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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Primary versus secondary contributions to particle number concentrations in the European boundary layer" *by* C. L. Reddington et al.

C. L. Reddington et al.

c.reddington@see.leeds.ac.uk

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Whilst reviewing the manuscript we discovered a bug in our model output analysis code, which has some impact on the statistical calculations and results of the study. In addition, the TNO (Cabauw) SMPS data presented in the paper are not corrected for (diffusional) losses in the inlet system and SMPS system itself. Below we present the necessary changes to the manuscript that result from the corrected analysis code and Cabauw SMPS data. The updated figures and tables can be viewed in the supplement. All values quoted in the manuscript that correspond to the corrected tables/figures have been updated accordingly. Any changes to the wording of the text as a result of the

corrections have been detailed below. The original text from the manuscript is shown in italic font and the corrected text is shown in regular font. All figure, table and section numbers refer to those used in the manuscript.

The bug in the analysis code resulted in an underestimation of the modelled mass (and volume) of the sea salt (SS) component and overestimation of the modelled mass (and volume) of the organic carbon (OC) and black carbon (BC) components. The errors in the calculated SS, OC and BC component volumes affects the calculation of the dry size of the aerosol distributions, and therefore impacts the shape of the modelled particle number and mass size distributions. There is no impact on the modelled total particle number concentration, but number concentrations in specified size ranges e.g. larger than 50 nm or 100 nm are altered when corrected for the bug. For more details on the corrections applied to the TNO SMPS data, please see Henzing (2011).

It is important to note that these changes do not alter the main conclusions of our study (listed below):

- The model shows good agreement with observed CCN-sized particle number concentrations at the surface (N_{100}) and aloft in the BL (N_{160}).
- The agreement between modelled and observed N_{50} is good if we assume a small emission size for primary carbonaceous aerosol.
- When we emit larger (more realistic) particles the model underpredicts the range of N_{50} observed. The underprediction is partly (but not entirely) compensated by including BL nucleation.
- The model underpredicts particle number concentrations $<$50 nm unless a BL nucleation mechanism is included.
- Fairly poor temporal agreement between model and observations (on hourly scale) precludes any attempt to identify the best nucleation mechanism from such a short dataset.

P18257, L23–25: *"At Cabauw, measurements below 30 nm were affected by noise, so for this location we restrict our analysis to D_p>30 nm. In addition, measurements at Cabauw have not been corrected for diffusional losses."*
The above text no longer applies to the corrected Cabauw SMPS data and has been removed. We also remove "(D_p>30 nm at Cabauw)" from L26.

P18270, L8–11: *"For example, the mean modelled N_{50} increases by ~80 % in the European BL between the BCOC_lg and BCOC_sm experiments, while N_{100} and N_{160} increase by ~40 % and ~20 %, respectively."*
The above sentence has been modified to the following to include the corrected percentage increases: "For example, the mean modelled N_{50} increases by ~60 % in the European BL between the BCOC_lg and BCOC_sm experiments, while N_{100} and N_{160} increase by ~45 % and ~20 %, respectively."

P18270, L21–23: *"For N_{160}, the model is biased slightly high in experiment BCOC_sm (NMB = 20 %, $m = 0.83$) and, in contrast to comparisons with observed N_{100}, N_{50} and N_{tot}, the agreement is improved in experiment BCOC_lg (NMB = 9 %, $m = 0.92$)."*
The text has been replaced with the following to account for the changes in the statistical results (see Table 3): "For N_{160}, the model bias is small in experiment BCOC_sm (NMB = 9 %, $m = 0.65$), but in contrast to comparisons with observed N_{100}, N_{50} and N_{tot}, we find the best agreement with observed N_{160} over the IOP is with the BCOC_lg experiment (NMB = -1 %, $m = 0.74$).

P18270, L24–L26: *"Including BL nucleation increases simulated N_{50} and N_{100} over Europe by ~10–50 % and ~5–20 %, respectively, depending on the mechanism and on the emission size of BC+OC particles."*
The above sentence has been altered to the following to include the corrected percentage increases: "Including BL nucleation increases simulated N_{50} and N_{100} over

Europe by ~10–40 % and ~5–20 %, respectively, depending on the mechanism and on the emission size of BC+OC particles."

