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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.

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We thank the referees for their helpful and constructive comments. We have responded to all the referee comments and have made alterations to our manuscript where appropriate. The referee comments are shown below in italic font and our responses are shown in regular font. Additional text included in the manuscript in response to the referee comments is shown in bold font.

J. Pierce (Referee)

General comments:

1. *The insensitivity of particles larger than 50 nm (and particularly particles larger than 100 nm) to BL nucleation in these results is pretty remarkable. In Pierce and Adams (2009) we discuss the dampened response of CN to nucleation (this wasn't anything terribly new then, we all are aware of it), but the extent here is pretty amazing (e.g. you are shutting BL nucleation off entirely, not just scaling it by 1-2 orders of magnitude!). I believe that this could use further discussion and potentially further analysis. Obviously the dampening comes from the reduction in survival probability of ultrafine particles due to an increase in the condensation and coagulation sinks once nucleation is turned on (e.g. Pierce and Adams, 2007, Kuang et al. 2009 and numerous studies out of Kulmala's group), but it would be good to discuss this and/or look into this some more.*

We find that the impact of BL nucleation on CCN-sized particle number concentrations in our paper is consistent with previous GLOMAP studies and with the results in Pierce and Adams (2009). We add the following text to Sect. 4.1.1 (P18270, L24):

"Including BL nucleation in the model increases the campaign mean N_{50} and N_{100} in the European BL by 23–36 % and 14–20 % respectively in the BCOC_lg experiment, and by 8–12 % and 5–8 % respectively in the BCOC_sm experiment. The increase in particle number concentrations depend on the nucleation mechanism (the smallest increase in N_{50} and N_{100} is achieved with the ACT mechanism; the largest with the ORG1 mechanism). These results are in-line with the mean enhancements to CCN found by Spracklen et al. (2008); CCN number concentrations at 1% and 0.2% supersaturation (CCN (0.2 %)) were found to increase by 30 % and 6–15 % respectively at European ground sites. Pierce and Adams (2009) also show a $\sim$5% increase in BL CCN (0.2 %) over Europe when activation BL nucleation is included in their model.

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The impact of BL nucleation on CCN-sized particle number concentrations is considerably smaller than for N_{tot} (given above) and for the total particle number concentration $>$3 nm (see Table 1). The dampened response of N_{50} and N_{100} to BL nucleation arises from an increase in coagulation and condensation sinks from an additional source of secondary particles, thereby reducing the survival probability of ultrafine particles and reducing the condensational growth of these particles to CCN sizes (e.g. Pierce and Adams, 2007; Kuang et al., 2009)."

2. *When comparing the measured and modelled timeseries (e.g. Table 6), I believe it makes more sense to do a ∼5–20 hour running average of the measurements (adjust the time to the average residence time of air in the box). There will be noise on the hourly timescale of the measurements that the model cannot capture because of the spatial resolution. This would make a more apples-to-apples comparison, and I believe that comparing the r^2 values from this analysis between different model simulations would then be more meaningful.*

The point of the hourly analysis is to capture the observed peaks in particle number concentration that occur due to BL nucleation. A 20-h smoothing would eliminate such events and therefore mask what we are trying to evaluate. Nevertheless, we agree with the reviewer that there will be noise on the hourly timescale of the measurements that the model is unable to capture (please see discussion in the Summary and Conclusions, P18285, L6–12). We also add the following text to Sect. 4.1.2 (P18274, L12):

"If we adjust the interval of the modelled and observed time series to better represent the average residence time of air in the model grid box (∼5–20 hours), the results of the significance tests are improved but the conclusions regarding BL nucleation remain unchanged. If we compare the model to a 20-hour running average of the measurements, the number of sites where the difference between modelled and observed $N_{<50}$ is statistically insignificant is increased to 12 out of 15 sites, but at all but 2 of these

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sites it is still necessary to include BL nucleation to capture the observations. For N_{50} and N_{100} the number of sites with an insignificant difference is increased to 12 and 13 sites respectively, but BL nucleation is only needed to capture the observations at $\leq$2 of these sites. These results confirm the conclusions from the hourly time series analysis; to capture ground-based observations of $N_{<50}$ we need to include BL nucleation in the model, but for CCN-size number concentrations only a fairly small contribution from BL nucleation (if any) is needed to capture the observations."

