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***Interactive comment on “The effect of coal-fired power-plant SO<sub>2</sub> and NO<sub>x</sub> control technologies on aerosol nucleation in the source plumes” by C. R. Lonsdale et al.***

**C. R. Lonsdale et al.**

chantelle.lonsdale@gmail.com

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Response to referees comments on “The effect of coal-fired power-plant SO<sub>2</sub> and NO<sub>x</sub> control technologies on aerosol nucleation in the source plumes” Lonsdale et al.,

We would like to thank both of the referees for their comments and discussion regarding this paper. Their time and effort is greatly appreciated. Below are specific responses to their comments.

Referee 1: However, the manuscript does not give any information about the particle sizes produced. This size distribution would be critical in comparing the model to measurements, either to the existing results of the Texas (Parrish) plume measurements or

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any other future power plant plume experiments. Differences or agreement between the model and the experiment could be a valuable information about a possible contribution of initially emitted new particles that do not need OH radicals to be produced, particles from SO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> production described by Srivastava (2004) inside the power plant.

And

Referee 2: Unfortunately the paper gives no information about the impact on particle number concentration (cm<sup>-3</sup>) at difference size regimes (e.g. from 3 – 100 nm, >100 nm, etc.). It would be really interesting if the authors could add some information on that.

Response: Plots of size distributions for the model vs measurements have been added for both 2000 and 2006 (Figures 7 & 8) with the following text added to the paper; 'The particle size distribution of the model simulations and flight measurements are shown in Figures 7 and 8. Figure 7 contains the simulated (solid lines) and aircraft-measured (dashed lines) size distributions for the 2000 case (high-2000 emissions and clean-2000 background in the model) with the size distribution averaged across the plume at the four transects made by the plane; 3.2 km (blue), 19.6 km (red), 39.5 km (green) and 54.5 km (purple) from the source stack. Figure 8 contains the model and aircraft-measured size distributions for the 2006 case (low-2006 emissions and polluted-2006 background in the model) at the three transects made by the plane; 4.8 km (blue), 35.9 km (red) and 53.0 km (green) from the source stack. It should be noted that the model domain ended at 50 km down wind of the stack, and the size distributions at this edge of the model domain are used for comparison to the furthest aircraft transects. Similar to the NPC rates in Figure 6, the model does better at predicting particles sizes further downwind in the plume. In both cases the model does not capture a measured mode with a median diameter around 20 nm at the closest transects. It is these 20-nm particles that are the source of the low model bias in Figure 6. This initial peak may be particles that formed in stack due to SO<sub>3</sub> emissions (Junkermann et al., 2011b; ;

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Srivastava et al., 2004). SO<sub>3</sub> emissions may be due to the FGD on one of the four units, or they may be due to the SCR on all four units, although the SCR was only installed for the the 2006 case and wouldn't influence the 2000 case. It is also possible that photolysis of HONO emitted by the power plant is contributing to enhanced OH concentrations near the source (Gonçalves et al., 2009, Elshorbany et al., 2010). The model does not simulate HONO and would miss this extra OH. We plan to investigate the role of SO<sub>3</sub> and HONO in future work. In both cases, the model captures the evolution of the size distributions increasingly well with later transects showing that the in-plume processes simulated by the model are dominating the size distribution outside of the first 10-20 km. However, the initial 20-nm mode, while diminished due to coagulation, grows to enhance ~100-nm particles that are not captured by the model.'

Referee 1: Also not mentioned, but possibly important for the emission szenarios is the change in the particle size distributions in the plumes from fine to ultrafine particles. The changes in NO<sub>x</sub> and SO<sub>2</sub> emissions in the short time scale from 1997 to 2010 for all the US power plants are possibly minor compared to the changes following the introduction of filters that removed most of the primary emitted fine particulate mass.

Response: The comment concerning changes in power-plant plumes due to primary emitted fine particulate mass was addressed in this paper already. It was mentioned that primary particles were not included in this study because such emissions are not released by the clean air emissions inventory and they are assumed to be almost negligible (filter efficiencies ~99%) during the time period spanning 1997 to present day when we have data. However, we have added more discussion to the paper for clarification based on this comment .

We did perform an analysis of primary particulate emissions influence on nucleation in the power plant plume, but did not include them in our manuscript because of the lack of quality emissions data. See the Figure 1 below . In brief, several model simulations were run with a range of primary particulate emissions rates in order to estimate the new-particle contributions in plumes prior to the installation of primary particle controls.

