Response to the reviewer's comments on the manuscript "New particle formation in the fresh **fl**ue gas plume increase the effective particle number emissions of a coal-**fi**red power plant by Mylläri et al.

May 5, 2016

Dear editor and reviewers,

we thank the reviewers for comments and constructive suggestions. According to these comments, including short comments given by Stevens et al. and Junkermann, we have prepared the response to the comments and revised the manuscript. A response to the reviewers' comments is presented below, including the original comments written in italics.

Sincerely,

Mylläri and co-authors

Response to comments from Reviewer #1

Original comments are presented below in italics.

The results of this work are interesting but major revisions are needed before the manuscript can be published in ACP. The performed measurements and model calculations are not explained adequately but some relevant information is missing. The authors should also make sure that they are correctly citing and interpreting the previous work published on the topic (see the short comment by R. Stevens), and they should discuss uncertainties related to their measurements (see the short comment by W. Junkermann). The language of the manuscript should also be checked. More specific comments are presented below.

We modified the manuscript according to these comments. We added more information regarding experiments and modelling. For these, please see the responses for detailed comments below.

In addition, we took into account the comments from Stevens et al. and Junkermann and made the following changes to the manuscript (underlining indicates new text):

"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 emission* from the stack and *the measurement results show correlation with the model especially at distances of 10-20 km*. Brock et al. (2002) argue the secondary particle formation to begin at 2 hour aged plume."

"In this study the power plant plume diluted to background levels in 2 km (200-400 seconds) which is faster than in other in-flight measurements (Stevens et al., 2012; Junkermann et al., 2011). This difference may be because *the dilution of plume and other processes are affected by source strength, background concentrations, and meteorology (*Stevens et al., 2012*).* In our study, we collected high-time-resolution data close to the power plant stack, which enable us to model the plume dilution on a detailed scale. From this, we were able to observe that while SO_2 and CO_2 were already diluted to

background levels at a distance of 2 km – in agreement with the dilution modeling - the effect of the source to the aerosol number concentration was distinguished at distances >2 km. We attribute this to nucleation taking place in the ageing plume"

"According to modelling results of Stevens et al. (2012), atmospheric new particle formation via coal combustion originated sulphuric acid nucleation begins at *1 km* distance from the source whereas the sulphuric acid formation begins right after emission. Our study therefore *supports this previous modelling work by showing that* 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."

Regarding comments from Junkermann, the diffusion losses of particles were included to the supplementary information and discussed in the main text:

"The data was recorded based on gps-coordinates which were used to calculate distances from the stack, and the distances were changed to correspond plume age using wind speeds 6.5 m s^{-1} and 4.0 m s^{-1} (LIDAR, S3)." Diffusion losses for the particles in the sampling lines were calculated based on the measurement setup. The calculation showed that nearly 70 % of the 2.5 nm particles in diameter was lost in the sampling lines and thus the total concentration shown in Figure 3 can be higher than shown here. The vertical lines denote the 2 km distance from the stack. Figure 3 shows the dilution time scale of the flue gas in terms of CO_2 and SO_2 in both operation conditions. Same trend in SO_2 and N_{tot} concentrations as observed in Table 1, was measured by instruments installed in the helicopter; in ``FGD+FF off'' situation the particle and $SO_2$ concentrations were higher than the ``FGD+FF on'' situation. It should be kept in mind that in ``FGD+FF off'' situation only one of the two flue gas cleaning systems was bypassed."

In addition, flight direction and wind direction data during the measurements were compared. Those can be seen now in Figure 1. The right wind directions were corrected to the manuscript. The date of the measurement and flight time information were included in the manuscript.

Page 1, line 14: Can it really be concluded based on this study that "flue gas nucleation is more efficient than natural atmospheric nucleation"?

We modified the sentence to "In addition, results indicate that flue gas nucleation is more efficient than predicted by using atmospheric aerosol modelling."

Page 2, line 21–23: This statement is too general. For example, SO2 does not affects the climate directly but because it forms sulfuric acid, which affects particle formation.

We modified the sentence according to comment. It is now "In addition to CO_2 emissions, known to have climatic effects, coal combustion causes emissions of other harmful pollutants like NO_x, SO_2, and particulate matter, all decreasing the air quality and increasing health related risks but also affecting climate directly and indirectly. For instance, SO_2 affect the climate indirectly because it tends to oxidize in atmosphere and form H_2SO_4, which affects particle formation."

Page 3, line 66–76: The aims of the study should be expressed more clearly in the end of the introduction.

