acp-2018-690: Advanced methods for uncertainty assessment and global sensitivity analysis of a Eulerian atmospheric chemistry transport model

by Aleksankina et al.

<u>Response to reviewer #2</u>

This discussion paper by Aleksankina et al. documents a global sensitivity and uncertainty analyses for the regional chemical transport model EMEP4UK, with the objective of quantifying the uncertainty in surface concentrations of air pollutants (ozone, nitrogen dioxide, and particulate matter below 2.5 um in diameter) and the contribution to that uncertainty from uncertainties in UK-only emissions. No uncertainties associated with model transport and/or chemical processes, or the lateral boundary conditions or driving meteorology were considered. I found the paper to be well organised, well written, and a really nice example of applying powerful statistical approaches to understanding model behaviour and uncertainties. The discussion on the sensitivity analysis itself was very interesting and shows how insightful this technique is. The paper will add to the growing literature base on the use of Gaussian emulation in quantifying uncertainties in geophysical models. I wholeheartedly recommend that the paper is accepted and published in Atmos. Chem. Phys.

Response: We much appreciate the reviewer's comments on the merits of the paper and their enthusiastic recommendation for its acceptance and publication. Thank you.

However, I have a few comments which I hope the authors will consider when submitting a revised manuscript:

1) *Intro: For the non-specialist, I think it would be worthwhile to include some basic introductory material on what you mean by sensitivity analysis versus uncertainty analysis.*

Response: The following text has been added to the Introduction (p2, L26):

"The main distinction between uncertainty and sensitivity analysis is that uncertainty analysis is performed to quantify model output uncertainty arising from the uncertainty in a single or multiple inputs, whilst sensitivity analysis is performed to investigate input–output relationships and to apportion the variation in model output to different inputs. Hence the sensitivity analysis allows conclusions to be drawn on the extent to which the overall variation in the modelled values is driven by variation in different inputs (Saltelli, 2002)"

2. Can you include some discussion on structural uncertainty?

Response: The following text has been added to the Introduction (p2 L17).

"There are various sources of uncertainty in a model; the sources range from structural or conceptual uncertainties about how well a given model represents reality to uncertainties in the model input data and physical and chemical constants, which have an effect on calculation results of the model."

3. *Intro: Note that aerosols affect climate through aerosol-cloud interactions and not only aerosol-radiation interactions*

Response: The first paragraph of the Introduction has been amended to now read as follows:

"Additionally, particulate matter and O_3 contribute to climate change through radiative forcing and aerosol-cloud interactions (for PM) (IPCC, 2013; Stevenson et al., 2013) and O_3 has an adverse impact on natural and semi-natural vegetation and crop yields (Teixeira et al., 2011)."

4. *Intro: Meta models have also been used in exploring climate sensitivity/climate response e.g. Murphy et al. (2004)*

Response: From our search of the literature we are assuming the reviewer is referring to the following paper: "Murphy, J. M., D. M. H. Sexton, D. N. Barnett, G. S. Jones, M. J. Webb, M. Collins, and D. A. Stainforth. Quantification of modelling uncertainties in a large ensemble of climate change simulations. Nature, 430, 768–772, 204." If so, we do not think it appropriate to include as an example of meta-model application in sensitivity analysis because the methodology for sensitivity analysis described in this paper is one-at-a-time (OAT) and is based on an ensemble approach.

5) *Section 2.1: Full names for SO2, NH3 etc.*

Response: Full names of chemical species are now added in the methods section where they first appear (p4, L19).

6. *Section 2.1: Can you include details of bvoc emissions scheme, and parameterisations for sea salt and dust emissions?*

Response: The following two blocks of text have now been added to section 2.1 (p4):

"Biogenic emissions of monoterpenes and isoprene are calculated by the model for every grid cell and time step according to the methodology of Guenther et al. (1993, 1995), using near-surface air temperature and photosynthetically active radiation as well as aggregated land-cover categorisations, as described in Simpson et al. (2012)."

And

"The details of the sea-salt generation parameterisation scheme used in the model are presented in Monahan et al. (1986) and Mårtensson et al. (2003). The boundary condition monthly average concentrations of fine and coarse dust are calculated with the global chemical transport model of the University of Oslo (Grini et al., 2005). The detailed parametrisation of dust mobilisation is presented in Simpson et al. (2012)."

7) *Table 2: Slight error with SNAP sectors for NH3_O (i.e. 10 should not be included!)*

Response: Thank you for spotting this typo which we have now corrected.