P18270, L28–P18271, L4: *"In the BCOC_sm experiment, the impact of including BL nucleation depends on the mechanism and the particle size range; for N_{50} the bias is smaller with the ACT and ORG2 mechanisms, but for N_{100} all nucleation mechanisms lead to a larger model bias. Since the model without BL nucleation slightly overpredicts the mean N_{160}, including BL nucleation only acts to increase the model bias in this size range."*
We have replaced the above text with the following to account for the changes in the statistical results (see Table 3): "In the BCOC_sm experiment, the bias in N_{50} is reduced by including BL nucleation; the smallest bias is achieved with the KIN and ORG1 mechanisms (-11 %). However, for N_{100} all nucleation mechanisms lead to a slightly larger model bias (although the NMB remains smaller than 10 %). The impact of BL nucleation on N_{160} is fairly negligible (increasing mean concentrations over Europe by less than 1 %), resulting in small changes in the model bias in this size range."

P18271, L5–7: We have replaced the following *"The slope of the linear regression and correlation coefficient between simulated and observed multi-site campaign-mean N_{50}, N_{100}, and N_{160} are not improved with BL nucleation."* with "When BL nucleation is included, there is little improvement (if any) in the slope of the linear regression and correlation coefficient between simulated and observed multi-site campaign-mean N_{50}, N_{100}, and N_{160}."

P18272, L6–8: *"Figure 4a shows that at 12 of the 15 sites the NMB_{hourly} is fairly large and negative (BCOC_lg, range -99– -84 %; BCOC_sm, range -87– -50 %)."*
The statistical values in the sentence above have been corrected to the following: "Figure 4a shows that at 12 of the 15 sites the NMB_{hourly} is fairly large and negative (BCOC_lg, range -98– -83 %; BCOC_sm, range -77– -33 %)."

P18272, L13–15: *"When some form of BL nucleation is included, the model-observation difference becomes insignificant at 7 sites, showing that, statistically, nucleation is an important process affecting $N_{<50}$ at at least half of the ground sites."*
The above sentence has been modified to the following: "When some form of BL nucleation is included, the model-observation difference becomes insignificant at 6 sites, showing that, statistically, nucleation is an important process affecting $N_{<50}$ at at least 40% of the ground sites."

P18272, L15–P18273, L4: *"For N_{50}, the model-observation difference is statistically insignificant at 7 of the 15 sites without BL nucleation. It is mostly the BCOC_sm experiment that captures the observations at these sites, apart from at Jungfraujoch where the model-observation difference is only insignificant when we assume large primary BC+OC particles. At these sites, the NMB$_{\text{hourly}}$ is very small (range -4 to 5 %), but at the remaining 8 sites with a significant difference the model bias is still fairly small (Fig. 4b): for 7 of the 8 sites, the bias is smallest with the BCOC_sm experiment (between -36 % and 15 %), the exception being Finokalia where the bias is smallest with the BCOC_lg experiment (-23 %).*
When some form of BL nucleation is included, the model-observation difference in N_{50} becomes insignificant at 3 out of the 8 remaining sites (Finokalia, Hyytiälä, and Mace Head). For these sites, BL nucleation makes an important contribution to N_{50}. For 6 sites where the difference was insignificant with experiments BCOC_sm and BCOC_lg, including BL nucleation increases the model bias, but at the 99% confidence level the model-observation difference remains statistically insignificant. Overall, with BL nucleation, the difference between modelled and observed N_{50} is insignificant at two thirds of the ground sites. Thus, the model with BL nucleation is in better agreement with the observations than the model without BL nucleation."
The results of the significance tests in the above paragraph have been corrected and the text has been modified to the following: "For N_{50}, the model-observation difference is statistically insignificant at 3 of the 15 sites (Jungfraujoch, Melpitz and Cabauw) without BL nucleation. It is the BCOC_sm experiment that captures the observations