And add the following text to Sect. 4.4 (P18281, L16):

"Increasing the 1-hour interval of the time series to 20 hours (an estimate of the average residence time of air in the model grid box), marginally improves the correlation between modelled and observed N_{tot} (average R^2_{hourly}=0.17 without BL nucleation; average R^2_{hourly}=0.10 with BL nucleation). The temporal correlation between the model with BL nucleation and observations remains lower than the model without BL nucleation, suggesting possible errors present in the modelling of BL nucleation events for this period."

3. *There is much discussion about the appropriate size of primary particle emissions to be used for global models because the mode size increases within the first several hundred kms from emission. This discussion is very justified, is a big problem for global models, and it is great that this paper performs sensitivity tests to determine how primary particle size affects the comparisons to observations. I too have struggled with how to account for these issues (Pierce, J.R., Theodoritsi, G., Adams, P.J., Pandis, S.N., Parameterization of the effect of sub-grid scale aerosol dynamics on aerosol number emission rates, Journal of Aerosol Science, 40, 385-393, 2009.)*
However, I do not necessarily agree with the level of favoritism of the large particle emissions in the text (e.g. P18279L9-11: "more appropriate for a global model", the numbered list on P18279L13-16 that does not include the possibility that smaller particles might be ok for the comparisons, P18286L8: "too small to be appropriate for global

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grid boxes"). While I agree it makes sense that the average sizes of primary particles in a gridbox must be larger than the primary particles very close to their emission, emissions can be occurring in many locations in a grid box and at various distances from the measurement locations. Therefore, the time-averaged data from any measurement site is representing some average processing time from the closest (or most influential) sources. The average processing times will be different from site to site (and they will certainly change with time/wind direction). In other words, the grid-box mean will generally not correspond to what is being measured.
These measurement-location-specific issues are difficult (impossible?) to capture in the global model. However, we need to understand that many measurement sites might normally be seeing less processed aerosol than others and that using smaller sized particles for emissions will give better agreement for these sites. It would be good to add discussion on this to the text.

It is important to highlight that in the discussion in Sect. 3.2, we not only take into account the growth of the emitted particles over the size of the model gridbox, but also the measured emission size of black carbon (BC) particles. To make the message clearer, we amend P18263,L3 to the following and include some additional references:

"....the peak emission diameter of the primary soot (BC) component is more likely to be around $\sim$50 nm or larger as observed by Baltensperger et al. (2002). A second mode, with a maximum in the range $D_{\mathrm{p}}\sim$40–120 nm, is observed in on-road, kerbside, and urban background number size distributions (e.g. Kittelson et al., 2000, 2006; Geller et al., 2005; Casati et al., 2007; Wehner et al., 2009; Weimer et al., 2009) and is associated with direct emissions of soot (BC) particles from from diesel and gasoline vehicles (e.g. Harris and Maricq, 2001)."

With regards to the AEROCOM-recommended emission size being too small for the large gridbox size of the model, our study is also not alone in making this statement (e.g. Spracklen et al., 2011). However the reviewer raises a good point and we add the

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following paragraph to the discussion in Sect. 3.2:

"The size distribution of primary BC+OC particle emissions averaged over the model grid box is likely to be more representative of the evolved size distribution of primary carbonaceous aerosol measured at rural background sites. It is important to note, however, that the grid-box mean size distribution will not necessarily correspond to the measured particle size at point locations. Measurement sites will be located at varying distances from aerosol emission sources which means the average processing time of the primary aerosol will also vary between sites, thereby influencing the physical properties of the particles measured. At sites where the observed particles are generally less processed than at other sites, assuming a smaller initial size for BC+OC particles may agree better with the observations and vice versa."