8) *Results Section 3.1: You say that there is a "substantial contribution of hemispheric background O_3 to UK ambient concentrations"? Can you be more quantitative here?*

Response: We cannot estimate the first-order effect of the background O_3 on the UK surface concentrations of O_3 as this input was not one of the perturbed inputs in this study. (We focused on perturbation of primary anthropogenic emissions.) It is not at all straightforward to quantify the background contribution to a secondary pollutant such as O_3 because not only is there import of O_3 and of O_3 precursors into the UK, but the UK is also a surface sink for O_3. The statement in our paper was based on Simpson et al. (2012) who state that "ambient ozone levels in Europe are typically not much greater than the Northern hemispheric background ozone". Additionally, in EMEP4UK a "Mace-Head" adjustment is applied to monthly boundary condition values of the O_3 concentrations. Hence in this paper the contribution of hemispheric background O_3 to the UK ambient concentrations is offered as a possible explanation of the lack of sensitivity of the surface O_3 to changes in the precursor emissions.

We have now added the citation to Simpson et al. (2012) to the end of the sentence in question.

9) *Results Section 3.1: You refer to the 'compensation of errors' as one explanation why the surface response is weak given the input uncertainties. Can you point to the literature for evidence of this statement? I've only seen "compensation of errors" only referred to in the context of process representation in models.*

Response: The phrase comes from Skeffington et al. 2007. In that paper the reason for narrowing of confidence limits for critical loads compared to those of the input parameters was explained to be due to a "compensation of errors" mechanism, but no further explanation was provided. Here, by compensation of errors we mean a situation when the variation in the output is less than expected. This could be caused by multiple inputs having an opposite effect on the magnitude of change in the output of interest.

We have changed the phrasing in our paper to "so-called compensation of errors".

10. *Results Section 3.3: One potential explanation for the seasonal change in sensitivity at Harwell to shipping emissions is the seasonal change in the wind direction which results in more NOx from shipping emissions being transported to the site. Can this be verified from the WRF meteorology used to drive the model?*

Response: The seasonal wind speed and direction for the year 2012 is shown in Figure A below, using the meteorology supplied from the AURN data as extracted using the openair package. It could be argued that there is some correlation between sensitivity index patterns in Figure 8 of our paper and the wind direction; however most likely the seasonality in NO_x sensitivity to shipping emissions is due to a combination of interacting processes within the model.

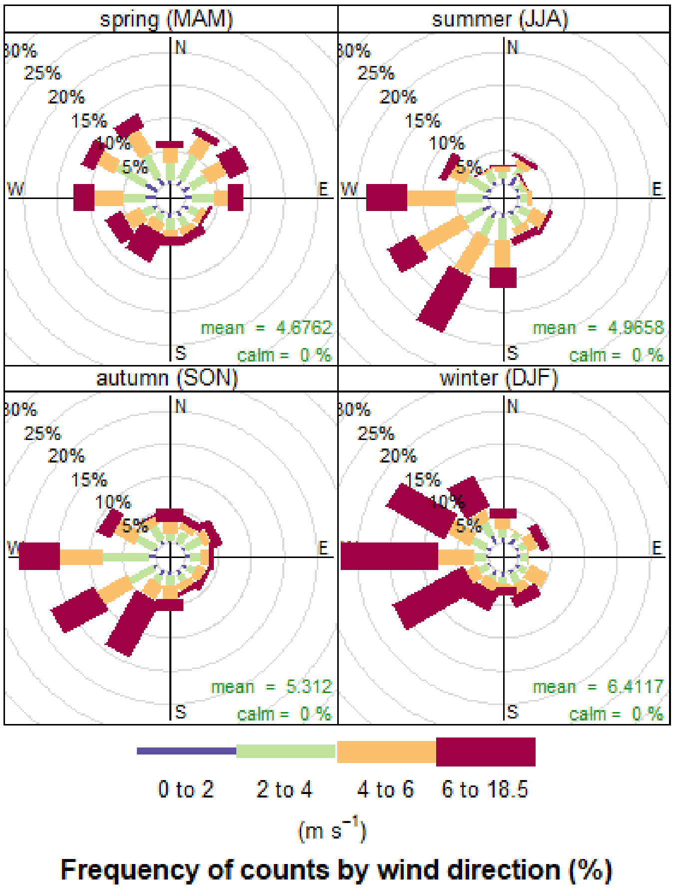

Figure A. Wind rose, Harwell AURN site, 2012.

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