple of subsequent sentences we already have in the text provide rationalisation, but for further emphasis we have now added the following sentence after the one in the original text: “This is primarily caused by the spatial distribution of major sources of SO_x emissions. As ~80% of UK SO_x 2010 emissions originate from large point sources (power plants, industrial facilities), which are not located in the heart of urban areas, associated emission reductions have the most profound effects in rural areas.”

20890.1: This could have been determined without any sensitivity model experiments by calculating the gas ratio (= available NH3 beyond that required for sulfate neutralization, divided by total inorganic nitrate + nitric acid, Ansari and Pandis, 1998) in the baseline model run. A map of that ratio over the UK would be useful for this work.

We agree that the NH_3 sensitivity of $PM_{2.5}$ formation could have been demonstrated by the method described, but we have demonstrated it as a consequence of the emissions sensitivities runs performed for this work. Moreover, it is not so straightforward since changes in emissions also change the dry/wet deposition ratio which also affects the lifetime and hence concentrations of $PM_{2.5}$ (Vieno et al., 2010).

20892.17: To be fair though, it may also be worth mentioning that NH_3 reductions would as well have additional benefits given their impacts on N deposition and ecosystems.

We entirely agree that NH_3 emissions reductions (Klimont and Winiwarter, 2015), and reductions in other precursors, will have other additional benefits on, for example N (and S) deposition (Adrian et al., 2015) and on ground-level ozone (with its associate human health and ecosystem impacts), aside from the impacts of these emission reductions on $PM_{2.5}$ that are the focus of this work. Our paper already pointed this out in a number of places. For example, right at the outset, at the end of the Introduction we state “It is also recognised that, whilst the focus here is on reduction in concentrations of $PM_{2.5}$ from the perspective of its impact on human health, the reduction of anthropogenic emissions in general will also have other benefits including on human health, on N and S deposition, and on ozone formation”; in the second paragraph of the discussion we write: “It is also recognised that reductions in NO_x and VOC emissions have the potential to deliver health benefits separately from their contribution to reduction in $PM_{2.5}$ through reductions in population exposure to surface NO_2 and O_3”; and at the end of the Discussion we write: “ Measures taken in the UK to reduce concentrations of ambient $PM_{2.5}$ and of precursor gases, both within and outside of areas of population, will have multiple co-benefits on human health, N and S deposition, ozone formation and radiative forcing, not

just in the UK but elsewhere." We therefore think we have adequately emphasised the other benefits of emissions reductions.

MINOR COMMENTS:
20882.18: "but" – what is being contrasted here? Maybe a different word would be better.

The word 'but' has been changed to 'however'.

20882.24: "observation" – conclusion? "observation" implies a measurement

The word 'observation' has been changed to 'conclusion'.

20883.9: should clarify that 3.2 million is exposure to ambient PM2.5 (as opposed to indoor, which is even larger)

The word 'ambient' has now been inserted to read '..to exposure to ambient PM2.5 concentrations prevailing in 2005…'

20884.1: I don't see how the mass concentrations masks composition. This could be re-written to be better, something like "while standards focus on PM2.5 mass concentration, meeting these standards are complicated by the considerable chemical heterogeneity. . ."

We agree the original phrasing was probably a bit difficult to interpret to understand what we meant. The reviewer supplies a good alternative phrasing to the start of this sentence which we have now incorporated.

20884.21: usually write old to new in citations years

Multiple in-text citations are now listed in chronological order of publication.

20885.16: reactions,

A comma has now been inserted after 'reactions' in this sentence.

20888.16: It would be interesting to also see the % reductions.

Throughout this work we focus on the absolute reductions in $PM_{2.5}$ (i.e. $\mu g\ m^{-3}$) derived from the UK emissions reductions simulations because it is the change in absolute amount of $PM_{2.5}$ that drives the change in impact of $PM_{2.5}$ (on health, or deposition, etc.) and because legislation for compliance on $PM_{2.5}$ concentrations is quantified by the amount of $PM_{2.5}$. Our work is an exploration, from the 'policy-maker perspective,' of sensitivities of UK $PM_{2.5}$ reductions to UK emissions reductions and it is the resultant absolute changes in $PM_{2.5}$ that are of relevance to policy-makers (and to the $PM_{2.5}$ impacts).

