Response to referees

We thank both referees for their thoughtful comments on our manuscript. Please see our comments below. To guide the reader, we have italicized the referees' comments.

Anonymous Referee #1:

This manuscript is a continuation for the previous work by Stevens et al. (2012, ACP) investigating new aerosol particle formation in power plant plumes. The novel feature of this paper is that it provides, to my knowledge, the first comprehensive parameterization of in-plume aerosol formation that can be used in large-scale modeling frameworks. I find the manuscript useful for the scientific community and definitely original enough to be published in Atmospheric Chemistry and Physics. There are no apparent errors in the manuscript, but the overall presentation could be made a bit more reader-friendly, especially what it comes to applying the derived parameterization by others. My detailed comments are given below.

The idea of providing advise how to use the parameterization in case some essential model input is missing is a very good one (see Appendix). I think the authors should provide some other type additional information as well, especially for those who are willing to program this parameterization by themselves. First, most of the equations used in the parameterization are not scale-independent in the sense that they contain a mixture of numerical values and physical quantities. As a result, the user needs to know in which units each of the quantities need to be inserted into these equations. This information should be given somewhere in the paper, either after each equation or in a separate table. Second, it might be worth summarizing somewhere (e.g. in Appendix) which model output corresponds to which equation, and which equations are really needed to calculate all the desired output quantities (are all the equations presented in section 3 necessary?).

Response:

We note that we have provided Fortran code of the parameterization as a supplement. However, we recognize that this will not be sufficient for some users, and we wish to make the use of the parameterization as easy as possible. We also thank the reviewer for pointing out that while the units for each input are available throughout the paper and in the tables, they are currently dispersed and not concisely listed for the reader. We have therefore included the units for all of the inputs and outputs when they are listed at the end of Sect. 1 and again at the beginning of Sect. 3. For other variables, such as OH_{eff}, $NO_{x,eff}$, and $SO_{2,eff}$, we now explicitly state the units where they first appear in the text.

As recommended by the reviewer, we have added a second appendix summarizing which equations are necessary to calculate each of the outputs of the P6 parameterization. If a user wishes to calculate all four outputs, they would indeed need nearly every equation listed in Sect. 3.

Referee #1: *When discussing the accuracy of the parameterization, it should be mentioned explicitly that while the parameterization does a good job in reproducing model simulations, its accuracy cannot be not better than the underlying theory, in this case the assumed linear dependency of the nucleation rate on the gaseous sulfuric acid concentration. It is clear that our understanding on in-plume sulphuric acid production and connection between the sulphuric acid and both nucleation and subsequent nuclei growth rate continues to be far*

from perfect.

Response:

We have added the following discussion of the nucleation scheme to the description of the SAM-TOMAS model:

"We note that it is clear that such an empirical scheme will not capture all of the variability in nucleation rates. However, an increase in the nucleation rate by a factor of 10 was found in Stevens et al. (2012) to increase N_{new} by a factor of about 3 for distances greater than 30 km from the source, and we will show that values of N_{new} span six orders of magnitude across the set of training data used for this study. As more accurate parameterizations of nucleation become available, we plan to itegrate them into SAM-TOMAS and incorporate the results into future versions of the P6 parameterization."

We have also added the following to the conclusions section:

"While the P6 parameterization reproduces well the behaviour of the SAM-TOMAS model, we note that it inherits the limitations of the SAM-TOMAS model. Aqueous-phase oxidation of SO_2 is not accounted for, and therefore f_{ox} may be underpredicted under cloudy conditions. Nitrous acid (HONO) and sulphur trioxide (SO_3) emission are not accounted for, and these processes may result in particle formation early in the plume. Nucleation rates are parameterized using an empirical fit proportional to H_2SO_4 concentrations. Despite these limitations, the SAM-TOMAS model has been previously shown to well represent the formation and growth of aerosol in coal-fired power-plant plumes (Stevens et al., 2012; Lonsdale et al., 2012). We therefore believe that the P6 parameterization captures well the variablity in new-particle formation and growth within sulphur-rich plumes."

