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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The aerosol distribution in Europe derived with the Community Multiscale Air Quality (CMAQ) model: comparison to near surface in situ and sunphotometer measurements" *by* V. Matthias

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The paper discusses the use of the EPA's Models-3 CMAQ (Community Multiscale Air Quality) model applied to Europe and, at a higher resolution, to the North Sea with particular reference to comparing "ground level" PM10 and aerosol optical depth (AOD) between the model and various measurement facilities. Overall, it is good to see work published relating to the application of the US code to the EU. The general approach appears sound and there is a large amount of statistical comparisons between ground-based measurements and model predictions. The main message is that, in these simulations (which use suitable inputs for Models- 3 CMAQ), the model always

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underpredicted total aerosol mass and thus AOD, the author suggests due to under representing organic aerosol (perhaps, partially, at the emissions inventory stage). It is a little worrying that these Models-3 CMAQ simulations are giving consistent underprediction of aerosol mass (of up to 60%) near the ground. Is this seen with the numerous other CMAQ simulations? And do other models, for the same domains, give better or similar results? I'd like to see comparisons made with both these.

A discussion with comparisons to other CMAQ results in the US has been added. In Europe the focus is on PM10 while it is on PM2.5 in the US, therefore the results are not always comparable. It is easier to compare the results for the inorganic aerosol components nitrate, ammonium and sulfate, where CMAQ's underprediction of aerosol mass in Europe is not that severe. Possible reasons for the underprediction of PM10 can best be discussed at Melpitz and Birkenes, the only two stations where the five most important inorganic aerosol components have been measured for a complete year. This has now been done more extensively in the text (section 3.3, line 629, line 660, sec. 4, line 886).

I note that the author refers to previous work to justify using only the dry aerosol mass in this paper. It would be interesting to include in this paper, reference to a statistical comparison also of the wet aerosol mass.

A short discussion is added in section 3.2 (line 392).

Whilst the paper is generally well written and contains much useful information, it would greatly benefit specifically from including maps showing the two domains and the position of the monitoring stations (Tables 1 & 2), eg overlaying measurements. It is not clear how the 2 domains are used and why most comparisons are on the coarse domain.

I added a map of the stations (new Figure 1) that shows their position. Symbols denote the different quantities that are measured at the individual sites. I hesitate to include

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more maps with results to keep the number of figures and tables limited and because the number of stations is usually too low to construct colour maps of the measurements. Most comparisons are done in the coarse domain because more stations are covered by this domain. The inner dommain is important for future investigations about nitrogen and B(a)P input into the North Sea. I added a sentence in the model description section to clarify this. Because the comparisons in the inner domain do not yield very different results to those in the coarse domain, they are not put into the focus of this study.

In places more references are required and generally the paper would benefit from closer proof reading - the interchange between using measurement station names and a shorthand key is confusing; there are many mismatchs of references to figures and tables and the actual figures and tables; some figures are missing and others are inadequately labelled; improved English spelling and grammar (less use of "also"!).

More references, particularly from work that has been done in the US have been added (section 3.3). The use of “also” is now less frequent and a closer proof reading will be done in the next publication step. All measurement station names always include the shorthand keys.

There is no “future/further work” section.

I added a paragraph at the end of the conclusions (line 955).

Some specific comments:

2.1 - Chemistry transport model The version of CMAQ being used should be made explicitly clear as early as possible both in Section 2 but also in the Abstract. The paper is based on version 4.5 but there are more recent versions available - it should be explained why v4.5 is most appropriate for this study.

It has been mentioned earlier now (in the abstract) and it has been explained that ver-

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sion 4.5 contains a sea salt parameteriasation, an important feature for PM10 studies in coastal regions (line 121).

The description of Models-3 CMAQ would benefit from explaining that it uses 3 modes for aerosol, that these presume logNormal mass-size distributions (for each mode) and that not all species are modelled in each of the 3 modes. Indeed, throughout the paper there seems to be no mention of CMAQ's Aitken mode - surely this has not been ignored?

The description has been expanded to explain those facts (line 104). The Aitken mode has been included in the comparisons although its contribution to the total mass is usually small (1-3 %). That's the reason why it is not been extensively discussed.

Model set-up: it's implied but not stated explicitly whether the new PAH method is used. I presume it is used but there is no discussion of how this addition scheme affects the model results - a significant omission.

Yes, CMAQ is used with the new PAH scheme and their contribution to the total aerosol mass is included in the PM10 results. The influence on the results presented here is negligible, because their concentration is about a factor of 10^4 lower than the total PM10. See line 139.

Model set-up: is the 18 km resolution grid "nested" in the sense that data from the 54 km grid is used to provide initial & boundary conditions (if so this should be clearly stated) or in the sense that it's a completely separate simulation but the domain happens to cover a geographical subset of the 54 km simulation?