And make some amendments to P18263, L16–L21: "The appropriate emission size distribution to assume for primary carbonaceous particles in a global model remains ambiguous. However, since the emission size distributions used by Stier et al. (2005) and Dentener et al. (2006) are representative of how the global aerosol modelling community treats the emission of carbonaceous aerosol, we use them here in our sensitivity study. We therefore have two scenarios for size of BC+OC particles at emission: large particles (BCOC_lg; Stier et al., 2005) and small particles (BCOC_sm; AEROCOM, Dentener et al., 2006). The emitted number concentrations predicted by these two experiments can be viewed as rough upper and lower limits to the modelled primary BC+OC particle number concentration."

We also make the following amendments to the text:

P18263,L12–13: We replace "it is likely that the particle size is too small to be appropriate" with "it is possible that the particle size is too small to be appropriate".
P18269,L29–P18270, L1: We replace "despite the large emission size being more appropriate for a global model." with "despite the large emission size agreeing better with measured roadside and urban BC particle size distributions (Sect. 3.2).".

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P18279,L10–11: We replace “more appropriate for a global model” with “more in-line with measured BC emission sizes in the literature”.
P18280,L1: We add the following text (as part of the numbered list on P18279): “Assuming a very small initial size for primary BC+OC particles (experiment BCOC_vsm) increases the modelled non-volatile particle number concentration, compensating for non-volatile residuals that may be neglected by the simple representation of non-volatile particles in our model. However, we believe a diameter of 25 nm assumed for D_{FF} in experiment BCOC_vsm, is unrealistically small for directly emitted BC particles from traffic sources and (taking into account sub-grid scale processing) this diameter may also be too small for the mean BC+OC particle size over a large model grid box (Sect. 3.2).”

We make some amendments and add some additional text to the Summary and Conclusions (P18286, L5–11):

“There is large uncertainty associated with the prescribed size distribution of anthropogenic carbonaceous (BC and OC) particle emissions in regional and global aerosol models. The assumption of an initial size distribution for primary particles in global models accounts for both the size of particles at emission and sub-grid scale aerosol processes and dynamics that influence the size and number concentrations of particles shortly after emission (Jacobson and Seinfeld, 2004; Pierce et al., 2009). Information on the effective emission size distribution of carbonaceous aerosol for large-scale models is lacking in mass-based emission inventories (e.g. Cooke et al., 1999; Bond et al., 2004), and thus far has only been provided by AEROCOM (Dentener et al., 2006). The widely used emission sizes recommended by AEROCOM assume particles that may too small to be appropriate for large model grid boxes, but the agreement with observed particle concentrations is generally much better than when we emit larger particles that may be more realistic.”

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Specific Comments:

1. *P18267 L12-19: If you have 2 externally mixed populations, why not put all nucleated particles into one population and all primary emitted particles (BC-OC-sea salt...) into another. Allow sulfate and SOA to condense onto both. When particles from the 2 populations coagulate with each other, put the resultant particle into whichever population had the LARGER parent particle. This formulation would give the exact contribution to CCN from nucleation versus primary emissions. You could present exact estimates of this rather than trying to tease out the influence of nucleation on CCN. I don't understand why you would put nucleated particles and sea salt into the same bins and why coagulation between particles in the two populations would always go into the same population (rather than whichever population had the bigger parent particle).*

To clarify, nucleated particles and sea salt are put into the same distribution but not the same bins because their mean particle sizes are very different. When two particles coagulate in the model the resultant particle goes into the size bin corresponding to the total size of the coagulated particle. Therefore, we are still able to calculate the exact contribution of e.g. particles from BL nucleation (Spracklen et al., 2008) or primary carbonaceous particles (Spracklen et al., 2011) to total CCN by turning off the sources of these particles in the model.

