

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

È. Lecœur and C. Seigneur

eve.lecoeur@cerea.enpc.fr

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Response to Reviewer #1

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

GENERAL COMMENTS

1. *Both the operational and the dynamic evaluations are based on a small number*
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of stations for PM_{2.5}. This means that the statistical indicators are not highly significant. The available data do not provide enough information for a full model evaluation and thus for assessing the link between meteorology and air quality. In particular, the impact on air quality of weather regimes in the parts of Europe having no station cannot be analysed. This issue needs to be discussed in the paper.

It is true that both evaluations are based on a small number of stations for PM_{2.5}, particularly for the dynamic evaluation, and we agree that this is a limitation of this study. For each year in the operational evaluation, we used all EMEP stations that provide at least 50% of the measurement data. The dynamic evaluation needed to be performed on stations that provide daily data for PM_{2.5} and its components for a common period of over at least a year. These requirements were used to increase the significance of the statistical indicators. Since PM_{2.5} measurements are relatively recent in Europe, the number of stations meeting these requirements is still small. Nevertheless, the ability of the model to reproduce the effect of meteorology on PM_{2.5} and its inorganic components is characterized at several stations, which are located in various parts of Europe and which provide different types of responses of PM_{2.5} and its components to meteorology. However, one should be cautious not to extrapolate the model dynamic performance to regions far remote from those stations. For example, the lack of PM_{2.5} chemical composition data in the western part of the domain (i.e., the countries near the Atlantic Ocean: Ireland, the United Kingdom, France, Spain and Portugal) is a limitation of this study. The results of studies on the impact of meteorology/climate change on PM_{2.5} compositions on that westernmost part of Europe should, therefore, be treated with caution. The results presented here provide the first dynamic evaluation of an PM_{2.5} air quality model with respect to meteorology, but we have added some text mentioning the limitations associated with the limited amount of data available.

2. *The authors perform an interesting evaluation of the model (operational and dynamic). Some deficiencies of the model are shown and it would be interesting to provide an analysis of the reasons/processes leading to these deficiencies.*

The model evaluation showed that nitrate and ammonium are overestimated. This overestimation results from a combination of various factors. First, artifacts in the measurement methods, due to the volatilization of ammonium nitrate from filters can contribute to the model overestimation, 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 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. Also, there is still significant uncertainty about ammonia emissions including their magnitude and temporal variability. Finally, taking the mean over 5 years to generate pseudo-climatological boundary conditions is also a source of uncertainties.

SPECIFIC COMMENTS

1. *Introduction: Only PM2.5 are studied. PM10 are also of interest. Moreover, there are more observations available for PM10. Why did you focus on PM2.5 ? This should be explained more clearly in the introduction.*

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We agree that there are more data on PM10 than on PM2.5, however, chemical speciation is available for PM2.5 and chemical speciation is needed to explain the response of PM2.5 to meteorology. This point is now specified in the introduction.

2. *Introduction, page 479, line 9-10: The goal of this study is to make a dynamic evaluation. This will likely serve the analysis of future simulations aiming at studying the impact of climate evolution on air quality. This could be added to make clear the link between the dynamic evaluation and the paragraph of the studies on the effect of climate change on air quality (starting on page 477 line 23).*

We added the following text : "Such an assessment of model performance appears needed since air quality models are increasingly being used to investigate the effect of climate change on future PM concentrations."

3. *Section 2, page 480, line 9-10. Specify if this is ECMWF forecasts or analyses which are used.*

ECMWF analyses are used. This is now specified in the text.

4. *Section 2, page 480, last paragraph: For the years 2000 to 2003, pseudo-climatological boundary conditions are created from the mean of 2004-2008. This choice needs to be argued. Since the emission policies tend to decrease emissions in Europe and in the North American continent, the pseudo-climatological conditions used (corresponding to 2004-2008) will not reflect the possible changes from 2000-2003 to 2004-2008. The use of the mean over 2004 would likely be closer to the years 2000-2003 than the mean of 2004-2008. Since*

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regional model results depend partly on the boundary conditions used, it might be one of the sources of uncertainty of the model results that could be discussed.

It is true that the emission control policies recently led to decreases in emissions in Europe and in North America, but boundary conditions also depend on the meteorology. Therefore, we believe that taking a mean over 5 years (2004-2008) is more robust than taking 2004 to generate the pseudo-climatological conditions for the previous years. Nevertheless, we agree that this is a source of uncertainties and we now mention it in the text.