Similar comment was given also by Reviewer #3. We modified the end of the introduction so that it describes better the aims of the study: "The aim of this study was to characterize how the atmospheric emissions from a 726 MW coal-fired power plant depend on flue gas cleaning, i.e. desulphurization plant and fabric filters (later referred as "FGD+FF off" and "FGD+FF on"). In addition to the stack measurements for pollutants, the study aimed to show how the flue gas cleaning affects real atmospheric concentrations of emitted CO_2, SO_2 and particles. The study included experiments conducted in the stack of the power plant, measurements conducted with a helicopter equipped with instruments for CO_2, SO_2 and particles, and flue gas plume dispersion and aerosol process modelling. "

Following text was removed from the end of the introduction because its information is included in the experimental section: "The power plant has two boilers with separate flue gas cleaning systems. The measurement equipment was installed into two locations; into a helicopter, which enabled the measurement of real flue gas plume in the atmosphere, for the first time, and into the stack. The measurements in the flue gas plume and in the stack were conducted for two different flue gas cleaning situations, i.e. when the flue gas desulphurization unit including fabric filters was in operation (later called as ``FGD+FF on'') and when it was bypassed (later called as ``FGD+FF off'')."

Page 3–4, Sect. 2: The section describing the measurements is missing some relevant information. First of all, it is not told how long the measurement period was (if the measurements were conducted during only one day or several days etc.) In addition, many instruments are mentioned (page 4, line 106–109) without explaining what they measure or what their working principle is. Also, when the weather conditions during the study are described (page 4, line 97–103) it is not explained if the given values for different variables are means for the measurement period or something else.

Unfortunately the original text did not describe the lengths of measurement periods. This information has been written now more clearly in the manuscript. Descriptions of instruments used in the study were modified. Now those include also citations for appropriate publications to guide reader to find more information if needed (LIDAR: Pearson et al., 2009; FPS: Mikkanen et al., 2001; CPC: Agarwal et al., 1980; SMPS: Wang and Flagan 1990; ELPI: Keskinen et al. 1992; EEPS: Mirme 1994). The weather conditions are now given by ranges (min-max) in main text and in one table presented in the supplementary material. Also this is now mentioned in the manuscript.

Page 6, Sect 2.1.1: The section describing how particle formation rates were calculated is unclear. The authors should explain more clearly what they did, and define what they mean with nucleation/formation rates (especially, it is important to know at which size they are determined). Showing the equations used for calculating the formation rates and growth rates would help at understanding the calculations better. If I understand correctly, the authors first calculate Jnuc (at some size smaller than 2.5 nm) by assuming that it depends on sulfuric acid concentration, and then estimate the particle formation rate at 2.5 nm by utilizing a revised version of the so-called Kerminen-Kulmala equation. It is unclear how realistic these calculations are with all the assumptions. Furthermore, it remains unclear if the authors determined particle formation rates only from the parametrization, or also using the measured particle size distributions, which would be very useful (see also the comment related to this on the next page).

The model description has now been clarified in the revised version of the manuscript. Moreover, the assumptions behind the model are discussed extensively in the last paragraph of Section 3.4.

Page 7, line 200: The correct terms are "electrical mobility equivalent diameter" and "aerodynamic equivalent diameter". In addition, it should be specified that the mean diameters are geometric means (as they probably are).

The correct terms were changed to the manuscript and it was specified that they actually are geometric means.

Page 7, line 206: It should be explained more clearly why the difference in the mean diameters indicates that the density is higher than 1 g cm-3.

"Figure 2 shows the particle number size distributions of flue gas in the stack in both cleaning conditions. These were measured using an electrical low pressure impactor (ELPI) and a scanning mobility particle sizer (SMPS) in both ``FGD+FF on/off'' cases. In the ``FGD+FF on'' case the SMPS measurement is a median value over few hours of operation due to low particle number concentrations in the stack. Based on the SMPS measurement the particle geometric mean electrical mobility equivalent diameter was 80 nm and the width of particle number size distribution (geometric standard deviation, GSD) was 1.45 for ``FGD+FF off'' case. In comparison, the geometric mean electrical mobility equivalent diameter was 31 nm for ``FGD+FF on'' and the width of particle number size distribution was 2.15. Based on the measurements using the ELPI geometric mean aerodynamic equivalent diameter was 141 nm and GSD was 1.41. The difference in mean diameters measured using the ELPI and the SMPS comes from the differences in size classification principles of these instruments and enables the determination of effective density of measured particles. The effective density measurement and calculation is based on the relation between the electrical mobility equivalent diameter and the aerodynamic equivalent diameter of the particle (see Ristimäki et al. 2002). In this study case the difference in equivalent diameters indicates effective density larger than unit density for emitted particles (approximately 3.1 g cm^{-3}). In comparison, Saarnio et al. (2014) used effective density of 2.5 g cm^{-3} to convert the electrical mobility diameter measured using SMPS to aerodynamic diameter. *When studying ``FGD+FF on'' case the particle concentrations were so low and thus accurate determination of mean particle size was not possible from the particle size distribution measured by the ELPI.*"

