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Interactive comment on "The size-composition distribution of atmospheric nanoparticles over Europe" *by* David Patoulias et al.

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1. *In this paper, the authors apply the PMCAMx-UF air quality model to a European domain to assess its ability to simulate nanoparticles over the domain. They suggest the model performs favourably, with the results generally within a factor of 2. They also assessed the simulated impact of organics, finding it leads to a large increase in N100 particles in parts of Europe. They also found using VBS improved model performance. In general, this work appears to be well done, though is rather limited in its scope of model evaluation, the core of the study.*

We do appreciate the positive assessment of our work. We have made several changes in the revised text in an effort to improve it and avoid any misunderstandings. These

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changes are described below following each comment of the reviewer (in italics).

2. *In a model application and evaluation such as this, it is important to provide some idea of what is driving the model. Is it the emissions of nanoparticles (which is a rather uncertain quantity, particularly the appropriate size distribution to be used given the model resolution: how is the very near field dynamics of traffic emissions treated?)? Is it boundary conditions? Is it nucleation, and if so, from emissions within the domain or from boundary conditions? (Note: there is rather little discussion of boundary conditions or emissions: they should discuss both in more detail and provide a spatial distribution of emissions by size. They can use NTotal, N10, N50 and N100, though as noted below, I would use N10-50, N50-100, N>100). Even if just for a one-week simulation, they should provide the results of four additional simulations: Halving the BCs on all PM, halving the BCs on species that might react to form condensable species, Halving the emissions. (They need not use changes of one half, but something where the response would be seen if it is important). In the end, the article should address, very precisely, why the simulated levels are what they are, by size. For now, there is a bit of that for organics.*

We have followed the suggestion of the reviewer and quantified the sensitivity of the model predictions to the boundary conditions and emissions. We performed eight additional simulations with: (i) 50 percent reduction of PM boundary conditions, (ii) 50 percent reduction of the boundary conditions for all gases, (iii) 50 percent reduction of just the SO_2 boundary conditions, (iv) set the SO_2 at the boundaries equal to zero, (v) 50 percent reduction of PM emissions at all sizes, (vi) 50 percent reduction of the emissions of all gases, (vii) 50 percent reduction of just the SO_2 emissions, and (viii) set the SO_2 emissions equal to zero. The effect of these changes on the particle number distribution was quantified. Summarizing the effect of the changes in the boundary conditions by 50 percent was less than 5 percent for all cases, showing that the boundary conditions were not a major driver of the simulation. On the other hand, the emissions of sulfur dioxide, other vapors and particles had a major effect with changes

of 10-35 percent for a corresponding 50 percent emission changes. Setting the sulfur dioxide emissions to zero resulted in changes of 40-70 percent in the concentrations in the different particle size ranges showing its importance for new particle formation and growth during this photochemically active period. The results of this sensitivity analysis are now discussed in a new sensitivity analysis section.

3. *It is interesting that their simulated results are spatially more uniform than might be expected. Looking at Fig. 5, the simulated results are typically about 2500-10000. The observations go rather lower. This requires more discussion. It also appears as though the results at Hyytalia are dominated by boundary conditions that are fixed in time... Is this true? If not, an interesting pseudo-steady state appears to be at work that should be explained. On the other hand, the simulation shows more variability than the observations in Fig. 8. Again, rather more discussions is warranted as this is the focus of the paper.*

The comparison of the model predictions with the observed values in Figure 4 for the 16 sites suggests that the model does a good job in capturing the observed variability in all size ranges. There is no evidence that the model fails to capture the observations at the low concentration levels. This is now mentioned in the revised paper.

Indeed, the model predictions at the station at Hyytiala are affected more than most other sites by the boundary conditions due to its location and the prevailing meteorology. A discussion of the sensitivity of the corresponding predictions and the average diurnal profiles for this station has been added. Also reasons for the discrepancies between observations and predictions for this important site are analyzed in additional detail.

4. *The title should be changed. This article is not focused on nanoparticles over Europe, but the simulation of nanoparticles over Europe. It also has almost nothing on the actual composition (they have a small piece on sensitivity to SOA formation). Much*

more information and analysis is necessary to have the more general title. I would propose "Spatial distribution of simulated nanoparticles over Europe".

We agree with the suggestion of the reviewer and we have added the word "simulation" to the title of the paper.