Reviewer T. Oxley

General Comments: This is a useful paper describing the sensitivity of UK PM2.5 concentrations and therefore health impacts to changes in primary and secondary pre-cursor emissions using the EMEP4UK ACTM.

We thank the reviewer for their supportive comments on the work.

I found that figure 6 not only doesn't add to the manuscript, but that it actually made it more difficult to follow because it took me some time to work out what the map was actually showing me, ie a 30% reduction in NH3 combined with a 30% increase in pPM2.5, which was a bit strange? The point the authors make regarding urban or rural impacts is valid, but I had already understood this from Figure 4.

We believe that Figure 6 is an impotant figure for this paper in that it provides a direct visualisation of the spatial pattern across the UK of localities where $PM_{2.5}$ reductions are most effectively derived through primary $PM_{2.5}$ emissions reductions as compared with through NH_3 emissions reductions (on the basis of applying 30% emissions reductions to one or the other). In this single visualisation, Figure 6 directly shows that for the largest urban areas in the UK, reducing primary $PM_{2.5}$ emissions is more effective at reducing $PM_{2.5}$ than the equivalent percentage emissions reductions in NH_3; whereas outside these urban areas, reducing NH_3 emissions is more effective at reducing $PM_{2.5}$ than the equivalent percentage emissions reduction in primary $PM_{2.5}$.

We think the caption text for this figure is clear in stating how this figure is derived and how it should be interpreted: first, the caption explicitly states that Figure 6a is the data in Figure 4b ($PM_{2.5}$ changes for 30% NH3 emissions reductions) minus the data in Figure 4f ($PM_{2.5}$ changes for 30% primary $PM_{2.5}$ emissions reductions) for the 2010 year, and similarly for Figure 6b for the 2030 future scenario; secondly, the caption also states that blue colours indicate where reductions in $PM_{2.5}$ for 30% reduction in NH3 emissions exceed the reductions in $PM_{2.5}$ for 30% reduction in primary $PM_{2.5}$ emissions, and vice versa for the red colours.

We therefore wish to retain Figure 6. It may be that the short title that was also present on this figure contributed to some misunderstanding in interpretation; we have now removed this title so that the caption alone provides the detail.

Specific Comments:
Is figure 2 really necessary as it is simply a population map which could be downloaded from www.ons.gov.uk

A central finding of our work is the different effects on $PM_{2.5}$ mitigation spatially that derive from UK emissions reductions of different $PM_{2.5}$ precursors, which in turn means that different precursor emissions reductions have different impact on population-weighted $PM_{2.5}$ compared with area-weighted $PM_{2.5}$. Understanding this point is helped by knowledge of the pattern of population density in the UK. Whilst we could just retain the summary data for the effects of the reductions on the UK national population-weighted and area-weighted $PM_{2.5}$ that appear in Table 1 and Figure 5, we believe that since our paper includes a number of maps of model simulations it is helpful for the reader to be able to see directly for themselves the geographical comparison between maps of spatial changes in $PM_{2.5}$ for different emissions reductions sensitivities and maps of the population density of the UK. We think this is particularly important for readers not familiar with the UK urban layout. We note this reviewer is UK-based so will presumably be very familiar with the locations of UK urban centres. This will not be familiar to many other readers.

Whilst we could just refer the reader to a URL, or put the population weighted map in supplementary information, since we are talking about one figure that we believe will be of considerable help to the reader in emphasising our findings we wish to retain this Figure in the main paper.

P20890, L14: I think it would benefit to remove Figure 6 and revise these paragraphs accordingly so that the discussion of 2010 and 2030 effects flows better. P20891, L1: Figure 8 I like. Figure 6b is definitely unnecessary having seen figure 8.

These two comments refer again to the issue of whether to retain Figure 6. See our response to this comment above.

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