**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Nucleation and growth of sulfate aerosol in coal-fired power plant plumes: sensitivity to background aerosol and meteorology" *by* R. G. Stevens et al.

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We thank both referees for their thoughtful comments on our manuscript. Please see our comments below.

Referee 1

 Referee 1: What I am missing here is some concrete recommendation(s) for large-scale modelers how to deal with the emissions leading to near-source new-particle formation. I am sure that there is something that can be recommended based solely on the performed analyses. For example, most modelers fix the total mass and size of emitted particles, after which these two quantities then dictate the number of emit-

ted particles. This is probably fine for larger primary particles emitted by the source. However, is it the best way to go for particles formed in the plume, especially when considering how much the size of newly-formed particles may vary depending on both the distance and extend of SO2 to sulfuric acid conversion? The authors certainly have some ideas on this issue and therefore I would encourage them to discuss it shortly in the paper.

Response: As shown in the sensitivity studies we have performed, the number and size of aerosol formed within a given power-plant plume can vary dramatically based on meteorology and background aerosol, and no fixed size distribution will provide an adequate representation under all conditions. We therefore do not wish to recommend any single aerosol size distribution for use under all conditions. We are currently creating a parametrization that would predict the median size and number of aerosol formed per kilogram SO2 emitted, as well as the fraction of SO2 oxidized and the fraction of H2SO4 formed that forms or condenses onto new particles, for given emissions, meteorology, and mean background aerosol condensation sink. We have added the following discussion to the conclusions section of the text:

"Until this parameterization is available, it may be wise to consider separately conditions under which it is likely there will rarely be significant aerosol nucleation within the plume. Based on our sensitivity studies, when OH concentrations are very low (for instance, at night) or when the background condensation sink is very high, it seems prudent to assume that all H2SO4 formed within the plume will condense onto the existing background aerosol, and that therefore aerosol mass should be increased without increasing number. Under other conditions, it may be preferable to assume the size distribution used by Adams and Seinfeld (2003) to the size distribution assumed by Dentener et al. (2006), as the former was closer to the results we obtained for every case where particle number concentration increased inside the plume. We wish to stress that this does not imply that it will be the better assumption under all conditions, but our results suggest that it may be the better assumption under conditions when

nucleation does occur."

Referee 1: Using roughly 50 lines in describing the OH parametrization in the model sound a bit long and technical issue as part of the main text. I think this part would better fit to an appendix.

Response: The material describing the OH parameterization has been moved to an appendix, and replaced with the following briefer description:

"We use a parameterization to estimate the concentration of OH in each model grid box based on the concentration of NOx in ppbv and the downward shortwave radiative flux (dswrf) in W/m2. While the NOx concentration is used to predict the concentration of OH, we do not currently have a chemical sink for NOx in the model, which will lead to an over-prediction of NOx later in the plume. The parameterization is an empirical fit to the results of many simulations from the detailed time-dependent photochemical box model described by Olson et al. (2006).

One process not accounted for in the OH parameterization is the effect of the presence of large amounts of highly reactive VOCs on OH production. The additional peroxy radicals from isoprene oxidation induce a shift in the peak OH production to a higher NOx level. To understand the potential effect of high VOC concentrations in our study, a second parameterization, referred to as the "high-VOC" case, was developed based on an isoprene mixing ratio of 1.5 ppbv (the 95th percentile value observed during INTEX-A). We refer to the original parameterization as the "low-VOC" case. The two parameterizations are outlined in detail in the appendix."

Referee 1: Page 24778, lines 5-8. This statement is not entirely correct. A Gaussian representation of a plume can be implemented in a way that allows mixing and chemical reactions within the plume and with ambient air mixed into the plume. This requires using grid boxes that grow in size as the plume is transported downwind from the source.

Response: While some of the effects that were described in this paragraph could be individually resolved with a Gaussian plume representation, such as the effect of the homogeneous background condensation sink on the processes, we are specifically referring to the regions of enhanced nucleation due to turbulent mixing in this sentence. Even allowing for grid boxes that grow in size as the plume is transported downwind, the heterogeneous structure of these regions cannot be resolved.