at these 3 sites, the observations at Jungfraujoch are also captured with the BCOC_lg experiment. At the sites where the model-observation difference is insignificant, the NMB$_{\text{hourly}}$ is very small (range -7 to 5 %). But at the remaining 12 sites with a significant difference the model bias is still fairly small (Fig. 4b): for all 12 sites the bias is smallest with the BCOC_sm experiment (between -43 % and 21 %).
When some form of BL nucleation is included, the model-observation difference in N_{50} becomes insignificant at 7 out of the 12 remaining sites. For these sites, BL nucleation makes an important contribution to N_{50}. At Jungfraujoch (where the difference was insignificant with experiments BCOC_sm and BCOC_lg), including BL nucleation increases the model bias, but at the 99% confidence level the model-observation difference remains statistically insignificant. Overall, with BL nucleation, the difference between modelled and observed N_{50} is insignificant at two thirds of the ground sites. Thus, the model with BL nucleation is in better agreement with the observations than the model without BL nucleation."

P18273, L4–6: The following sentence has been removed from the text: *"However, if we consider individual sites the agreement between model and observations deteriorates slightly at some locations with BL nucleation and the model bias increases."*

P18273, L7–21: *"For N_{100}, we find that at 10 sites there is a statistically significant difference between the model and observations in experiments without BL nucleation. Although this is a higher proportion of sites than for N_{50}, at the sites where the difference is significant the NMB$_{\text{hourly}}$ is generally smaller for N_{100}. For 5 sites the bias is smallest with the BCOC_sm experiment (between -16 % and 14 %), and for 4 sites the bias is smallest with the BCOC_lg experiment (between -18 % and 9 %). At 1 site (Jungfraujoch), there is a large negative bias with both model experiments (BCOC_sm, -69 %; BCOC_lg, -80 %).*
When BL nucleation is included, the model-observation difference in N_{100} is no longer significant at an additional 3 sites (Hyytiälä, Vavihill and Melpitz). However, at 3 of the 5 sites where the difference was insignificant with experiments BCOC_sm and BCOC_lg

(Schauinsland, Aspvreten and Mace Head), adding BL nucleation results in overprediction of N_{100} and the model-observation difference becomes significant. In total, the model with BL nucleation is able to capture the observations at one third of the ground sites."

The results of the significance tests in the above paragraph have been corrected and the text has been modified to the following: "For N_{100}, we find that at 12 sites there is a statistically significant difference between the model and observations in experiments without BL nucleation. At the 3 sites where model-observation difference is statistically insignificant, again it is the BCOC_sm experiment that captures the observations. This is the same proportion of sites as for N_{50}, but at the sites where the difference is significant the NMB_{hourly} is generally smaller for N_{100}. For 9 sites the bias is smallest with the BCOC_sm experiment (between -19 % and 18 %), and for 2 sites the bias is smallest with the BCOC_lg experiment (-32 % at Cabauw and 9 % at Finokalia). At 1 site (Jungfraujoch), there is a large negative bias with both model experiments (BCOC_sm, -69 %; BCOC_lg, -81 %).

When BL nucleation is included, the model-observation difference in N_{100} is no longer significant at an additional 4 sites (Hyytiälä, Vavihill, Monte Cimone and Aspvreten). However, at 1 of the 3 sites where the difference was insignificant with experiment BCOC_sm (Schauinsland), adding BL nucleation results in an overprediction of N_{100} and the model-observation difference becomes significant. In total, the model with BL nucleation is able to capture the observations at almost half of the ground sites."

P18274, L2–11: *"It is more difficult to draw conclusions about the contribution of BL nucleation to N_{50} and N_{100} because of the limited number of sites where nucleation is needed to explain significant model-observation differences (3 out of 15 sites). In addition, when we take into account the ± 10 % uncertainty of the measurements (Wiedensohler et al., 2010), N_{100} can be explained at all 3 of these sites without the need for BL nucleation. In total, the difference between the model (without BL nucleation) and observations (± 10 %) is statistically insignificant at 10 sites for N_{50} and 11 sites for N_{100}. Including BL nucleation in the model, the observations can be captured within*

± 10 % at an additional 2 sites for N_{50} (Finokalia and Hyytiälä) and an additional 3 sites for N_{100} (Hohenpeissenberg, Ispra and Košetice)."