Referee #1: *Finally, even though it is extremely important to investigate the sensitivity of the parameterization to various input parameters, I am still not convinced about the usefulness of Figures 2 to 6 in their current form. Each sub-plot of these figures contain a large number of lines, and very few of them a really informative. I do not say that some of these figures should be removed from the paper, I simply encourage the authors to reconsider if all this information could be provided in a bit more compact way.*

Response:

We agree with the reviewer that the figures were dense, and that the sensitivities could be shown more elegantly. We therefore have re-plotted these figures, after removing the median value from each plotted line. Where we plot f_{ox} or f_{new}, we have subtracted out the median value of each line, and where we plot M_m or N_{new}, we have divided out the median value of each line, so that the sensitivities to the inputs are highlighted. We show the new figures here as figures A-E at the end of the responses.

Anonymous Referee #2

The authors present a parameterization of sub-grid sulfate formation for use in global and regional-scale models, which, due to their coarse resolution, cannot directly represent new particle formation in sulfur-rich plumes. The parameterization utilizes commonly available input parameters from global- or regional-scale models. While the authors developed and

present a really useful parameterization, I would expect at least one concrete application of this parameterization in a global model. The current manuscript, as is, is more suited for publication in GMD rather than ACP. Based on the presentation of results/figures the implications for global modelers are not necessarily clear.

To warrant publication in ACP the authors ought to demonstrate how the use of this parameterization affects, for example, aerosol particle number and mass concentrations in a global model. In fact, the Conclusions section does not provide any further or new insights into the issues raised (which rightly motivate and warrant such a parameterization) in the Introduction. I am certainly not suggesting a full-blown assessment or comparison of a global model with and without the parameterization. Doing so could also help to outline how other groups could implement the parameterization.

Response:

We agree with the referee that the implications of implementing the P6 parameterization into a global model were not made clear in the manuscript. We currently plan to perform a complete comparison of the results of a global chemical-transport model with and without the parameterization as well as looking at the effects of emissions controls as a future manuscript. We would prefer to publish the global model results along with the full discussion that we feel will be necessary. However, some implications for global aerosol number and mass are clear from the training cases used to create the P6 parameterization. We have added the following discussion of the implications of the P6 parameterization to the conclusions section:

"The median value of f_{ox} predicted by the P6 parameterization (0.0098) for the training cases is much less than the fraction of emitted SO_2 mass added as sub-grid-sulphate by Adams and Seinfeld (2003) (0.03) or Dentener et al. (2006) (0.025). Additionally, we excluded night-time cases from our training data, where no oxidation of SO_2 and no new-particle formation would be predicted by the P6 parameterization. Consequently, we expect that predictions of total aerosol mass near sulfur-rich point sources using global-scale models implementing the P6 parameterization will be less than those using the Adams and Seinfeld (2003) or Dentener et al. (2006) assumptions. Additionally, as the median values of both N_{new} and M_m predicted by SAM-TOMAS were less than those predicted by Adams and Seinfeld (2003), we expect than both globally-averaged aerosol number concentrations and globally-averaged CCN concentrations would be less than those using the Adams and Seinfeld (2003) assumption, with large regional differences (e.g. less CCN formation using the P6 parameterization under cloudy, polluted conditions than sunny, low-background-aerosol conditions). It is our intent to perform a complete comparison of the results of a global chemical-transport model with and without the parameterization as a future work."