For this study it was necessary to put the carbonaceous particles in a separate distribution in the model in order to track the number concentration of BC/OC particles separately to compare with observations of non-volatile particles. In addition, we have also only focussed on the contribution of primary carbonaceous aerosol to CCN-sized particle number concentrations in our paper, rather than the contribution of all primary particles. The suggested model set-up would be useful for looking at total primary (and secondary) contributions to CCN with just one model simulation. We therefore thank the reviewer for their suggestion and we will certainly consider this model set-up in future studies quantifying the contribution of primary and secondary aerosol to

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CCN. However, for this study we believe it is unnecessary to re-run the model with this configuration.

2. *Section 4.1.2: I don't understand how the "primary aerosol" stats and "BL nucleation" stats are aggregated together when you have many different model simulations. Can you please explain more clearly?*

We agree that the explanation for grouping the experiments together is lacking from this section. We add the following text to Sect. 4.1.2 (P18272, L1):

"In this section, we essentially test the significance of all the plausible primary aerosol experiments and BL nucleation experiments and so group the model simulations into those without BL nucleation (simulations 1–2, Table 2) and those with BL nucleation (simulations 3–10, Table 2). The range in the first set of experiments represents the uncertainty in the assumed emission size distribution for primary BC+OC and the range in the second set of experiments represents the uncertainty in the empirical BL nucleation parameterisation used in the model."

We have also altered the headings in Fig. 5 to "model experiments without BL nucleation" and "model experiments with BL nucleation" to make it clearer what each figure refers to.

3. *P18282 L17: "In particular, we neglect the impact of cloud cover on incoming radiation on OH concentrations". Really? Is this because you are running in a mode where OH is offline and monthly averaged? I find it hard to believe that TOMCAT doesn't predict these online.*

OH concentrations in the version of GLOMAP used in the study are specified using 6-hourly monthly-mean 3-D gridded concentration fields from a TOMCAT simulation with detailed tropospheric chemistry (Arnold et al., 2005). The oxidants are read in at 6-

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hour intervals and linearly interpolated onto the model timestep. The modal version of GLOMAP can be run with coupled chemistry but this is not available for the bin version of the model.

4. *P18285 L10-14: Spracklen et al. (2006) and Hyytiala data. When I was reading the section on timeseries, I kept scratching my head thinking, "I thought BL nucleation in GLOMAP does great in Hyytiala", so I'm glad this was mentioned here. I would appreciate any additional insight into this... have things in the GLOMAP changed greatly since then? Is there a major difference between the seasons tested in Spracklen 2006 and this paper?*

There have been no large changes in the model since the study by Spracklen et al. (2006) so it is more likely that any difference in model performance is down to the different period studied (March 2003 versus May 2008) e.g. different meteorology, air masses, pollution levels, condensation sink etc. In March 2003, there were very clean periods where several distinct nucleation events were observed, which make it easier for the model to capture the variability in CN. But in fact, if we compare the same model set-up as used in Spracklen et al. (2006) (activation BL nucleation mechanism and AEROCOM-recommended BC/OC emission sizes), the agreement between model and observations at Hyytiala is also reasonably good for the EUCAARI IOP (frequency distributions overlap by 84%) and the model bias (-9%) is smaller than in Spracklen et al. (2006). It is the correlation between hourly mean modelled and observed N_{tot} that is poor in our May 2008 study, but since Spracklen et al. (2006) do not show the correlation coefficient between model and observations it is difficult to compare the model performance on the hourly scale.

In Spracklen et al. (2006), the model has a tendency to overestimate peak particle concentrations during nucleation events and also to underpredict the rate of decrease of particle concentration after an event. These factors are both evident in the modelled

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CN timeseries at Hyytiala in our study, which is likely to contribute to the poor correlation. But without a detailed evaluation of the factors controlling BL nucleation at this location (gas-phase concentrations of SO_2, H_2SO_4, monoterpenes) we are unable to fully explain the poor correlation at Hyytiala for this period.