New citation related to the calculation of effective density of particles was included into the manuscript:

Ristimäki et al., 2002 On-line measurement of size distribution and effective density of submicron aerosol particles; Journal of Aerosol Science, vol 33, num 11, pp. 1541-1557.

Page 8, line 209–211: Can it be concluded that the mean aerodynamic diameter and the mean mobility diameter are similar in this case, when there seem to be so few data points from ELPI based on Fig. 2?

This was again very valuable comment. It is true that in "FGD+FF on" case the effective density cannot be determined accurately. The sentence was modified.

Page 9, line 240–242: It is not clear for me where these "closest background values" were obtained and why they were subtracted from the measurements.

The closest background values referred the upwind side (from the stack) measurements between headwind and downwind approaches. We modified the sentence to consider only the mean background

concentration during the flight loops at the upwind side of the stack: “Respectively, the N_{tot} reached nearly to background concentrations after 200 seconds and 300 seconds. The background gaseous concentrations for each measured gaseous component were 403 ppm and 2-8 ppb, for CO_2 and SO_2 respectively. The boundary layer mixing started during the ``FGD+FF on'' measurements and thus the background values measured from the upwind side flight loops from the stack were averaged and subtracted from both ``FGD+FF on/off cases''.”

Page 9, line 249: Can it be concluded that particle concentration reached this high value as the concentration seems to fluctuate very strongly based on Fig. 3?

It is true that the data fluctuated significantly. We modified the text which is now “Based on Figure 3a, for ``FGD+FF off'' case the background particle concentration was 1430 cm-3, after 200 seconds the concentration was at the background level and after 400 seconds it increased significantly, even up to average level of 5 000 cm-3.”

Page 9, line 255: These increases cannot be seen in Fig. 3.

Descriptions regarding to increases were removed from the text.

Page 11, line 266. How these nucleation rates were calculated? If they were calculated using measured particle concentration, authors should explain the method they used. Also, these values should then be compared to the modelled values. It would also be useful to show the time series of nucleation rates; large ranges are now given which do not really tell how high nucleation rate was most of the time.

These formation rates were calculated from mean slope of increasing concentrations.

Page 12, line 278: It is too vague to state that “after that more small particles (and some larger particles are detected”. The authors should specify which particle sizes they mean, and how much more these were observed.

That is true. Taken the measurement uncertainties regarding the measurements by the EEPS into account, we modified the paragraph. It is now “The particle size distribution measurement made by using the EEPS (SI5) supports the results for total particle number measurement made by the CPC (Figure 3), i.e. in terms of particles the flue gas is diluting in 0-300 seconds in ``FGD+FF off''. In addition, the particle size distributions measured by the EEPS indicates slight increase of nanoparticle concentrations during the dilution and dispersion of the flue gas in the atmosphere”.

Page 12, line 301–303: The values of error and R^2 for different cases should be shown for example in a table.

The values for MREs and R^2 are now shown in table form.

Page 12, line 305–306: It would be good to show a figure about SO2 concentration.

Figure about the SO_2 concentrations was added to the supplementary information.

Page 14, line 319–320: The atmospheric concentration of sulfuric acid can vary a lot depending on the environment. Therefore, it would be good if the authors referred here also to some articles reporting sulfuric acid concentration at a site similar to this study.

We included two citations for studies reporting atmospheric sulfuric acid concentrations: Mikkonen et al., (2011, ACP), which report that H_2SO_4 concentrations varied between $1.86 \cdot 10^5$ -$2.94 \cdot 10^6$ molec cm^{-3}, and Sarnela et al., (2015, AE) which reported the sulfuric acid concentrations of 0.38-0.75 ppbv for Finnish industrial and non-industrial area.

Page 14, line 322: Based on Fig. 6, OH concentration seem to be practically zero in the beginning of the simulation. Is that true?

That is not true, for some reason the y-axis was limited from 1e2 onwards. The y-axis scale has now been changed.