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The primary particles were emitted into the 1997 and 2010 simulations used in the paper. Historical rates and sizes derived for this study were acquired from Hegg et al., 1985, Hegg and Hobbs 1980. The median diameter and sigma for the primary particles were 500 nm and 2.2, respectively. We use  $6E14$  particles  $s^{-1}$  as a lower bound and  $1E16$  particles  $s^{-1}$  as an upper bound. In the 1997 simulations, there is no NPC even in the absence of primary particles. In the 2010 simulations, only the  $1E16$  particles  $s^{-1}$  rate is sufficient to impact particle formation. Thus it is possible the the ash particles may have affected particle formation in the plumes, but better-constrained ash emissions are needed to make sufficient estimates.

Hegg, D.A. And Hobbs, P.V.: Measurements of gas-to-particles conversion in the plumes from five coal-fired electric power plants, *Atm. Environ.*, 14, 99-116, 1980  
Hegg, D.A., Hobbs, P.V., and Lyons, J.H.: Field Studies of a power plant plume in the arid southwestern United States, *Atm. Environ.*, 19, 1147-1167, 1985

Referee 1: The two figures 4 and 6 show a difference of more than an order of magnitude in the particle production rate. Compared to 2006 with further reduced NO<sub>x</sub> emissions the OH concentration should be even higher, see figure 1. How can this be explained?

Response: For clarification, Figure 4 compares the time ranges of 1997 and 2010 (the full time range available from the Clean Air Market Data). Figure 6, however, is comparing a shorter time frame of only 2000 to 2006 (the time of the available flight measurements from the TexAQS of 2000 and 2006), therefore the NO<sub>x</sub> reduction is less for Figure 6 (see the NO<sub>x</sub> emissions for the 4 years in question in Figure 2), providing different OH concentration differences for each of these figure.

Referee 1: Figure. 3 is used as an illustration for relatively clean and polluted background conditions. For readers that are not that familiar with size distributions, can these data be converted into something like PM<sub>2.5</sub> values?

Response: Unfortunately PM<sub>2.5</sub> values cannot be shown because measurements

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were performed for particles smaller than  $\sim 600\text{nm}$ , this may exclude a large portion of the  $\text{PM}_{2.5}$  mass. The total mass of particles smaller than  $600\text{ nm}$  ( $\text{PM}_{0.6}$ ), however, was calculated to be  $6.9\ \mu\text{g m}^{-3}$  for the clean-2000 background and  $9.3\ \mu\text{g m}^{-3}$  for the polluted-2006 background. The following has been added to the text for additional aid in understanding the size distributions; “ The total mass of particles smaller than  $600\text{ nm}$  ( $\text{PM}_{0.6}$ ) for the 2000 background and 2006 background was  $6.8\ \mu\text{g m}^{-3}$  and  $9.3\ \mu\text{g m}^{-3}$  respectively. The difference between the  $\text{PM}_{0.6}$  values of the two cases are more similar than the difference between the condensation sink values because the size distribution for the clean-2000 case is skewed towards larger diameters (see Fig. 3).”

Referee 1: The second half of Section 5.3, "Observational evidence and comparison" is difficult to read. This might be due to the frequent jumps in the text from 2000 (high) to 2006 (low) emissions and 2000 (low) and 2006 (high) background conditions. An additional table could help as the figures 3 and 6, which are necessary for comparison with these results most probably are not very close to the text in the final version.

Response: Rather than referring to the 2000 background, 2006 background, 2000 emissions and 2006 emissions cases, we now say clean-2000 background, polluted-2006 background, high-2000 emissions and low-2006 emissions for clarity. We made a table, but we found that was not as useful as the name change and did not add value to the paper, so we did not include it.

Referee 2: In the abstract most of the main results of the paper are described very generally and qualitatively. E.g. “: : These results suggest that  $\text{NO}_x$  emissions may strongly regulate particle nucleation and growth in power-plant plumes.” Or “: : highlight the substantial effect of background aerosol loadings on this process”. I think you should be more quantitative and give specific numbers when using words such as “strong”, “substantial”, etc.

Response: We now have specified that the nucleation increased by an order-of-

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magnitude when the NO<sub>x</sub> emissions were reduced by 10x in the sentence before the first example. For the 2nd example, we've added, "(the more polluted background of the 2006 case caused more than an order-of-magnitude reduction in particle formation in the plume compared to the cleaner test day in 2000)."

Referee 2: The main conclusions from the results are two and are contradicting. When looking at the Parish power-plant the effect of NO<sub>x</sub> is strong and results in a considerable enhancement of particle formation. But when looking at the US median of the power plants the particle formation rate is decreasing. These two are equally highlighted in the paper/abstract, and thus the reader is rather confused. Since the Parish plant is an extreme case (as explained in page 19699) due to extremely high NO<sub>x</sub> decrease, this should be pointed out in the abstract. The general conclusion about the effect of the controls to US plants is that nucleation is on average decreased. This should be more emphasized in the abstract.