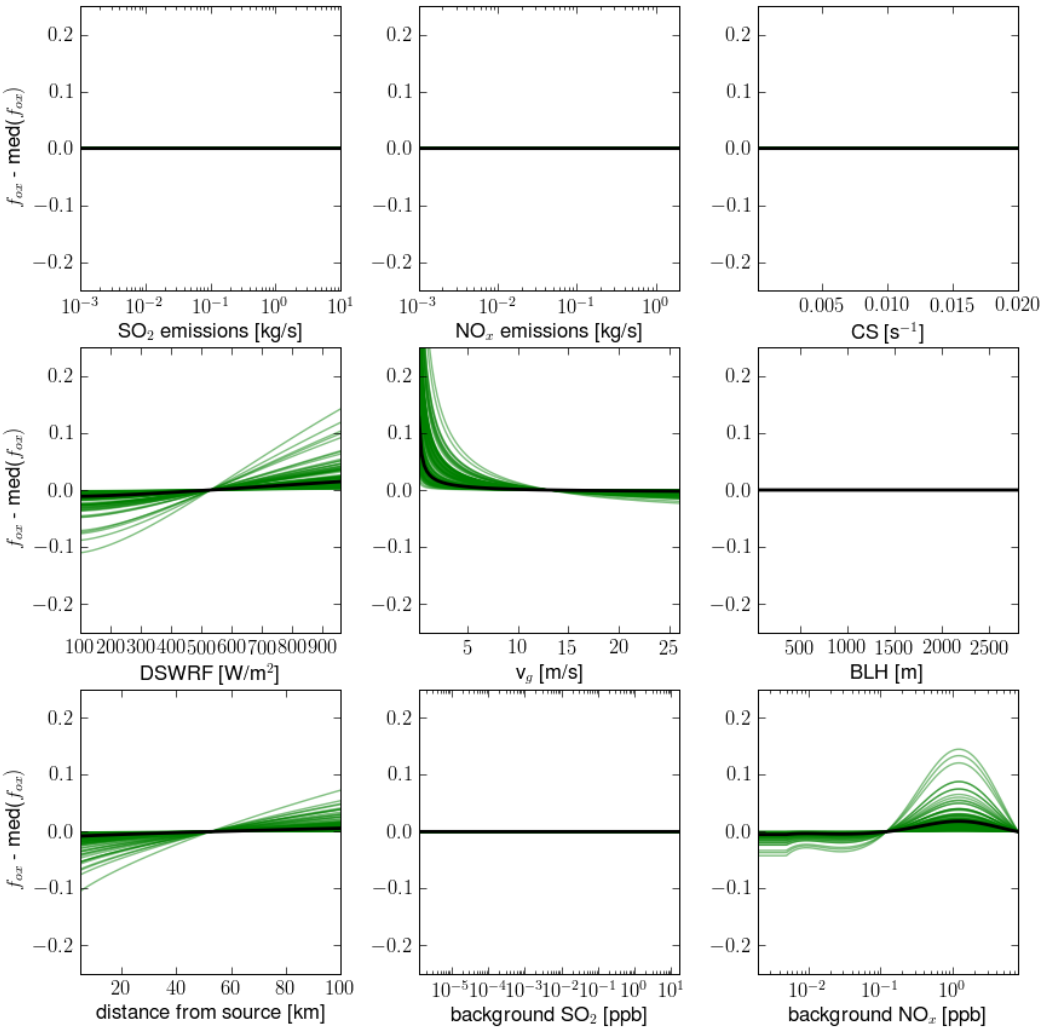

Figure A: Sensitivity of f_{ox} to each of the inputs for 100 randomly selected sample inputs. The black line denotes the median value case. The median value of each plotted line is subtracted from its values to highlight the sensitivities to the inputs.