Page 15, line 332: To which time periods the mean values presented here correspond? Reporting the mean values of nucleation rate for the whole measurement period is not reasonable.

The mean values are calculated over the whole measurement periods.

Page 15, line 330–340: As mentioned above, the nucleation rate calculations are not explained clearly and therefore it is difficult to follow when the authors mention different nucleation/formation rates. For example, it is not clear for me, if "apparent particle formation rates" calculated from CPC data are presented somewhere in the manuscript. It is also not explained how growth rates were determined. In addition, the authors should refer to some article more relevant for this study than Kulmala et al. (2001) when discussing previous observations on growth and formation rates.

The phrase "apparent particle formation rates" was removed and changed to "Furthermore, in our measurements the particles were detected at the lowest CPC detection limit which was 2.5 nm, $J_{2.5}$." In addition more relevant article was added Stoltzenburg et al., 2005.

Page 15, line 353–354: What if too low values of nucleation rates are due to the incorrect assumptions of the model? The authors should add some discussion about the uncertainties related to different assumptions in their calculations.

The uncertainties in the model calculations are discussed in the last paragraph of Section 3.4.

Page 15, line 357–358: It would be good if authors referred here to the observations of OH concentration made in some environment similar to the site of this study,

Two citations were included: Berresheim et al., (2002 JGR) which reports median value for OH concentration was $0.25 \cdot 10^6$ molec cm^{-3} in Mace Head, and Kuang et al. (2008 JGR) which reports OH concentrations of 10^6-$2 \cdot 10^7$ molec cm^{-3} in Atlanta.

Page 15, line 360: Why NOx concentrations are not shown?

We used in the Gaussian model the NO_x concentrations from the stack measurements. In principle, we think that it wouldn't bring major benefits to show the time series NO_x because it is practically similar than for time series for CO_2.

Page 16, Table 2: It should be stated clearly in the table caption that the results shown there are from a parametrization. In addition, it is not clear for me what is meant by "(1 cm-3 (600 s)-1)".

The caption was modified. The values in the table were recalculated to $cm^{-3}s^{-1}$

Page 16, line 366: The authors could refer here also to some articles discussing the role of other compounds besides sulfuric acid in atmospheric particle formation.

We refer now on Ehn et al. (2012) related to role of ELVOCs in atmospheric particle formation, and on the study of Arnold et al. (2012), which reports sulphuric acid concentrations for combustion engine engine exhaust but also indications on organosulfates in particle formation:

Ehn, M., Kleist, E., Junninen, H., Petäjä, T., Lönn, G., Schobesberger, S., Dal Maso, M., Trimborn, A., Kulmala, M., Worsnop, D.R., Wahner, A., Wildt, J., Mentel, T.F. Gas phase formation of extremely oxidized pinene reaction products in chamber and ambient air (2012) Atmospheric Chemistry and Physics, 12 (11), pp. 5113-5127.

Arnold, F., Pirjola, L., Rönkkö, T., Reichl, U., Schlager, H., Lähde, T., Heikkilä, J., Keskinen, J. First online measurements of sulfuric acid gas in modern heavy-duty diesel engine exhaust: Implications for nanoparticle formation (2012) Environmental Science and Technology, 46 (20), pp. 11227-11234.

Page 17–18, line 408–460: The conclusions of the study about the climate effects of the emissions of a coal-fired power plant should be stated here in a more concise and clear way. Now the conclusions remain rather vague.

The aim of the conclusions and discussions regarding climatic effects was to link the results with larger topics. Based on this comment and nearly similar comment from Reviever #3, we decided to remove the last paragraphs of Discussion and Conclusions sections. In addition, we modified the conclusion also based on short comments from Stevens at al. and Junkermann et al.

Page 1, line l4: This sentence should be rephrased.

We modified the sentence to ”In addition, results indicate that flue gas nucleation is more efficient than predicted by using atmospheric aerosol modelling.”

Page 5, Fig. 1: It should be explained in the figure caption what different colors (blue and black) mean.

The caption was rephrased: “Helicopter flight routes. The wind blew in the angle of 216°±5.51° (based on LIDAR data) and the flight direction was 213°±4.14° (based on GPS data for helicopter) in ``FGD+FF off'' (blue circles). Corresponding angles for ``FGD+FF on'' case (black circles) were 220°±6.25° (wind direction based on LIDAR data) and 223°±5.66° (flight direction based GPS data for helicopter). The triangular shapes (black and blue lines) show the helicopter GPS coordinates that have been taken into account in the calculations.”