5. *The comparison of results to the flight data is remarkable. They really do need to show what is contributing to the results, particularly how the bump at 800 m exists given the vertical diffusion found in most air quality models. The bump at 800 m is very interesting and really understanding it is key. Given the model's ability to capture this, I was surprised there was no discussion of it. They could take the analyses done to assess the processes leading to their simulated levels discussed above and use that.*

This is a good point, but unfortunately the explanation for the bump at the 800 m is rather mundane. There were few measurements at this altitude in a few flights in days with relatively high particle number concentrations. These created the apparent bump in the measurements. The model captured these high concentration periods so it predicted the same bump for the average concentration profile. We now explain in the revised paper that the average profile is the result of averaging of measurements and predictions from different flights and simulations. There were relatively fewer measurements in altitudes above 600 m. The number of samples at different altitudes changed for each flight creating additional variability in the measured profiles.

6. *The manuscript also does not provide any information on how well the model captures aerosol composition. If the model is within a factor of 2 on total number, but off by a factor of 2 on OC or sulphate mass, this has important implications, particularly if the differences are in different directions.*

We have followed the suggestion of the reviewer and added a section comparing the model predictions for PM1 composition with the corresponding measurements. Overall, the model reproduces the observations of inorganic aerosol components (sulfate,

nitrate, ammonium) reasonably well (e.g., errors in the average concentrations of less than 0.5 μg m^{-3} in the Italian sites) but it tends to underpredict the organic aerosol concentrations. For example, the OA in San Pietro Capofiume is underpredicted by approximately 40 percent. This underprediction of the organics is the major reason for the underprediction of the condensational sink shown in Figure 7. Based on previous work with the sister model PMCAMx in Europe, the chemical aging processes, that are not simulated in this version of PMCAMx-UF, should be able to explain a significant fraction of the missing OA. The role of these processes, as explained elsewhere, is the topic of a follow-up publication that is currently under preparation.

7. *I think it would be better to show their results by size groups, e.g., N<10, N10-50, N50-100 and N>100. This would better demonstrate the variability in the different sizes, and if, for example, one size range is much more uniform than the others.*

We have followed the suggestion of the reviewer and added selected results in the Supplementary Information using the suggested size distribution ranges. We would prefer to keep though the current size ranges in the main paper for consistency with previous studies (Fountoukis et al., 2012; Baranizadeh et al., 2016).

8. *It appears that WRF is applied without nudging, but is simply reinitialized every three days. Why? To what degree does the reinitialization impact the system? How does the performance degrade after multiple days?*

We have tested the performance of WRF with different types of initialization and nudging. The three-day reinitialization has been chosen because of its simplicity and the fact that the corresponding WRF predictions remain consistent with all the measurements. The measurements are pre-processed by the WPS package, which provides each atmospheric and static field with fidelity appropriate to the chosen grid resolution of the model. The performance of WRF for Europe against observed meteorological variables has been the topic of several recent studies (Jimenez-Guerrero et al., 2008;

de Meij et al., 2009; Im et al., 2010; Argueso et al., 2011; Garcia-Diez et al., 2012) showing good performance. A brief discussion of this point has been added to the paper.

9. *In summary, the submitted manuscript is a good start on what can be a nice contribution to the literature, though it is currently too limited in what they have done and what they explain. The authors should provide a more extensive analysis on what processes drive their simulations. At a minimum, they should: 1. Provide a set of calculations showing how the model responds to changes in emissions and boundary conditions, and the role of sulfate nucleation, and the origin of the SO_2 (BC or emissions inside). 2. Provide some information on how well the composition is captured is also needed. Without such further information it is difficult to say whether the model results are reasonable or not. 3. Provide more detailed information on the model application, including vertical cell spacing, overall performance for more species. How well does WRF capture the meteorology? 4. Change the title to include "simulation" as the article does not really focus on the distribution of nanoparticles (as observed).*

We have done our best to address the comments of the referee. More specifically, the results of eight sensitivity simulations are now described in a new sensitivity analysis section (please see response to Comment 2). A brief evaluation of the performance of PMCAMx-UF in reproducing the PM composition observations has been added (please see response to Comment 6). We have also added in the Supplementary Information a table with the vertical levels of the model and a brief justification of our choice for the WRF reinitialization (response to Comment 8). The title has been changed to clarify that this is a modeling study (response to Comment 4).

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