We have added the following to the text: "These turbulent eddies create regions within the plume with especially high and low concentrations of NOx and SO2 that alter the nucleation rate within the plume. This in turn causes the contribution of nucleation to the condensation sink to be inhomogeneous within the plume. As seen in Fig. 1, these turbulent mixing effects can cause the nucleation rate to vary by a factor of two within the plume, even at the same distance from the source. These inhomogeneous regions of enhanced nucleation, evident at the downwind plume edges, cannot be resolved using a model that assumes a pre-defined Gaussian plume".

Referee 1: Table 2. The table is not understandable by itself. Please add information on where to locate the explanation for the different cases given in the first column of the table.

Response: The text "The labels 400x400x40m and 800x800x40m refer to the two model resolutions used in this study. The A-6, Vehk, Meri, and Yu10 nucleation schemes are discussed in Sect. 4.1. The REM, MAR, and URB aerosol backgrounds are discussed in Sect. 4.2. The sunny and cloudy cases and the high-VOC and low-VOC cases are discussed in Sect. 4.3." has been added to the caption for Table 2.

Referee 1: Figures 4, 7, 8 and 9: The term "additional particles" should be clearly defined, and in such a way that the reader finds this information easily when looking at the figures.

Response: We have added the following to p. 24781, line 18: "We approximate this value in the model by subtracting the original background concentrations from the par-

ticle number concentration, and then we divide this by the background-corrected SO2 mass concentrations and integrate across the plume." We also include a reference to this ("see Sect. 3.1") in the caption for each of these figures.

Referee 2

Referee 2: In a number of cases throughout the text it would improve the paper to add a quantitative comparison to support qualitative statements: e.g., P24780, L8: "agree quite well"; P24780, L18: "agree better...slight low bias".

Response: We have changed the statements mentioned by the reviewer to the following. We have also quantified other qualitative statements throughout the text.

p. 24780, line 8: "agree quite well" has been replaced with "differ by less than 5%"

p. 24780, line 18: This sentence has been rephrased as follows: "The modeled concentrations of particles smaller than 50 nm are within 10% of the observed concentrations at the remaining transects, and the modeled concentrations of particles larger than 50 nm in diameter are 36% and 22% lower than the observed values at the third and fourth transects, respectively"

Referee 2: P24787, L13. I don't understand why this is the case: "pre-existing particles smaller than 30 nm that were mixed into the plume; however, these are an artifact of our calculation and should not truly be considered new particles. " Please explain and if possible clarify in the manuscript.

Response: As mentioned in our response to the last point by Referee 1, when calculating additional particles we subtract out the original aerosol background. As the background particles smaller than 30 nm grow beyond the 30 nm cutoff, the total number of particles larger than 30 nm increases. If we compare the number of particles larger than 30 nm at some distance downwind of the power-plant to the original background concentration, we see an increase, however this is not due to new-particle formation but instead due to condensational growth of pre-existing sub-30 nm particles. The

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sentences in question have been changed to the following:

"There are, however, additional particles larger than 30 nm for the Vehk case because of condensational growth of pre-existing particles smaller than 30 nm that were mixed into the plume. This causes the concentration of particles larger than 30 nm to increase above the original background concentration; however, these are an artifact of our calculation and should not truly be considered newly-formed particles."

Referee 2: P24787, L21. Change "Spracklen, 2008" to "Spracklen et al., 2008" and "Sihto, 2006" to "Sihto et al., 2006". There may be others I did not spot, please carefully check throughout.

Response: These citations, and others, have been corrected.

Referee 2: P24787, L18. Add "(equation 1)" after "fitting parameter A".

Response: This has been added.

Referee 2: Figs 3b, 4, 6b etc. I found these plots hard to read and this may be worse when they appear smaller in the ACP version. Restricting the y-axis to a narrower range in plots 3b and 6b, making lines thicker etc might make the plots more readable.

Response: The suggested changes have been made to Figs 3b, 4, 6b, 7, 8, 9, and 10b.

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