We have replaced the paragraph above with the following to be consistent with the corrections made to the results of the significance tests: "The results of the t-tests show that the model with BL nucleation also gives the best overall agreement with observations of N_{50} and N_{100}, capturing the observations at two thirds and almost half the sites respectively. However, if we take into account the ± 10 % uncertainty of the S/DMPS measurements (Wiedensohler et al., 2010), N_{50} and N_{100} can be explained at almost all of these sites without the need for BL nucleation. In total, the difference between the model without BL nucleation and observations (± 10 %) is statistically insignificant at 8 sites for N_{50} and 10 sites for N_{100}. Including BL nucleation in the model, the observations can be captured within ± 10 % at an additional 4 sites for N_{50} (Hyytiälä, Mace Head, Vavihill, Schauinsland) and an additional 2 sites for N_{100} (Hohenpeissenberg and Košetice). Therefore at the majority of ground sites, it is difficult to detect the contribution of BL nucleation to N_{50} and N_{100} within the uncertainty of the observations."

P18274, L21–P18275, L8: *"The dependence of the best-fit assumption of BC+OC particle emission size on site location can be seen clearly. At Finokalia and Jungfraujoch, the range of observed concentrations is captured best with larger primary BC+OC particles, with distribution overlap values of 55 % and 78 %, respectively. But at all other sites, the BCOC_lg experiment not only underpredicts N_{50}, but also underpredicts the range of concentrations observed (average overlap of 43 %). The range of observed N_{50} is captured much better at most sites when smaller BC+OC particles are emitted in the model (average overlap of 66 %).*

Including BL nucleation increases the range of simulated N_{50} in experiment BCOC_lg and improves the agreement between modelled and observed distributions (average overlap of 53–58 %, versus a mean of 43 %). At 6 sites the distribution overlap becomes equal to or greater than experiment BCOC_sm. The impact of BL nucleation is fairly small on the range of N_{50} predicted by experiment BCOC_sm, and at roughly two-thirds of the sites the distribution overlap is decreased slightly (average 62–64 %,

depending on the mechanism). At 8 sites, the range of observed N_{50} is captured best with experiment BCOC_sm (with or without BL nucleation)."
The paragraphs above have been replaced with the following to account for small changes in the distribution overlap percentages (see also corrected Fig. 6): "Figure 6 shows there is some dependence of the best-fit assumption of BC+OC particle emission size on site location. For example, at Jungfraujoch the range of observed concentrations is captured best when larger primary BC+OC particles are emitted, with a distribution overlap of 78 %. But at all other sites, the BCOC_lg experiment not only underpredicts N_{50}, but also underpredicts the range of concentrations observed (average overlap of 42 %). The range of observed N_{50} is captured much better at most sites when smaller BC+OC particles are emitted in the model (average overlap of 67 %).
Including BL nucleation increases the range of simulated N_{50} in experiment BCOC_lg and improves the agreement between modelled and observed frequency distributions (average overlap of 53–56 %, versus 42 % without BL nucleation). The impact of BL nucleation is fairly small on the range of N_{50} predicted by experiment BCOC_sm (average overlap of 67–68 % versus 67 % without BL nucleation), and at 9 sites the distribution overlap is decreased slightly. At Finokalia and Hyytiälä the distribution overlap becomes greater in experiment BCOC_lg than in BCOC_sm. However, at two thirds of the sites, the range of observed N_{50} is captured best with experiment BCOC_sm (with or without BL nucleation)."

P18277, L21–26: ''*The impact of BL nucleation on number concentrations at the large end of the size distribution is relatively small, with an average increase of ~ 2 % in the mean simulated $N_{160-1040}$ for each flight. But in general, the overall agreement between mean modelled and observed $N_{160-1040}$ is improved with BL nucleation, particularly in the BCOC_lg experiment. This can be interpreted as a decreasing influence of primary emissions aloft in the BL compared with observations at the surface."*

Alterations have been made to the paragraph above to account for the changes in the modelled particle number concentrations $>$160 nm: "The impact of BL nucleation on number concentrations at the large end of the size distribution is relatively small, with an average change in the mean simulated $N_{160-1040}$ for each flight of $\sim$2 %. But in the BCOC_lg experiment, the overall agreement between mean modelled and observed $N_{160-1040}$ is generally improved with BL nucleation. This can be interpreted as a decreasing influence of primary emissions aloft in the BL compared with observations at the surface. For number concentrations in this size range the BCOC_lg experiment gives slightly better agreement with the aircraft observations which is consistent with comparisons with the ground-based observations."