Page 7, line 172: The equations should be numbered and shown on their own lines (also elsewhere in the manuscript).

These changes have been made like proposed.

Page 9, line 223: This sentence should be rephrased.

The sentence was changes to: “Figure 3 shows the measured flue gas plume concentrations as a function of plume age.”

Page 10, Figure 3: The figure is unclear. It is difficult to see different line as they are on top of each other. It might also be a good idea to present at least particle concentration data using a logarithmic scale. On the line 247 authors refer to Fig. 3a, but there are no “a” and “b” marked in the figure. It would also be good to mention in the figure caption or in the text which instrument was used for measuring Ntot shown in the figure.

The figure has been improved so that data series can be better compared with each other. The scaling of the axis is still linear (no concentration differences of several magnitudes) but the N_{tot} curve is in a different position compared to CO_2 and SO_2. Labels a) and b) were added and the measurement method for N_{tot} was added to the caption.

Page 11, Figure 4: The color bar is missing the label.

The color bar label was added.

Page 12, line 272: “changing in same ages” should be rephrased.

The sentence was removed.

Page 12, line 274–276: The sentence starting with “Figure 4...” is unclear.

The sentence was removed.

Page 12, Fig. 5: This figure is unclear due to errorbars, and it is difficult to separate red lines from each other. In addition, it is not explained in the figure caption which lines represent which stability classes.

The Figure 5 was modified according to the comment: errorbars were removed and legend was modified to show stability classes.

Page 16, line 370: “this studied power plant” should be rephrased.

The sentence was modified to “Each power plant (over 50 MW) in EU has emission limits for SO_2, NO_2, and particle mass concentrations, for the studied power plant the limits are 600 mg Nm^{-3} (210 ppm), 600 mg Nm^{-3} (290 ppm), and 50 mg Nm^{-3}, respectively.”

Page 16, line 371: “Table 1 results with” should be rephrased.

The sentence was modified to “Comparison of the results in Table 1 with these emission limits shows that the emissions were clearly below these limits when the power plant operation was normal (``FGD+FF on'').”

Page 16, line 375: It is not clear for me what is meant by “the amount of H2SO4 of new aerosol particles”

The sentence was modified to “In addition to primary emissions, flue gas cleaning systems seemingly affect also the compounds which can act as precursors for new particles; e. g. SO_2 tends to oxidize in the atmosphere to SO_3 and, further, to form H_2SO_4 which can nucleate or condensate to particle phase.”

Page 17, line 399–400: This sentence is unclear and it should be rephrased.

The sentence was modified to "For instance, rough estimates for particle number emission factors can be calculated by comparing the measured particle number concentration with the simultaneously measured CO2 concentration of the flue gas plume (see e.g. Saari et al. 2016). By utilizing this method, for particles existing in the flue gas plume in ages of 25–55 seconds the emission factor in respect of CO_2 was $2.0 \cdot 10^{10}$ $(g\ CO_2)^{-1}$ and in ages over 400 seconds $8 \cdot 10^{10}$ $(g\ CO_2)^{-1}$ in the "FGD+FF off" case. Similarly, in the "FGD+FF on" case the emission factors were $4 \cdot 10^{9}$ $(g\ CO_2)^{-1}$ (for aerosol dispersed 55-85 seconds in the atmosphere) and $3.74 \cdot 10^{10}$ $(g\ CO_2)^{-1}$ (for aerosol dispersed more than 500 seconds in the atmosphere). "

One reference was added: Saari, S., Karjalainen, P., Ntziachristos, L., Pirjola, L., Matilainen, P., Keskinen, J., Rönkkö, T. (2016) Exhaust particle and NOx emission performance of an SCR heavy duty truck operating in real-world conditions. Atmospheric Environment, 126, pp. 136-144, 10.1016/j.atmosenv.2015.11.047.

Response to comments from Reviewer #3

Overall, the major problem I see with this paper is lack of discussion on uncertainties related to their measurements (see detailed short comment by W. Junkermann). I urge authors to compare and contrast the results of their work and those presented in Junkermann et al. (2011, 2015) and others.

We have taken into account the comments from W. Junkermann in our revised manuscript. This includes adding the date of the measurement, wind direction from LIDAR, helicopter flight directions and sampling losses for particles in helicopter measurements into the manuscript. In addition, we checked that the manuscript text includes relevant information on the question related to semi-volatile particles in stack measurements.

