

Interactive comment on “Dynamic evaluation of a multi-year model simulation of particulate matter concentrations over Europe” by È. Lecœur and C. Seigneur

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Response to Anonymous Referee #2

The authors would like to thank the reviewer for evaluating this work and for the helpful comments and suggestions.

SPECIFIC COMMENTS

1. *Emissions (page 480, lines 14-18): I guess that you do not perform any horizontal disaggregation from the EMEP inventory because your model run at $0.5^\circ \times 0.5^\circ$. But, how do you perform vertical and temporal disaggregation from the inventory ? You used MEGAN model from biogenic emission, do these emissions include NO_x from soils ? If it is true, how do you considered NO_x from SNAP10 ? Have you considered emissions from forest fires ?*

disaggregation from the EMEP inventory because your model run at $0.5^\circ \times 0.5^\circ$. But, how do you perform vertical and temporal disaggregation from the inventory ? You used MEGAN model from biogenic emission, do these emissions include NO_x from soils ? If it is true, how do you considered NO_x from SNAP10 ? Have you considered emissions from forest fires ?

It is correct that the model horizontal grid spacing matches the EMEP inventory resolution so that no horizontal disaggregation was needed. Surface emissions were injected into the model surface layer. Elevated point source emissions were injected into model layers based on the EMEP table for the vertical distribution of anthropogenic emissions. Temporal profiles by source sectors are also provided by EMEP. MEGAN biogenic emissions do include NO_x emissions from soils. These emissions were removed from SNAP10 of the EMEP inventory to avoid double counting. Emissions from forest fires were not included, which could lead to some PM underestimation in summer; however, modeling the impact of forest fires is still an area of research due to uncertainties in emission rates and smoke plume heights. These points are now mentioned in the manuscript.

2. *Boundary Conditions (page 480, line 19). You use MOZART-4 model to gas-phase and aerosol species at the boundaries. How many species (gas and aerosol) from MOZART-4 do you considered in the boundaries ? How have you matched the MOZART-4 VOCs to CB05 ? Specifically, how have you managed the lumped species called BIGALK, BIGENE and TOLUENE from MOZART in your CB05 mechanism ?*

Sixty species from MOZART-4 are considered in the boundary conditions. The MOZART-4 VOC species were matched to CB05 following information available in Emmons et al. For example, BIGALK is considered as ALK3 and thus matched

to 3.0 PAR, BIGENE to 1.0 PAR + 1.0 OLE and TOLUENE to TOL (instead of a combination of TOL and XYL). The aerosol species were also matched to Polair3D species. Most of the species were directly matched to Polair3D, except OC1 and OC2, which were converted to 2.25 PSOAP + 2.88 PSOAP + 3.87 PSOAP. These coefficients, which depend on the SVOC/POA and OM/OC ratios are from Couvidat et al. (Atmos. Chem. Phys., 13, 983-996, 2013). These points are now clarified in the paper.

3. *Aerosol module (page 481, line 9). Do you consider aerosol dynamic ? Do you consider nitrate aerosol in the coarse fraction ?*

Aerosol dynamics (nucleation, condensation, evaporation, and coagulation) and nitrate aerosol in the coarse fraction are considered.

4. *Section 2.2 PM2.5 spatial distribution and chemical composition over Europe. This section is very descriptive and it does not explain the reason of such patterns. I am surprised about the high contribution of organic matter in Scandinavia in Figure 3. Do you have any reason for that ? It seems like organic matter contribution in this area came from boundary conditions ? Could it be unrealistic ?*

MEGAN shows high biogenic emissions of terpenes over Scandinavia. Moreover, there is significantly less PM2.5 over this region compared to the rest of Europe. A combination thereof explains the high contribution of organic matter in Scandinavia. We have added some discussion of the spatial patterns of the PM2.5 component concentrations in the revised text.

5. *Section 3. Operational evaluation and section 4. Dynamic evaluation. Why don't*
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you evaluate NO₂ concentration ? Do you have any idea about this performance in your model system ?

Elevated NO₂ concentrations are mostly an issue at urban locations. The spatial resolution used here (50 km × 50 km) is not suitable for such an evaluation. For example, the model was evaluated for NO₂ over Europe and showed errors on the order of 70% (Sartelet et al., Atmos. Environ., 41, 6116-6131, 2007). The model was also evaluated for NO₂ over the Paris region (Roustan et al., Atmos. Environ., 45, 6828-6836, 2011) and showed an error of 67% and a bias of -48% for a grid spacing of 5 km. A different evaluation over the Paris region (Briant et al., Atmos. Environ., 68, 162-173, 2013) showed that it is necessary to have a subgrid-scale treatment to obtain satisfactory performance for NO₂.