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M. Schulz (Referee)

General remarks:

1. *The isolation of the importance of nucleation is a strength of this paper. However, I wonder if there should not be a more explicit paragraph in the discussions/conclusions on other processes influencing size distributions, such as cloud processing, inhomogeneity of source size, dynamics of semi-volatile size distributions.*

This is a good suggestion. We add the following text to the Summary and Conclusions:

"It is important to note there are processes in addition to BL nucleation, condensational growth and primary particle emissions that influence the particle number size distribution over Europe, such as cloud processing, wet/dry deposition, coagulation, dynamics of semi-volatile size distributions etc. These processes have not been explored in detail in this study, but we are working towards a more complete uncertainty analysis of the GLOMAP model (Lee et al., 2011)."

We have already touched upon the inhomogeneity of source size (P18275, L13): "...the emitted primary particle size/number concentration is more variable across Europe than assumed by the constant emission size distribution prescribed in the model..."

We add the following to the conclusions: 'The particle number emission inventory will also better represent the inhomogeneity of source size and how this influences the particle number size distribution in the BL."

2. *Chapter 4.1.2 and further use of statistics is used to underpin the importance of nucleation for reproducing hourly evolution of number concentrations. That is of course a possible explanation for improved correlation in some sites. However, I wonder if also a random process adding ultrafine particles would improve the statistics. This suspicion enters since the baseline simulation of e.g. BCOC_sm has probably a very*

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flat, damped time series of number concentration evolution, due to missing processes to create temporal fluctuations on the order of hours. I think a random adding of number would be a useful additional valid null hypothesis to compare the experiments with BL nucleation with.

This is an interesting suggestion and provides a useful test of the performance of a deterministic model against random fluctuations. However, we believe this analysis would be extensive and beyond the scope of this study. It would be necessary to randomise several factors of the nucleation event such as the magnitude, time of occurrence, duration, location, spatial extent etc. We will certainly consider applying this test to future evaluations of the modelled CN timeseries.

3. *Also - shouldn't one try to evaluate a normalized diurnal cycle of number concentration. Isn't there a diurnal signal from nucleation to be expected?*

We have carried out an evaluation of the modelled diurnal cycle of particle number concentration, but have not included this analysis in the paper. The reason for this is that we find by averaging the diurnal cycle over the campaign period, the signal from BL nucleation is smeared out and mixed with pollution events so that a strong nucleation signal is only visible at less than half the ground sites. In addition, we find the results of the evaluation lead to the same conclusions as the evaluation of the hourly CN time series which is already shown in the paper. For example, the correlation coefficient between the campaign average model and observed diurnal cycle of N_{tot} is reasonably good with BL nucleation (average $R^2 = 0.34$), but decreases when BL nucleation is included, suggesting that the model with BL nucleation is unable to adequately capture the observed nucleation events at most sites for this period. We will include a detailed evaluation of the modelled diurnal cycle in future work where we focus more strongly on identifying the best-fit nucleation mechanism.

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4. *I find the discussion on the primary OCBC size distribution already pretty good, and the authors are applauded for that. However, some aspects on the discussion of the potential non-validity of AeroCom size assumption could be extended:*
1) I believe it is worth discussing more that the spread of the primary distribution is considerably smaller in Stier et al. This has an impact on the number of particles and this is probably as large as the mode diameter chosen.

This is a good suggestion. We have made some amendments to P18261, L17–23 and added some additional text on the spread of the primary distribution:

"The factor $\sim$2 difference in the recommended values for D imply very different BC+OC number concentrations (for fixed mass); AEROCOM requiring emitted number concentrations to be a factor ~ 8 higher than Stier et al. (2005) for fossil fuels.