Comparison of the results of our study with the results reported by Junkermann et al. cannot be made very straightforward because their study is actually very different when compared with our study. This is e.g. due to the lack of information shared regarding emission reduction systems used in their study, different time scales studied and different particle size ranges covered. However, we see that our study and studies reported by Junkermann et al. fulfills quite efficiently each other. Junkermann et al. (2015) studied "Size distributions measured within the centre of the first (12 km) and last (130 km) plume crossing" while our study focused on particles very near the source. Thus we made comparisons at very general level; e.g. Junkermann et al. (2011) reports that "our results show that flue cleaning technology actually introduces a new problem, in the form of increased emissions of ultrafine particles". Role of ultrafine particles is topic also in our study. In addition, Junkermann et al 2015 reports that the particle size increases as a function of plume ageing in the atmosphere. This has connection with our study, where new particle formation was measured in the flue gas plume.

Page 1, Line 9-10: What makes you think that these are actually newly formed particles in the flue gas plume when you are not measuring sub-3nm particles? New particles are those closer to 1 or 1.5 nm in

diameter, smaller than most instruments can measure (e.g. PSM can measure particles of 1 nm diameter which was not operated in this study). Please justify this?

Good comment. We modified the abstract according to this comment. “However, the atmospheric measurements indicated that new particles larger than 2.5 nm are formed in the flue gas plume, even in the very early phases of atmospheric ageing.” Unfortunately we did not have the PSM in our measurement setup.

Page 1, Line 13: There is no sufficient evidence to conclude "flue gas nucleation is more efficient than natural atmospheric nucleation" and also, it contradict to what authors discuss on page 14, line 333-336.

This similar comment was also given by Reviewer #1. We modified the sentence.

Page 3, Line 67-74: "The power plant has two boilers with separate ----------bypassed (later called as “FGD+FF off”). This all should moved to Experimental section, and instead discuss here measurements planned and major goals of this study.

This similar comment was also given by Reviewer #1. We moved the sentences to experimental section and modified the end of the introduction more to discuss the aims of this study.

Page 4, Line 96: What do you mean by weather conditions were quite stable? I would like to see time series or table for detailed weather conditions during the measurement time period, if available at all? You can include it in supplementary material.

We meant by stable weather conditions that variations in wind direction and wind speed, temperature, radiation and visibility were small. Those variations were included in the text already in the original manuscript; however, we added an additional table to the supplementary. Finally, we removed “quite” from the sentence.

Table 1/2: In both the tables, I would like to see nucleation mode particles in the particle size range of 2.5 nm to 15 nm diameter alongside total number concentration (as shown by Ntot). I believe you report Ntot from CPC measurements and SMPS calculated Ntot will give Ntot>15nm so CPC Ntot minus SMPS Ntot would probably give you rough estimate of nucleation mode particles.

Unfortunately, our measurement data does not allow this kind of analysis. Our measurement setup in the stack was designed so that we could study the non-volatile particles in the flue gas. Thus we do not have data from stack measurements related to nucleation mode particles.

Fig. 3: This figure needs to be improved significantly. The font size for all axes is too small. Why delta Ntot shown? It makes more sense to show Ntot. It is difficult to differentiate the background lines for SO2 and CO2, simply remove it and state the background values in the figure caption, that should be enough. Include labels "a" and "b" as you used in the text. Also include nucleation mode particles (2.5-15 nm diameter range) as a function of plume age as suggested above.

Thank you for your very good comment. This was pointed out also by Reviewer #1. We have modified the figure like you and Reviewer #1 proposed. ΔN_{tot} is shown the net effect of the particle formation. Regarding to nucleation mode particles (proposed particle size range 2.5-15 nm diameter range), please see our response on previous comments. In addition, the calculation based on CPC and EEPS data increases significantly uncertainties thus decreases the quality of data so much that we would not like to present it this paper.

Page 11, Line 265-279: Authors state that "EEPS particle size distribution data is noisy, can show maximum of 67% wrong compared to SMPS, and also can not be compared with CPC data" If that is so then Fig. 4 should be removed or at least moved to supplementary for readers and discuss in the main text very briefly.

The figure was moved to Supplementary Information.

Page 14, Line 330-331: " According to the scheme applied here", be specific and give details of scheme applied here or in the methods section.

One equation was added to the manuscript from Lehtinen et al. (2007). The sentence was modified " According to the scheme applied here (see equations 4 and 5)…"

Page 16, Line 386: I do not agree with authors stating that " but this study shows that nucleation can take place in lower SO2 concentrations." From Table 1 and figure 3, the SO2 concentrations were much higher than a ppb level and with 10^5-10^6 cm^-3 OH concentrations and lower CS, it should form sufficient H2SO4 for particle nucleation to occur.