It's a nested run and this was already stated in section 2.1 of the manuscript.

3.2 - PM10 results It is not explicitly stated whether the use of PM10 and "total aerosol mass" are interchangeable in these simulations - the version of Models-3 CMAQ used

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uses a modal representation of aerosol mass so summing the masses of aerosol outputs would not neccessarily be equivalent to PM10.

Yes, summing all CMAQ modes is not necessarily the same as PM10, nevertheless I treated them to be the same. This could result in a slightly larger underestimation of the total aerosol mass because PM10 is usually somewhat lower than total suspended matter (TSP). This is now explained in the paper (line 396). The effect on the results will be small, because typically 90 % of the modelled aerosol mass is in the I and J modes (representing PM2.5).

I note that the model output for "ground layer" is, as expected, the lowest model layer. It would be interesting to explicitly state the value of the top of this (terrain-following) layer at each measurement site and further work could involve higher resolved layers near the ground to investigate whether this improves the simulations. The author should clarify why it is more appropriate to use these data than the model's dry deposition values.

I'm sorry but I don't understand what is meant with the last comment about the model's dry deposition. Why should it be used to be compared with concentration measurements?
The lowest layer is typically 36 m thick, representing a σ-level of 0.995. This is now mentioned in section 2.1 (line 154).
The effect of the layer thickness and the number of vertical layers was already tested with less vertical layers (9 and 12 layers). This led, as expected, to lower concentrations in the lowest layer. Higher resolved layers would likely lead to increased concentrations in the lowest layer. It is beyond the scope of this paper to investigate this effect in more detail.

It is also unclear the difference between the 54 and 18 km resolution domains (see earlier comments), particularly why the focus of the comparison in on the 54 km resolution

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domain.

It is now stated in section 2 that the 18 km grid is setup over the North Sea because future model runs will be done to investigate the atmospheric input of pollutants into the North Sea (line 142). The focus of the comparisons in this paper is on the 54 km grid because the transport of pollutants from whole Europe into the North Sea region is of utmost importance for their deposition in this region. Because our work at GKSS aims at multi-annual runs it is computationally too expensive to set up a 18 km grid for whole Europe.
Additionally, more EMEP measurement stations are available in the 54 km grid that allow comparisons to measurements in different regions. The model results of the 18 km grid are only briefly discussed in the paper because they do not give additional information, particularly the model performance is not significantly better in the 18 km domain. This is stated in the text (section 3.2, line 493).

The statistical analysis would benefit from a brief introduction explaining the purpose, physical meaning and limitations of each statistical measures. Specifically, the number of points in the sample should be stated (eg Table 3); and eg (relative) standard deviation is the spread of daily means with respect to the annual mean and the comparison of this measure between the model and the measurements says little concerning the model's ability to capture the time evolution of aerosol concentrations, although it does indicate that the range of values is broadly similiar. Given the low correlation coeffs (Table 3) I would ask the author to give the time-series figures earlier in his discussion and he should consider providing (a summary of) such time-series for all monitoring stations and also on 18 km resolution.

In my opinion, these statistical measures are so basic that almost all interested readers should know them. If not, it is very easy to look them up. What these measures mean for the comparisons is given in the text. I added the number of points in Table 3 and gave the numbers for Table 5 in the Table caption. I explained in the text why I added

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geometric mean and geometric standard deviation, as proposed by Reviewer 2, to the Tables.
A reference to the time-series figures is now given earlier, but I hesitate to show all time series as the reviewer proposes. These Figures would then be very small and it would be impossible to learn more from them than the summarizing numbers in the Tables. I prefer to give two examples in larger size, so that some of the details can be captured by the reader. The correlation coefficients are low at some of the stations but in the range of values that can be expected from other model results (eg Kahnert, 2004). The time series at DE07 and CH02 that are shown indicate that the low correlation coefficients arise from the fact that some of the higher PM10 values, particularly in summer, are not represented in the model results. This is now stated in the text (line 421).
The statistics of the monitoring stations in the 18 km grid are discussed in the text when it was necessary. At most of the stations the results do not differ significantly from the results in the 54 km grid and therefore the numbers would duplicate results that are already given.

The discussion concerning log-normal distribution of aerosol mass needs a sharper focus, for examples: i) why do we expect a single logNormal dist - what about the Aitken (or coarse) modes? ii) "a" in the Table 4 (etc) needs defining iii) it would be useful to state, explicitly, which values of chi^2 and "a" are required to pass the given statistical tests to remove the need for the reader to use look-up tables. Currently it's very hard to interpret these tables.

