Interactive comment on "New particle formation in the fresh flue gas plume increase the effective particle number emissions of a coal-fired power plant" *by* F. Mylläri et al.

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We believe that the authors present valuable measurements and analysis in this article, but we feel that there are several misrepresentations of our own work.

Starting on line 56, the authors state: *"Stevens et al.(2012) and Lonsdale et al. (2012) have compared these measurements to modelling results, which were based on emission inventory values. Modelling results indicated that secondary particle formation occurs in the plumes after 10–20 km from the stack, [..]"*

Additionally, starting at line 381, the authors state: *"The power plant plume dilutes to background levels in 200 seconds which is faster than indicated in other in-flight mea-*

surements (Stevens et al., 2012; Junkermann et al., 2011). According to modelling results of Stevens et al. (2012), atmospheric new particle formation via coal combustion originated sulphuric acid nucleation begins at 5 km distance from the source whereas the sulphuric acid formation begins right after emission. Experiments of this study indicates that the nucleation may take place in the aged plume and being the most effective after 400 seconds, corresponding approximately 2 km distance from the emission source in atmosphere. Also this distance is significantly less than 5 km distance indicated by Stevens et al. (2012). Thus, this study indicates that atmospheric nucleation in power plant plumes takes place faster than the models and measurements have suggested before."

We note that the time required for a plume to dilute to background concentrations depends on the strength of the source, the background conditions, and meteorology. It would not be surprising, for example, if a power plant with lower emissions in a more polluted environment diluted to background conditions more quickly than the specific case studies discussed in Stevens et al. (2012) and Lonsdale et al. (2012).

In Fig. 1 of Stevens et al. (2012), we show that the SAM-TOMAS model predicts positive nucleation rates at distances as low as 1 km from the emissions source. Many of the other figures from the paper (Fig 2b, 4, 6b, 7, 8a, 9a, and 10b) show increases in modelled total particle number due to new-particle formation at distances less than 2 km from the source for both case studies and with different model configurations. Figures 4 and 6 from Lonsdale et al. (2012) show increases in the net particle contribution rate at distances less than 5 km, and these increases are due to new-particle formation occurring within the power-plant plume. Therefore our model results, previously published in Stevens et al. (2012) and Lonsdale et al. (2012), show new-particle formation occurring in power-plant plumes at distances less than 5 km from the source.

Additionally, in-flight measurements were performed within the Parish power-plant plume at distances of 5 km or less in the years 2000 and 2006. Some of these observations are shown in Stevens et al. (2012) and Lonsdale et al. (2012). In both

years, the observations show greater particle number concentrations than those simulated by SAM-TOMAS, suggesting even greater new-particle formation at distances less than 5 km than that represented by the model. We have discussed direct emissions of SO3 and HONO as potential sources for the discrepancy between the model and the observations, but we have shown that these do not seem to be necessary for the SAM-TOMAS model to reproduce particle number and size at greater distances from the source.

We have stated in our work that new-particle formation within a given plume may be suppressed close to the source by high NOx concentrations, but this does not mean that new-particle formation will not occur at all. Additionally, the distance from the source at which NOx concentrations fall to levels optimal for OH formation depends on the NOx emissions, the background NOx concentration, and the plume dilution rate. One would expect that in the plume of a power plant with lower NOx emissions under conditions that dilute the plume more quickly, new-particle formation could occur closer to the source, all else being equal (Lonsdale et al., 2012). As power plants continue to lower NOx emissions through controls, we would expect nucleation begin closer to the the source.

Line 450: *"The formation of these particles in the power plant plumes should be properly parametrized to implement power plants more efficiently e.g. in air quality and climate models."*

A parameterization of new-particle formation in power-plant plumes for regional and global models exists, and is freely available. Please see Stevens and Pierce (2013). Fortran 90 code for the parameterization is included as supplementary material to the article. Further, it has been used in the GEOS-Chem model (Stevens and Pierce, 2014).

Posted on behalf of Robin Stevens, Chantelle Lonsdale and Jeff Pierce.

References:

Lonsdale, C. R., Stevens, R. G., Brock, C. A., Makar, P. A., Knipping, E. M., and Pierce, J. R.: The effect of coal-fired power-plant SO2 and NOx control technologies on aerosol nucleation in the source plumes, Atmos. Chem. Phys., 12, 11519-11531, doi:10.5194/acp-12-11519-2012, 2012.

Stevens, R. G., Pierce, J. R., Brock, C. A., Reed, M. K., Crawford, J. H., Holloway, J. S., Ryerson, T. B., Huey, L. G., and Nowak, J. B.: Nucleation and growth of sulfate aerosol in coal-fired power plant plumes: sensitivity to background aerosol and meteorology, Atmos. Chem. Phys., 12, 189-206, doi:10.5194/acp-12-189-2012, 2012.

Stevens, R. G. and Pierce, J. R.: A parameterization of sub-grid particle formation in sulfur-rich plumes for global- and regional-scale models, Atmos. Chem. Phys., 13, 12117-12133, doi:10.5194/acp-13-12117-2013, 2013.

Stevens, R. G. and Pierce, J. R.: The contribution of plume-scale nucleation to global and regional aerosol and CCN concentrations: evaluation and sensitivity to emissions changes, Atmos. Chem. Phys., 14, 13661-13679, doi:10.5194/acp-14-13661-2014, 2014.