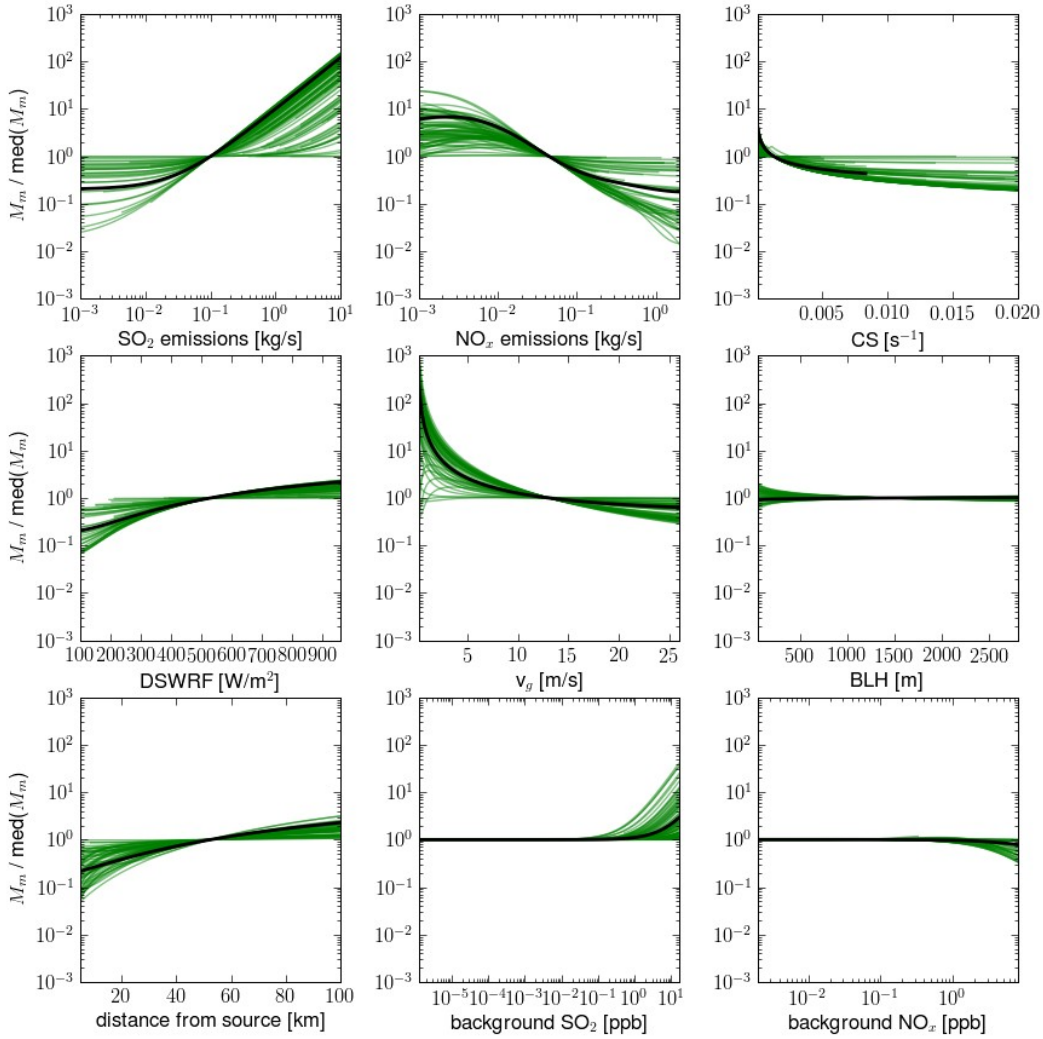

Figure B: Sensitivity of M_m to each of the inputs for 100 randomly selected sample inputs. If nucleation is not predicted by the P6 parametrization, no value is shown. The black line denotes the median value case. Each plotted line is divided by its median value in order to highlight the sensitivities to the inputs.