There is some confusion here about the relation between the log-normal size distributions and the log-normal distribution of the probability to find a distinct particle mass concentration in the atmosphere. I think there is no relation. The size distribution could also be gamma or normal, we would also find a log-normal distribution for the daily means of the PM10 mass concentrations. In my opinion, the fact that we find such a distribution is related to the changes in the meteorological conditions.
"a" in the Table 4 has been changed to "KS" as proposed by Reviewer 2.

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The values for χ^2 and "KS" (former "a") are now given in the text (line 482).

The discussion concerning the discrepancies between model and measurement values seems to be implying that a higher resolution works better but yet the figures provided do not justify this. Indeed, the same statistical analysis should be undertaken for the finer grid as for the coarse grid so that the reader can make a valued judgement.

A higher resolution only works better if the measurement station is in inhomogeneous terrain (eg if it's a coastal site or a mountain site). This is clearly said in the text. However, the differences in the statistical parameters, except for the bias, are small. No additional information would be provived if they would be given in an additional table. On the contrary, this might confuse the reader because the paper already contains 10 Tables. It is said in the text that no improvement in terms of better statistical parameters (except the bias) of the comparisons is achieved with the finer resolution (line518).

3.3 Chemical Composition Rather than looking at two sites in detail and then summarising briefing other sites' data, I would outline all available data, plot all species (inc total aerosol mass) as timeseries (as well as table of stats) and then discuss en masse rather than separateout two sites for special attention. (Or an explicit case should be made not to take this approach.)

It would be too much to plot all species for all sites. These two sites have been chosen because they are the only sites where 5 inorganic species (sulfate, nitrate, ammonium, sodium, chloride) and PM10 were measured simultaneously for a complete year. Therefore, these two stations allow to investigate the possible reasons for the PM10 underestimation in the CMAQ model results and this has been done here. I explained this in the text (line 561).

Measurement stations should also be named using their shorthand key for ease of comparison with earlier tables/figs).

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Shorthand keys are now used for all stations.

Any comments on why nitrate is not captured well at Birkenes, esp. in comparison to Melpitz?

This is not easy to answer. What can be said is that in summer at Birkenes the model results show a clear shift from particulate nitrate to HNO_3. This shift, that can also be seen at other stations, is more pronounced at Birkenes than at other places. At this stage it is hard to say what the reason might be and it is beyond this paper to investiagte this in more detail.

Fig 6, which may have addressed some of the above points, is a repeat of Fig 5.

This was an error that occurred while the manuscript was processed at the Copernicus Office. It will be corrected in this version.

"Not specified aerosol" is vague - which of CMAQ species are the authors referring to (and what are the initial/boundary conditions for these species and how are they treated within CMAQ)? Given that CMAQ models aerosol water as a separate entity, although the author has decided to model only dry aerosol, how can the "not specified aerosol" be partly water? (Depending upon how the author addresses this point, amendments will also be required to the Conclusions.)

"Not specified aerosol" is the difference between the total mass (or PM10 here) and the specified components sulfate, nitrate, ammonium, sodium, chloride. This is now better explained in the text (line 619). Concerning the measurements this may include water although the filtersamples are weighed at RH = 50 % and it is assumed (by the data providers) that they are “dry”. Therefore it can be partly water if we speak about measurements. In the model results, water is excluded, and “not specified aerosol” means everything except water, sulfate, nitrate, ammonium, sodium and chloride.

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What is the magnitude of the organic aerosol and EC at the sites and does this difference wrt model help account for the model's systematic underpredictions?

Unfortunately, there are only very few measurements available that cover a longer time, therefore OC and EC could only be evaluated for NO01 in 2001. Here, the model underestimates the measurements by a factor of 3 (measured mean (OC+EC) 1.1 μgm^3, modelled 0.4μgm^3). Certainly, the underpredicted OC and EC is a large fraction of the "missing" aerosol, but it is difficult to say how large that fraction is. Even if OC and EC were measured, the measurements usually still have a large gap between total mass and the sum of the species. This might in large parts also be organic aerosol. Therefore the model's underpredictions might be even larger than the factor of 3. This cannot be said more precisely because measurements were mising at the other sites.

3.4 Aerosol Optical Depth
No mention of AOD being function of wavelength. How is equ (1) amended to take account of this?

Equation (1) should be valid in the mid visible. The AOD from the Aeronet measurements was taken at 500 nm. This is added in the text (line 697) and in the table caption.

It would be useful to plot the Aeronet measurement stations on a map to get a bearing of their positions relative to the other measurement stations eg given AOD dependence on aerosol mass and composition, are we looking at the same data in a different manner, or data at very different sites? The former would allow, for example, an evaluation of the Malm approach.