P18280, L25–P18281, L5: *"However, because of the large variation in the observed number concentrations in this size range, we find that the differences between the simulated and observed means are not statistically significant at the 99 % confidence level. The observations lie between the two size distributions predicted by the model suggesting the measured non-volatile particle size distribution does not only consist of BC, but is likely to include contributions from non-volatile organic matter, in addition to contributions from sea salt particles (Jennings and O'Dowd, 1990; O'Dowd and Smith, 1993) and mineral dust. The latter two species have not been included in the modelled non-volatile particle number concentration, but are likely only to make substantial contributions in the super-micron size range."*
The paragraph above has been altered to the following to account for changes in the corrected modelled number size distribution at large sizes: "Figure 9 shows the observations lie closer to the modelled size distribution of aged BC+OC (SO_4/BC/OC) and the difference between the integrated flight-mean modelled (aged BC+OC) and observed non-volatile particle distributions is statistically insignificant at the 95 % confidence level. The difference between the modelled BC-only size distribution and the observations is statistically significant in the measurement range, which suggests that the measured non-volatile particle size distribution does not only consist of BC, but

is likely to include contributions from non-volatile organic matter. In addition, sea salt particles (Jennings and O'Dowd, 1990; O'Dowd and Smith, 1993) and mineral dust may contribute to the measured non-volatile particle number concentrations, which are not included in the modelled size distribution in Fig. 9. However, these species are only likely to make substantial contributions in the super-micron size range."

P18281, L8–10: We have altered the following sentence, *"Conclusions regarding the best nucleation mechanism are hard to draw because of the limited number of ground sites where BL nucleation is needed to explain significant model-observation differences."* to be consistent with the corrected results of the significance tests: "Conclusions regarding the best nucleation mechanism are hard to draw because of the difficulty in detecting a statistically significant impact of BL nucleation on CCN-sized particle number concentrations within the uncertainty in the ground based observations."

P18282, L2–4: The statistical values for CS have been corrected from *"(NMB = -18–12 %, m = 0.66–0.90, R^2 = 0.77–0.80, average R^2_{hourly} = 0.25)"* to the following: "(NMB = -29–6 %, m = 0.53–0.69, R^2 = 0.73–0.74, average R^2_{hourly} = 0.25)"

Detailed below are the changes made to the Summary and Conclusions to reflect the corrections made to the text in the previous sections.

P18283, L26–28: The following sentence has been removed from this section: *"A t-test showed the difference between the modelled and the observed N_{50} and N_{100} was statistically insignificant at the 99 % confidence level at half and one third of the sites, respectively."*

P18284, L1–6: *"The mean number size distribution at sizes smaller than 50 nm diameter ($N_{<50}$) was underpredicted in model experiments without BL nucleation. The difference between modelled and observed $N_{<50}$ was found to be statistically signif-*

icant at all ground sites. The average overlap of modelled and observed frequency distributions of N_{50} and N_{100} was over 65 % (with small primary BC+OC particles), but less than 50 % for $N_{<50}$ without BL nucleation."
Minor changes have been made to the paragraph above: "The mean number size distribution at sizes smaller than 50 nm diameter ($N_{<50}$) was generally underpredicted in model experiments without BL nucleation. The difference between modelled and observed $N_{<50}$ was found to be statistically significant at all ground sites. The average overlap of modelled and observed frequency distributions of N_{50} and N_{100} was over 65 % (with small primary BC+OC particles), but less than 55 % for $N_{<50}$ without BL nucleation."

P18284, L22–24: *"...but we found a small but significant difference was removed at 3 out of the 15 sites for both N_{50} and N_{100} by including BL nucleation."*
The sentence above has been replaced with the following: "...but we found by including BL nucleation a small but significant difference was removed at 7 and 4 of the 15 sites for N_{50} and N_{100} respectively."

P18286, L19: *"at 11 out of 15 sites."* has been replaced with "13 out of 15 sites."

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C9917/2011/acpd-11-C9917-2011-supplement.pdf

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