Good comment. The part has been modified based on the short comment by Stevens et al. and the sentence was removed. The nucleation indeed does take place (see Fig.5 in the revised manuscript). However, according to the model calculations, practically all of the nucleated particles (having an initial size of 1.5 nm), did not reach detectable sizes due to coagulational scavenging.

Page 1 : Suggest revising the title to "New particle formation in the fresh flue gas plume from a coal-fired power plant in Helsinki increases the effective particle number concentrations".

We think that the location of the power plant is not important; however the emphasis of the title should be in the flue gas cleaning technologies which are, in our opinion, far more important in terms of the emissions. Based on this we modified the title: "New particle formation in the fresh flue gas plume from a coal-fired power plant: effect of *flue gas cleaning"*

Page 1, Line 5: Replace "nucleation particle precursor formation" with "particle formation precursors"

Replacement was made.

Page 2, Line 23: replace "particles" by "particulate matter".

Replacement was made.

Page 2, Line 25-27: " However, with proper combustion and flue gas cleaning ---------- than CO2 can be decreased", support this statement by reference

The references were added (Helble, 2000; Saarnio et al., 2014), they were missing from the text but were present at the references list.

Page 3, Line 100-101: " The background aerosol concentrations for ---------- were: CO2 403 ppm, SO2 less than 2-8 ppb." Do you mean to say the background gaseous concentrations were?

The sentence was corrected to consider "the background gaseous concentrations".

Page 4, Line 107: correct as "TSI Inc."

This was corrected.

Page 4, Line 115-116: Particle size distribution was measured in what size range?

Particle size range was added "… from 5.6 nm to 560 nm".

Page 10, Line 236: I believe, the atmospheric background SO2 concentration is much lower than 25 ppb.

Good comment. We checked the background SO_2 concentration and corrected the value into the manuscript to be "2-8 ppb". These values are from helicopter flight upwind the stack.

Fig.5: replace " (above) and 'c' and 'd' (below)," by " (top panel) and 'c' and 'd' (bottom panel)," if that is what you meant to say.

Corrections were made as suggested.

Page 7, Line 182: Authors state that "background concentration of NOx is practically zero". It should be justified.

The sentence was changed to "To support this, the study of Pirjola et al. (2014) indicates that in the harbour area close to the power plant studied here the NO_x concentration level is typically clearly lower than 100 ppb."

Page 13, Line 323: I think, the atmospheric background value of CS is slightly lower than you report, usually of the order or 10^-3 s^-1.

This value was calculated based on the SMEAR III station data and it is at the ground level, thus the CS given in our manuscript might be an overestimation of the real CS at the stack height in the atmosphere.

Fig. 6: figure caption needs to be improved e.g. time development of [H2SO4] (blue line), ...The OH concentration seems to be zero or lower than 10^2 initially, please re-scale y-axis

We agree. Y-scale of the figure was changed.

J25 refers to particle formation rate at particle size of 2.5 nm, right? if so then replace J25 to $J_{2.5}$.

It was replaced.

Page 15, Line 352: it should read as - they are "relatively" low. It is very difficult to compare these values with literature values as you do not mention the time of measurement (see specific comment)

Information about time of flights and the date were added to the manuscript. “The measurements were made 24.3.2014 in two separate one hour periods (see specific times from S2, the black rectangulars; the first illustrates “FGD+FF on” case and the latter “FGD+FF off” case). “

Page 16, Line 407: "Based on that knowledge, it can be assumed that the formed particles are more scattering than absorbing." can we really conclude this based on results presented here?

We significantly modified the discussion and conclusion sections regarding to this comment (removed most of text). Nearly similar comment was given also by Reviewer #1. The aim of these texts was to bring larger perspective into the paper.

Page 16: the last paragraph of 3.5 discussion section is not in light of results presented in this work. e.g. in countries like China and India, there are other major sources e.g. transportation which contribute significantly to degraded air quality (total particulate matter). I would suggest to remove this paragraph completely and discuss your results in light of previous studies on particle formation in power plant plume rather than generalizing way beyond the scope of this study.

The paragraph was removed.

Page 18: The last paragraph of conclusion section mostly like introducing the topic, instead it should state what are the implications of this study.

The paragraph was removed.

Finally, list of new references included in the manuscript:

Agarwal, J. K. ja Sem, G. J. (1980). Continuous Flow, Single-particle Counting Condensation Nucleus Counter. *Journal of Aerosol Science*, (11), 343–357.