The stations are also plotted in the new Figure 1. As well as the PM measurements, they represent a Central European data set, but with a focus on France. Some of the stations are very close together (eg. Melpitz and Leipzig). In fact, one could try to evaluate the Malm approach based on the measurements but this needs at least some information about the PBL height. Additionally one has to assume that the PBL is well

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mixed and that no additional layers aloft influence the results. It will be a separate, but certainly interesting study.

I did not follow the explanation of why Clermont Ferrand's simulated AOD is an overestimate - surely all modelled AOD data include contributions from all model layers? Given the importance of correct RH, it would be interesting to know whether the model is using similiar RH values as to those recorded by the measuring stations (but we appreciate this is a test of the uncoupled met model).

The model terrain height in Clermont Ferrand's grid cell is lower than the measurement station's altitude, because of the coarse grid resolution. The modelled values are therefore not representative for a mountain station that in many cases will be less polluted than other places in the valleys (e.g. because CLE is above the PBL in the morning hours).
It has been checked before whether the meteorological input compares well with observations. On average this is the case, the results are submitted to Env. Fluid Dynamics in a separate paper (line 268). The MM5 results tend to overestimate RH by about 10-15 %.

It is generous to say from Fig 7 that for Lille the model's 6 wks' average represents the measurements. I cannot comment for those plots not shown - if they are relevant they should be shown. Perhaps replotting with diff wrt average (for each of model & measurement) will be a better illustration?

Lille is shown in Figure 7 and the reader can judge the comparisons. Again, I think that it is better to show two examples that are typical for the results of the modelled and measured AOD than to show all of them in a rather small format. The discussion can also be follwed if not all plots are given.

Need to expound, or cite other work, re statement that biogenic emissions are underestimated in the model.

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Primary biogenic particles are not included in the model and also not in the emissions. This is written in the text (section 3.3, line 623). The amount of secondary biogenic aerosols must be considered as very uncertain because here only monthly average values from a global data base were taken. It is the common opinion in the scintific community that secondary organic aersol formation from terpenes and isoprene is likely underestimated in current SOA formulations (Yin et al., 2004, Fuzzi et al. 2006, Kanakidou et al., 2005, the papers are now cited in the manuscript).

The text refers to Kishinev in Table 10 but it's not present - need to add to Table.

Kishinev is the same as "MOL" in the table. The short hand key was changed to "KIS".

Would the author care to comment as to why the model fails to produce a similiar frequency distribution for AOD as to that measured (Fig 8)? Fig 8 should use same axes in both plots for ease of comparison.

The model fails to produce the high AOD values and one possible explanation is that strong Saharan dust events are not captured by the model. Saharan dust is often lifted in very high altitudes and then the dust can be transported over long distances and contribute significantly to the AOD in the Mediterranean region. This is hardly reproduced by the model because the model study is not designed to treat these events correctly, particularly on the emissions side.
In Figure 8 the different scales are used for a better comparison of the distribution funcions despite their different medians. If the same scale would be used, the main differences in the distribution function would not be so easily visible. Therefore I would prefer to stay with the different axes ranges.

4. Conclusions
Whilst mention was made that the model performed less badly (re PM10) in Winter in section 3.2 it was only for the German station(s?) (and not so for Switzerland & Austria) - this should be clarified in the Conclusions.

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It has been clarified in the conclusions (line 833). In Austria and Switzerland large deviations between measurements and model results also appear in winter.

The discussion of Saharan dust should be not be introduced in the Conclusions - a mention must be made earlier (3.4). It may be useful, as future work, to use a larger (perhaps coarser) domain that includes North Africa to provide temporally variant boundary conditions for the 54 km resolution domain capturing dust events.

The Saharan dust problem is now also discussed in section 3.4 (line 810). In my opinion an extension of the domain would not be sufficient. It would make more sense to take BCs from a dust model or to include algorithms that represent wind blown dust in CMAQ (maybe similar to what has been done for sea salt).

The Conclusions' firm assertion that the AOD variablity is "correctly produced" by the model appears to contradict Section 3.4's discussion of Fig 8. The Conclusions should be amended to account for this.

An explanation has been added that the log-normal distribution function is not followed by the data taken at Avignon (line 802).

Some specifics: Fig 1: which resolution is this or is it a mixture of resolutions? and why has the plot been cropped (as illustrated by the x-axis and y-axis range not starting at 1)?

The figure is in 54 x 54 km^2 resolution. It has been cropped to exclude boundary effects from the figure.

Section 3.4 - refers to equation 2 but presumably it means equation 1 and the reference to Table 4 is misleading.

I'm sorry for this. It's a latex processing error.

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Thanks to Michael Bane for his precise comments. It really helped to improve the manuscript and make it better readable.

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