5. *Section 2.2, page 482, comments on figure 2e and 2b: there is a large gradient on sea salts and organic matters in the North West part of the model domain. This indicates significant differences between MOZART boundary conditions and Polyphemus/Polair3D. How do you explain these differences ?*

Since Figure 2b refers to sulfate, we discuss sea salt (Figure 2e), sulfate (Figure 2b), and organic matter (Figure 2f) below. The sulfate gradient in the north-western part of the model domain in Figure 2b results in part from the choice of colors and concentration intervals in the caption. These parameters have been changed (see Figure 1 below) and we see that there is a general north-south gradient and an ocean-land gradient, which seem realistic. Therefore, we do not think that the MOZART-4 boundary conditions are creating an unrealistic sulfate gradient. The sea salt gradient in the northwestern part of the model domain in Figure 2b is a consequence of two factors. On one hand, sea salt emissions are high in the northwestern part of the domain (see Figure 2). On the other hand, sea salt emissions in MOZART-4 are computed online using the parameterization of Mahowald (Mahowald et al., J. Geophys. Res., 111, D05303, 2006), whereas sea salt emissions in Polair3D are computed offline

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in the preprocessing of Polyphemus, using the parameterization of Monahan (Monahan et al., Oceanic whitecaps and their role in sea-air exchange processes, Dordrecht, p.167-174, 1986). The differences in these two parameterizations contribute to explain the spatial gradient. Figure 2f shows an ocean-land gradient for organic matter. Since both anthropogenic and biogenic emissions are higher on land than over water due to the presence of vegetation and human activities, this gradient seems realistic.

6. *Section 3.1, page 484 first paragraph: This paragraph describes the sources of uncertainties in the observational data. An estimation (even rough) of these uncertainties would add useful information to the evaluation.*

Artifacts associated with nitrate and ammonium measurements occur due to evaporation (or condensation) of semi-volatile ammonium nitrate from the particles collected on the filter due to fluctuations in temperature and relative humidity and/or pressure drop across the filter, which perturb the gas-particle equilibrium. In California, these uncertainties have been estimated to be up to 30% (Herring and Cass, 1999); however they could be less in Europe where most ammonium nitrate formation occurs during the cold season. An unbiased uncertainty of 15% has been reported for nitrate measurements in Europe (Schaap et al., Atmos. Environ., 38, 6487-6496, 2004); this is now mentioned in the text.

7. *Sections 3.2 and 3.3: These sections provide comparisons with existing evaluations of other model simulations. The authors compare their statistics on 9 years (only on July August for AQMEII comparison) over the few stations they selected to statistics obtained with different sets of stations and in different time periods. This is not obvious why the authors have chosen this strategy. To be more meaningful, these comparisons could have been done on the same periods as*

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used in the other models and on a larger set of stations (closer to what was used in the other model simulations).

The model used here, Polyphemus/Polair3D, was part of the AQMEII project and has already been compared to other models (Sartelet et al., Atmos. Environ., 53, 131-141, 2012). Therefore, we did not repeat this specific comparison here (we now mention this point in the text). However, we believe that it was important to ensure that model performance did not degrade when simulating longer time periods, hence our comparison between our multi-year simulation and the AQMEII results. We clarify this point now in the text.

8. *Section 3.3: It would be useful to have the number of observations used to evaluate the 4 one-year simulations.*

Done.

9. *Section 4.2: Two indicators are used in the dynamic evaluation: correlation and regression coefficient. I assume that is a linear regression which is used. This information is needed. If this is a linear regression which was chosen, this does not fit the variations of the correlation with the lag which are not linear as expected.*

This is a good point because some plots of the correlation versus day lag indeed do not show linear relationships over 10 days. Since we show in the paper that the variations of PM_{2.5} as a function of the day lag is not significant if the day lag is greater than 3, we re-performed the linear regression for a day lag ranging from 0 to 2. On this day-lag interval, the variations of the correlation as a function

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of the day-lag fit a linear regression analysis.