The emission size distribution used by Stier et al. (2005) has been adapted from AEROCOM recommendations to fit the standard deviation of the size modes in their model. As a result, the spread of the primary distribution in Stier et al. (2005) ($\sigma = 1.59$) is considerably smaller than the spread of the AEROCOM-recommended distribution ($\sigma = 1.8$). Reducing the spread of the assumed emission size distribution from $\sigma = 1.8$ to $\sigma = 1.59$, increases the emitted number concentration by a factor of $\sim$1.8, if the value of D was to remain equal. It is important to note that in GLOMAP-bin, we are free to specify any shape distribution within the resolution offered by the 20 size bins, but use the two values of σ as specified above. The difference in the parameters assumed by Stier et al. (2005) and Dentener et al. (2006) corresponds to an overall factor $\sim$4.4 difference in the emitted number concentrations of fossil fuel BC+OC particles."

We have also carried out the following amendments taking into account the difference in the assumed value for σ:
P18278, L26: we alter "a factor of $\sim$8" to "a factor of $\sim$4.4"
P18279, L11: we alter "more than a factor of $\sim$10" to "more than a factor of $\sim$6"
P18287, L7: we alter "a factor of 10" to "a factor of 6"

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2) The assumption of rather small initial emitted sizes might be valid if the simulation of particle dynamics is captured by the time evolution and vertical mixing of aerosol particles in the global model. The length of time steps, the vertical layering and mixing in the model and the particle dynamics may help capturing the aerosol size evolution dynamics by the model even if no nucleation is included but if small primary particles are assumed. In a way the AeroCom distribution might reflect poorly parameterized particle formation through nucleation.

We agree with the reviewer and we have already acknowledged that this compensation may be occurring in the Summary and Conclusions (P18286, L13–15): "It is likely that the BCOC_sm experiment is compensating for missing particles from BL nucleation by increasing the primary particle number, and thus agrees better with observations."

3) Measurements showing larger sizes in urban areas might not reflect primary emission sizes.

We agree that measurements showing larger sizes in urban areas might not reflect primary emission sizes, however roadside and laboratory size distributions measurements of vehicular emissions also suggest larger mode diameters for fossil fuel BC particles than recommended by AEROCOM (e.g. Harris and Maricq, 2001; Kittelson et al., 2006). We also add some discussion on the traffic-related nucleation mode observed in on-road or kerbside size distributions (see respnse to comment "5)" below). These measurements, carried out close to the source, are more likely to reflect primary emissions sizes for traffic (fossil fuel) sources.

4) The particle dynamics are clearly a subgrid process to a global model. Without more explicit modelling on finer scale, all size assumptions are necessarily a parameterization.

We agree and have inserted the following into P18260, L26:

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"The assumption of an initial size distribution for primary particles in global models accounts for both the size of particles at emission and sub-grid scale aerosol processes and dynamics that influence the size and number concentrations of particles shortly after emission (Jacobson and Seinfeld, 2004; Pierce et al., 2009). In GLOMAP, the primary particles are "emitted" assuming an initial size distribution and then the size and number of particles are allowed to evolve during atmospheric transport."

We also add some additional text and references to P18262, L1–4: "Although the emitted mass is generally conserved during transport and dispersion over the GLOMAP grid box ($\sim$200 km at European latitudes), the number size distribution of primary particles shortly after emission can be altered significantly by (sub-grid scale) atmospheric dynamic processes such as dilution, condensational growth, heterogeneous and self-coagulation, evaporation, and nucleation (e.g. Kittelson, 1998; Wehner et al., 2002; Zhu et al., 2002; Zhang and Wexler, 2004; Zhang et al., 2004; Roldin et al., 2010). Explicit modelling of these subgrid-scale processes would be too computationally expensive for a global CTM, which is why an assumption of the emission size distribution is necessary."

5) A larger size of BC non-volatile, primary particles might be likely. But how much is that impacting the overall number size distribution, which is a result of other condensing and nucleating aerosol components?