Response: We have added the following sentence to the abstract, "Thus, the US power plants, on average, show a different result than was found for the W.A. Parish plant specifically, and it shows that the strong NO<sub>x</sub> controls (90% reduction) implemented at the Parish plant (with relatively weak SO<sub>2</sub> emissions reductions, 30%) are not representative of most power plants in the US during the past 15 years. These results suggest that there may be important climate implications of power-plant controls due to changes in plume chemistry and microphysics, but the magnitude and sign of the aerosol changes depend greatly on the relative reductions in NO<sub>x</sub> and SO<sub>2</sub> emissions in each plant."

Referee 2: How are organics treated in the TOMAS algorithm? Do organic vapors assist the growth of particles <100 nm in the model implementation? If not then this should be mentioned in the paper and the fact that organics probably do play a significant role information and growth which could considerably affect some of the paper's results.

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Response: Secondary Organic Aerosol (SOA) is currently not accounted for in this version of the model. We have added the following text to the manuscript, “The formation of Secondary Organic Aerosol (SOA) is not considered in this study. Our assumption is that H<sub>2</sub>SO<sub>4</sub> formation will dominate growth in the SO<sub>2</sub>-rich power-plant plumes, and we were able to reproduce most of the growth in two plumes in Stevens et al. (2012) (also see Figures 7 and 8). However, the lack of SOA is an additional uncertainty in this study.”

Referee 2. I think that some other modeling studies which looked at the impact of reducing SO<sub>2</sub> and/or NH<sub>3</sub> emissions on aerosol nucleation should be referenced here (e.g. Jung et al. 2010; Fountoukis et al., 2012, etc.)

Response: We have added the following sentence to the paper, “Regional changes in nucleation and growth are also expected due to changes in pollution controls (Jung et al. 2010; Fountoukis et al., 2012), and these effects must be considered along with the in-plume nucleation changes considered here.”

Referee 2: The choice of nucleation parametrization is also an issue here. Although I haven't looked at Stevens et al. 2012 results, there is clear evidence now that NH<sub>3</sub> could also be involved in the formation process and a more appropriate nucleation parametrization (e.g. a ternary nucleation scheme) could give different results. Could the authors comment on that?

Response: It was mentioned that in Stevens et al. 2012 several different nucleation schemes were modelled against measurements (including ternary nucleation), and activation nucleation provided the best agreement at the two sites studied (including the W.A. Parish plant). Below is Figure 8 from Stevens et al. (2012), which has this comparison of particle number >3 nm and particle number >30 nm for 5 simulations using different nucleation schemes at the W.A. Parish plant. Unfortunately, there was only NH<sub>3</sub> measurements for one comparison case, so this was the only case where we were able to evaluate nucleation schemes with NH<sub>3</sub>. A-6 and A-7 are simulations using

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activation nucleation with  $A=10^{-6}$  and  $10^{-7}$  s $^{-1}$ , respectively. Vehk is the Vehkamäki et al. (2002) binary scheme (no nucleation predicted). Meri is the Merikanto et al. (2007) Ternary scheme. Yu is the Yu et al. (2010) ion-mediated nucleation scheme assuming an ion-pair production rate of 10 pairs s $^{-1}$ . We also tested the Napari et al. (2002) ternary nucleation using a scale-factor of  $10^{-4}$ , but its results were very similar to the Merikanto scheme in this case. Since activation with  $A=10^{-6}$  s $^{-1}$  performed the best here, and we have not had ammonia information for any other test case, we have continued to use the activation scheme with  $A=10^{-6}$  s $^{-1}$ .

Regarding the role of NH<sub>3</sub> slip from power plants on nucleation in the powerplant plumes, we have a paper in preparation regarding this, and we have the following sentence in the paper, “Since NH<sub>3</sub> is potentially an important enhancer of aerosol nucleation rates (Merikanto et al., 2007; Kirkby et al., 2011), this too may affect particle formation. This influence on particle formation is not explored in this paper, but is explored in Gong et al. (2012).”

Gong, L., Lewicki, R., Griffin R., Lonsdale, C., Stevens, R., Pierce, J., Malloy, Q., Travis, S., Bobmanuel, L., Lefer, B. and Flynn, J. (2012) Atmospheric ammonia measurements and implications for particulate matter formation in Houston, TX, in prep, for Atmos. Env., 2012.