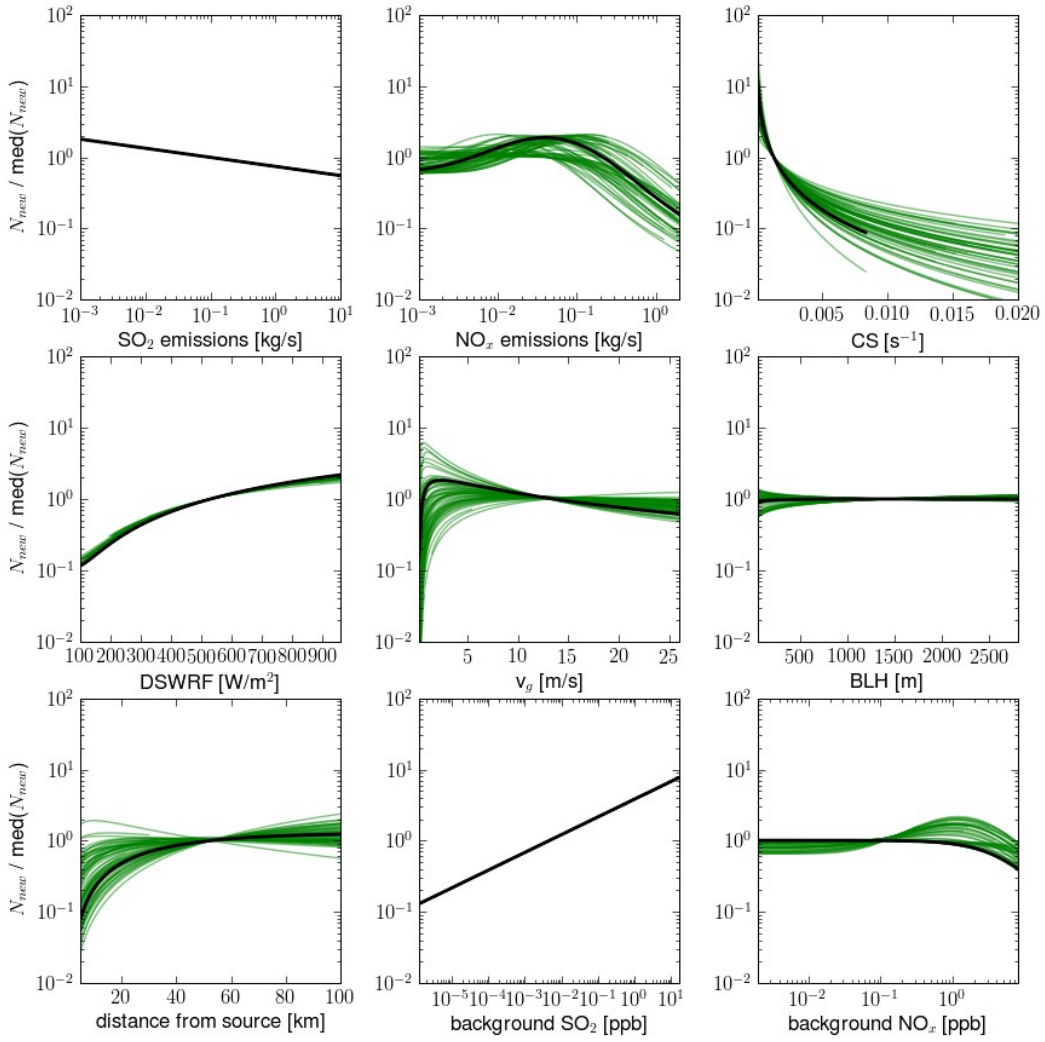

Figure C: Sensitivity of N_{new} to each of the inputs for 100 randomly selected sample inputs. If nucleation is not predicted by the P6 parametrization, no value is shown. The black line denotes the median value case. Each plotted line is divided by its median value in order to highlight the sensitivities to the inputs.