We add the following paragraph to the discussion in Sect. 3.2:

"However, assuming a larger emission size that is consistent with measurements of primary BC/non-volatile particles may neglect possible contributions to the total size distribution from particles formed via homogeneous nucleation and condensation processes in the vehicle exhaust (e.g. Abdul-Khalek et al., 2000) and/or combustion-generated nanoparticles of OC (e.g. Sgro et al., 2008). It is important to note that we class particles formed through homogeneous nucleation shortly after emission (either in the vehicle tailpipe or in the emission plume) as primary particles in the model, since they

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are formed from emitted precursor gases on sub-grid scales. Semi-volatile particles produced via this process may undergo gas-to-particle partitioning with atmospheric dilution; involving evaporation and possible re-condensation onto surfaces of larger particles in the exhaust plume e.g. soot or background aerosol (e.g. Zhu et al., 2002; Zhang et al., 2004). These processes make it difficult to quantify their contribution to the average BC+OC number size distribution over the model grid box."

General remarks to all figures:

1. *Since one emphasis of the paper is to see how different sensitivity experiments reproduce the different observed size properties, something pretty dense in information, it would be much more digestable if each experiment is identifiable with the same colour throughout all figures.*

We agree with this suggestion and have updated the colour scheme in the figures accordingly.

2. *I would also like to request that a legend, e.g. with colored experiments names is added to all figures. It would be so much more readable if experiment abbreviations and associated color (now even changing from graph to graph) are not "hidden" in the figure caption.*

We agree with this suggestion and have added a legend (with experiment name and corresponding colour) to figures where possible.

Specific remarks:

1. *P18268, L24: "the modelled size distribution looks very different than the primary distribution"*

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This is probably right, but it is hard to see in the figures. The size distributions in figure 2 and 3 are plotted quite differently. Please add a characteristic size distribution (mean over all stations from "reference" simulation?) to figure 2. I think that would be illustrative and easy to do.

This is a good suggestion. We have added a figure showing the campaign mean modelled size distribution over all sites for the BCOC_sm and BOCO_lg experiments.

2. *P18270, L4 : "decrease in the spatial correlation between the model with BL nucleation and observations (BCOC_lg, 0.12–0.52; BCOC_sm, 0.48–0.68)"*
The information in the brackets is hard to understand, since you refer not only to BCOC_lg/sm experiments, but to all experiments, right?

The information in the brackets refers only to the spatial correlation of the experiments with BL nucleation. However, we agree that the information is not communicated very clearly in this sentence. We have amended the text to the following:

"Including BL nucleation in the BCOC_lg experiment reduces the low bias of the model but does not fully explain the shortfall in N_{tot}. In addition, the magnitude of the slope of the linear regression between modelled and observed N_{tot} remains low ($m = 0.22$–0.26) and there is a decrease in the spatial correlation between model and observations with the ORG1 (R^2 =0.35) and ORG2 (R^2 =0.59) mechanisms. These results suggest possible errors in the modelling of nucleation events (discussed in Sect. 4.4), which may be a reason why BL nucleation is unable to explain the shortfall."

Additional changes and corrections:

Below are some general changes we have made to the manuscript.

We alter P18254,L17 to the following: "...by ∼4–5 orders of magnitude..."

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We alter P18264, L3–6 to the following: “This process involves the reaction of biogenic monoterpenes with O_3, OH and NO_3 (assuming the reactivity of alpha-pinene) to form a gas-phase oxidation product with a 13% molar yield (Spracklen et al., 2006). This first stage oxidation product can form SOA through condensing with zero vapour pressure onto pre-existing aerosol (Spracklen et al., 2006, 2008).”

We alter P18265,L8–9 to the following: “We assume that the concentration of organic vapour ([organic]) can be represented by the gas-phase concentration of the first stage oxidation product of monoterpenes (described above).”

We add the following to P18261, L11: “Grid-level and size-resolved particulate emission factors for traffic sources have been provided by Zhang et al. (2005), but the grid scale used in the study ($\sim$300 m) is far smaller than the grid box size of most large-scale models.”“

We alter P18262, L3 to the following: “We encounter further uncertainty associated with the assumed size distribution for primary BC+OC emissions...”

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