6. *Overall, the merit of the paper would significantly increase if the authors could more clearly identify current model deficiencies, point to specific chemical/dynamical processes in Polyphemus/Polair3D responsible for the underestimations, and suggest ways to improve those. Do you have any idea about the overestimation of PM10 and PM2.5 daily concentrations ? It is really surprising, since most models in Europe underestimate PM10.*

The overestimation of PM10 is slight (18.4 μg m⁻³ simulated against 17.3 μg m⁻³ in the measurements), but it is correct that Polyphemus tends to simulate greater PM10 concentrations than other models over Europe (Sartelet et al., Atmos. Environ., 53, 131-141, 2012). We believe that the comprehensive treatment of organic aerosols in Polyphemus, which leads to reasonable agreement with observations of carbonaceous aerosols (J. Geophys. Res., 117, D10304, 2012), can explain in part this slight overestimation because the nitrate overestimation is not compensated by an organic underestimation (as it was the case in some

earlier modeling studies). The overestimation of daily PM_{2.5} can be explained by the overestimation of nitrate, which is a greater fraction of PM_{2.5} than of PM₁₀. We now elaborate on those points in the paper.

7. *You also indicate that Polyphemus/Polair3D overestimate nitrate. Do you have any idea about this behavior ? Could it be related to ISORROPIA thermodynamic equilibrium ?*

We do not think that this behavior could be related to the ISORROPIA thermodynamic equilibrium because mass transfer between the gas phase and the particles is fast for fine particles and, therefore, equilibrium is a good assumption. As most CTMs, Polair3D overestimates nitrate (e.g., AQMEII study, Solazzo et al., Atmos. Environ., 53, 75-92, 2012). There are artifacts in the measurements methods, due to the volatilization of ammonium nitrate from filters, although an evaluation of nitrate measurement methods in Europe did not show any significant bias (Schaap et al., Atmos. Environ., 38, 6487-6496, 2004). In addition, the overestimation of nitrate could be due to the slight underestimation of the sulfate by the model ($2.2 \mu\text{g m}^{-3}$ simulated against $2.3 \mu\text{g m}^{-3}$ in the measurements). Thus, not enough ammonia is consumed by sulfate favoring the formation of additional ammonium nitrate. Finally, there is still significant uncertainty about ammonia emissions including their magnitude and temporal variability.

8. *In the comparison to other model evaluation I recommend you to clarify that the models use different configurations (emission disaggregation, boundary conditions, and meteorology, among others), are run for different years, with different horizontal resolution, and different set of air quality stations for the evaluation, etc.*

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Done.

9. *I recommend you to complete the tables A1, B1, B2, B3, and B4 with the year of the simulation, the name of the chemical-transport model, and the number of the stations include in the evaluation when possible.*

Done.

TECHNICAL CORRECTIONS

1. *Be consistent using PM_{2.5} of PM_{2.5} with subscript (especially in figures 2, 4, 5, 7 8 and 9).*

Done.

2. *Be consistent using ozone or O₃ along the text. The same for SO₄²⁻ or sulfate, NO₃⁻ or nitrate, etc..*

Done.

3. *Pg 484, line 2: "sites are remote rural background stations".*

Done.

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4. *Pg 486, line 5: remove "Daily PM10 is well estimated" by "Daily PM10 is overestimated".*

Done. We replaced it by "Daily PM10 is slightly overestimated", since the average of the simulation is $16.6 \mu\text{g m}^{-3}$ (against $15.9 \mu\text{g m}^{-3}$ in the measurements).

5. *Pag 487, line 3-4: remove "See Appendix B" by "Table B1".*

This section refers to all Appendix B, since it is about the evaluation of PM2.5 and its main components. Table B1 only refers to PM2.5.

6. *Pag 490, line 18: replace "data" by "measurements".*

Done.

7. *In Pag 492 line 29, Pag 493, line 1 you talk about soil dust. How do you include this ? Include some comments in section 2.1.*

We meant mineral dust. It has been clarified in the text. Mineral dust comes from the boundary conditions because there are no major arid areas within the modeling domain.

8. *Pag 495, line 9: "... over Poland in winter than in summer".*

Done.

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9. *Table 1. Caption: talk about the correlation coefficient. Include the temporal base of the statistics, hourly or daily ? Adjust significant digits in Table 1.*

Done.

10. *Table 2. Caption: include units (%). Used ozone as O_3 , to be consistent. Adjust significant digits in Table 2.*

Done.

11. *Table 4. Describe the meaning of DJF and JJA.*

Done.

12. *Table A1. Insert the reference of the data, I think Sartelet et al. (2012).*

Done.

13. *Table B1. Indicate that the data corresponding to "This study" represent the average (2000-2008).*

Done.

14. *Figure 2. Caption: include the year (2000-2008).*

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Done.

15. *Figure 3. Caption: include the year (2000-2008) and the units (%).*

Done. The years have been included in the caption, but there is no unit for this figure as it represents a fraction (between 0 and 1) and not a percentage.

16. *Does Figure 6 show SO₂ emission or concentration ? Please correct it accordingly with the units.*

Figure 6 shows SO₂ surface emissions. The unit has been changed to $\mu\text{g m}^{-2}$.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 475, 2013.