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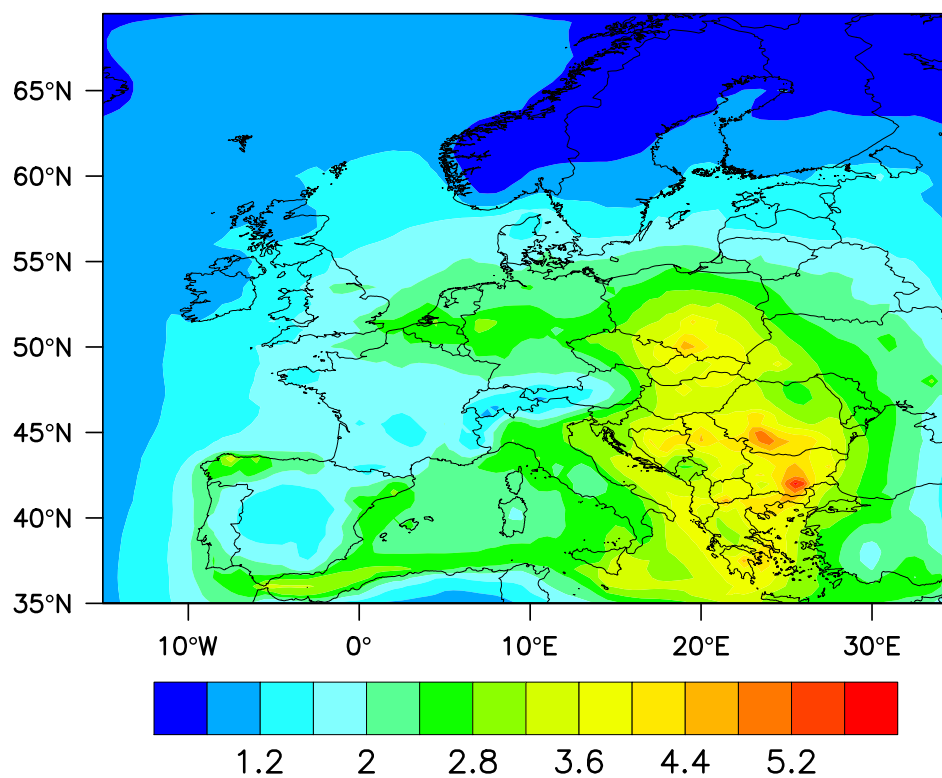


Fig. 1. Nine-year (2000-2008) averaged concentrations of sulfate, expressed in $\mu\text{g m}^{-3}$.

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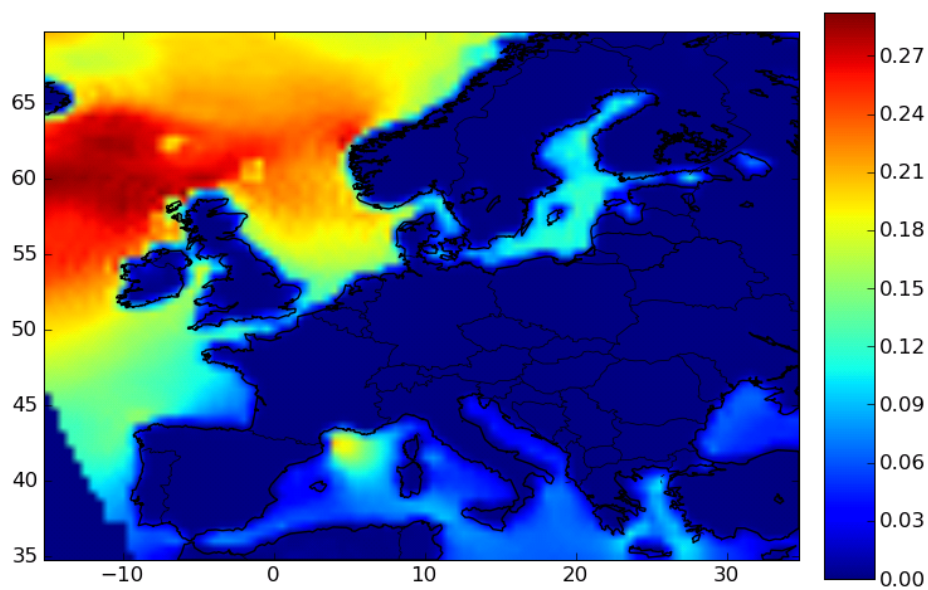


Fig. 2. Surface sea-salt emissions, expressed in $\mu\text{g m}^{-2}$.

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