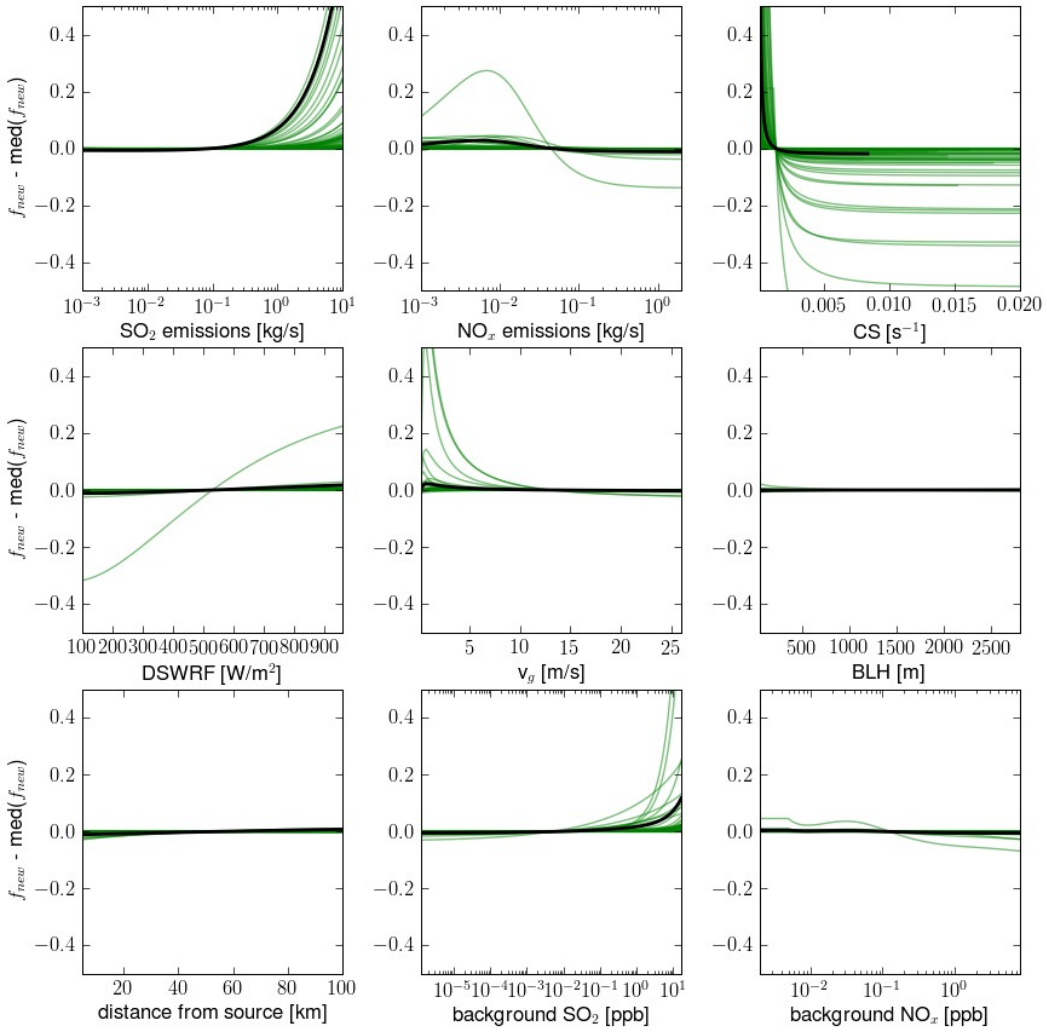

Figure D: Sensitivity of f_{new} to each of the inputs for 100 randomly selected sample inputs. If nucleation is not predicted by the P6 parametrization, no value is shown. The black line denotes the median value case. The median value of each plotted line is subtracted from its values to highlight the sensitivities to the inputs.

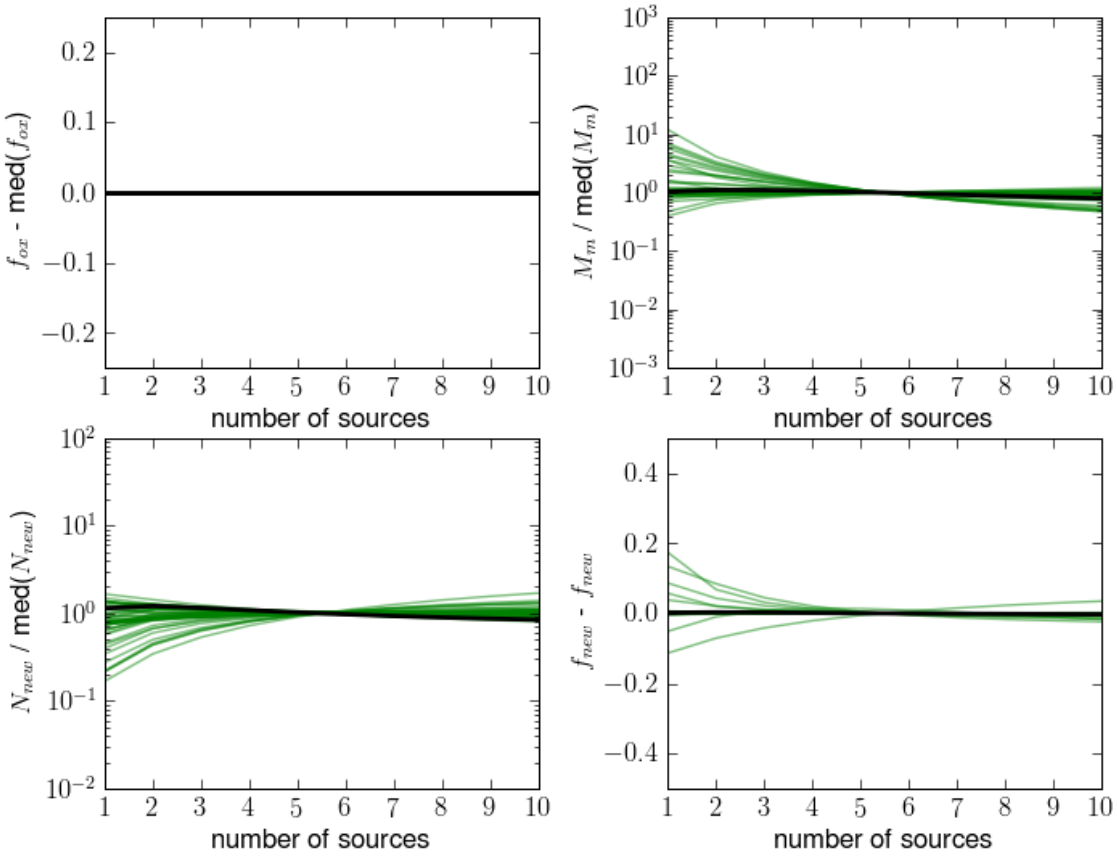

Figure E: Sensitivity of fox, Mm, Nnew, and fnew to the assumed number of emission sources, while keeping total emissions of SO2 and NOx constant, for 100 randomly selected sets of inputs. If nucleation is not predicted by the P6 parametrization, no value is shown. The black line in each figure denotes the median value case. In order to highlight the sensitivities to the number of sources assumed, for f_{ox} and fnew, the median value of each plotted line is subtracted from the line, and for M_m and N_{new}, each plotted line is divided by its median value.