Referee 2: On page 19699 the authors state that “a full analysis of the effect of power-plant emissions changes on aerosol concentrations using a regional chemical transport model is planned for future work, which will yield a more comprehensive estimate than provided here.” What do you think would be the advantages of using a regional CTM compared to the model used here?

Response: We have modified the sentence to better state our intentions, “However, a full analysis of the effect of power-plant emissions changes on aerosol concentrations using regional and global chemical transport model (using a parameterized version of sub-grid nucleation based on results from the SAM-TOMAS model) is planned for

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future work. This analysis will allow for emissions changes to be considered over the full range of atmospheric conditions and aerosol properties to be predicted further from the sources.”

Referee 2: On page 19691 the authors say “The large-scale meteorological forcing of the SAM simulations : : :”. What is the scale used in the meteorological input? Wouldn't sub-grid meteorological variability be another source of uncertainty?

Response: We are not sure if you are asking about sub-grid meteorological variability in the large-scale forcing data (32 km x 32 km horizontal resolution, now mentioned in the text) or in the SAM model (400 m x 400 m horizontal resolution). SAM resolves turbulent motions explicitly for scales larger than 400 m, and SAM predicts sub-grid diffusion for motions smaller than 400 m x 400 m using a Smagorinsky-type scheme. However, any systematic (non-turbulent) variability in meteorology (e.g. flow around hills or buildings) won't be captured by the SAM model. This can lead to uncertainty in the model (and likely lead to errors in plume flow during the simulation of the Conesville, Ohio plant in the Stevens et al., (2012) paper). We have added text regarding the systematic variability in the model. “Although SAM resolves turbulent flows on spatial scales smaller than the, any systematic (non-turbulent) variability in meteorology (e.g. flow around hills or buildings) won't be captured by the SAM model, and this could lead to uncertainties in the plume dispersion.”

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 19683, 2012.

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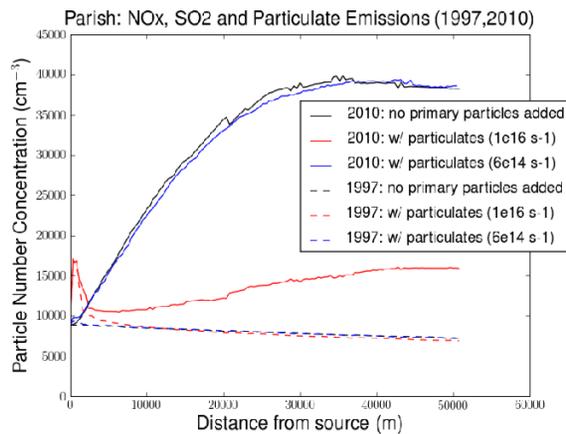
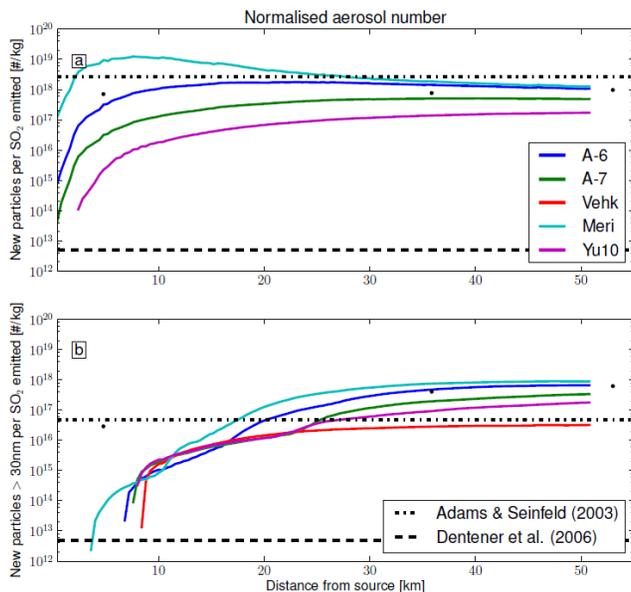
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Fig. 1.

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**Fig. 8.** (a) Total additional predicted particles and (b) additional particles larger than  $30 \text{ nm kg}^{-1} \text{ SO}_2$  versus distance from the Parish power-plant, using several nucleation schemes. See Sect. 3.1 for a description of the calculation. Values are averaged over the plume. Model resolution is  $400 \times 400 \times 40 \text{ m}$ . Black dots indicate aircraft observations, solid colored lines indicate model results, black dashed lines indicate the values used by Dentener et al. (2006), and black dashed-dotted lines indicate the values used by Adams and Seinfeld (2003).

Fig. 2.