Arnold, F., Pirjola, L., Rönkkö, T., Reichl, U., Schlager, H., Lähde, T., Heikkilä, J., Keskinen, J. First online measurements of sulfuric acid gas in modern heavy-duty diesel engine exhaust: Implications for nanoparticle formation (2012) Environmental Science and Technology, 46 (20), pp. 11227-11234.

H. Berresheim, T. Elste, H. G. Tremmel, A. G. Allen, H.-C. Hansson, K. Rosman, M. Dal Maso, J. M. Mäkelä, M. Kulmala. 2002. Gas-Aerosol relationships of H_2SO_4, MSA, and OH: Observations in the coastal marine boundary layer at Mace Head, Ireland. Journal of Geophysical Research, vol 107, no. D19, 8100, doi:10.1029/2000JD000229

Ehn, M., Kleist, E., Junninen, H., Petäjä, T., Lönn, G., Schobesberger, S., Dal Maso, M., Trimborn, A., Kulmala, M., Worsnop, D.R., Wahner, A., Wildt, J., Mentel, T.F. Gas phase formation of extremely

oxidized pinene reaction products in chamber and ambient air (2012) Atmospheric Chemistry and Physics, 12 (11), pp. 5113-5127.

J.J. Helble. A model for the air emissions of trace metallic elements from coal combustors equipped with electrostatic precipitators, Fuel Processing Technology, 2000, 63, 125-147.

Keskinen, J., Pietarinen, K. and Lehtimäki, M. (1992) Electrical Low Pressure Impactor, J. Aerosol Sci. 23, 353-360.

C. Kuang, P. H. McMurry, A. V. McCormick, F. L. Eisele. 2008. Dependence of nucleation rates on sulphuric acid vapor concentration in diverse atmospheric locations. Journal of Geophysical Research, vol. 113, D10209, doi:10.1029/2007JD009253.

Mikkanen, P., Moisio, M., Keskinen, J., Ristimäki, J., Marjamäki, M. *Sampling method for particle measurements of vehicle exhaust*. SAE Technical paper (2001), 2001-01-0219.

Mirme, A.. *Electric aerosol spectrometry*. Ph. D. Thesis, Tartu University, 1994.

Pearson, G., Davies, F., Collier, C. An analysis of the Performance of the UFAM Pulsed Doppler Lidar for observing the Boundary Layer. J. Atmos. Ocean. Tech., 26, 240–250, doi:10.1175/2008JTECHA1128.1, 2009

J. Ristimäki, A. Virtanen, M. Marjamäki, A. Rostedt, J. Keskinen. 2002 On-line measurement of size distribution and effective density of submicron aerosol particles; Journal of Aerosol Science, vol 33, num 11, pp. 1541-1557.

Saari, S., Karjalainen, P., Ntziachristos, L., Pirjola, L., Matilainen, P., Keskinen, J., Rönkkö, T. (2016) Exhaust particle and NO_x emission performance of an SCR heavy duty truck operating in real-world conditions. Atmospheric Environment, 126, pp. 136-144, 10.1016/j.atmosenv.2015.11.047.

N. Sarnela, T. Jokinen, T. Nieminen, K. Lehtipalo, H. Junninen, J. Kangasluoma, J. Hakala, R. Taipale, S. Schobesberger, M. Sipilä, K. Larnimaa, H. Westerholm, J. heijari, V.-M. Kerminen, T. Petäjä, M. Kulmala. 2015. Sulphuric acid and aerosol particle production in the vicinity of an oil refinery. Atmospheric Environment, 119, pp. 156--166.

K. Saarnio, A. Frey, J.V. Niemi, H. Timonen, T. Rönkkö, P. Karjalainen, M. Vestenius, K. Teinilä, L. Pirjola, V. Niemelä, J. Keskinen, A. Häyrinen, R. Hillamo. 2014 Chemical composition and size of particles in emissions of coal-fired power plant with flue gas desulphurization, Journal of Aerosol Science, vol 73, 14-26.

Stolzenburg, M. R., P. H. McMurry, H. Sakurai, J. N. Smith, R. L. Mauldin III, F. L. Eisele, and C. F. Clement. Growth rates of freshly nucleated atmospheric particles in Atlanta, J. Geophys. Res., 2005, 110, D22S05, doi:10.1029/2005JD005935.

Wang, S. C. and Flagan, R. C. (1990). Scanning Electrical Mobility Spectrometer. *Aerosol Science and Technology*, (13), 230–240.