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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosols in the CALIOPE air quality modelling system: validation and analysis of PM levels, optical depths and chemical composition over Europe" *by* S. Basart et al.

Anonymous Referee #2

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Referee Comment to the paper:

Aerosols in the CALIOPE air quality modelling system: validation and analysis of PM levels, optical depths and chemical composition over Europe

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The paper presents calculations and evaluation of a recently developed modelling system CALIOPE, focusing on aerosols. CALIOP integrates CMAQ (AERO4) model, which calculates anthropogenic, biogenic and sea salt aerosols, and BSC-DREAM8b model accounting for mineral dust from North African deserts. The calculations were

performed for Europe for 2004 and compared with observations from EMEP and CREATE databases. The average correlation between calculated and measured daily PM10, PM2.5 and AOD is 0.57, 0.47 and 0.51 for all sites. The model is shown to considerably underestimate concentrations of PM10, PM2.5 and individual aerosol compounds, as well as AOD. In particular, the model was applied to assess the impact of African dust on air quality in southern Europe.

The paper deals with a relevant subject of chemical transport modelling on a regional scale, with a particular focus on PM. In general, the paper is reasonably well structured and in general easy to follow, though there are also some unclear formulations or omitted information in the test (some of those are commented below). The work can be considered for publication in ACP after some revision of the paper. In particular, the language of the paper should be considerably improved, including correction of typos, checking on work usage and re-formulating unclear statements.

To my opinion, the paper shows too little effort to investigate the reasons and to explain the large underestimations of concentrations of PM10, PM2.5 and practically all individual aerosol compounds. 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 CALIOPE responsible for the underestimations, and suggest ways to improve those (in addition to listing general known uncertainties (e.g. p. 20587, 17-28)). Furthermore, it could be recommended to use for model evaluation some later years, for which much more data on PM concentrations and chemical composition are available (see for ex. same website www.emep.int). That would facilitate a more profound evaluation of model performance and gaining better insight in the nature of modelling inaccuracies.

The points to be considered by the authors: 1. Concerning calculated dust from North African deserts, the CALIOPE covers only very northern parts of Africa. It is unclear from the paper whether any boundary conditions are implemented to account for dust fluxes from the rest of Sahara? Also, it appear from the paper that the CALIOPE is the

only model in Europe which “includes a non-climatic representation of Saharan dust transport” (p. 20578, 10-12, p. 20598, 11). What about for example CHIMERE model (Menut et al., 2009, JGR., 114, D16202), or EMEP model (EMEP Report 4/2004 – 4/2011) on http://www.emep.int)?

2. In the paper, higher correlations of calculated PM10 with observations compared to calculated PM2.5 are explained by accounting for natural dust (e.g. p. 20576, 12-14). However, the model calculates considerable contribution of dust to both PM10 (25%) and to PM2.5 (as much as 20%) (p. 20587, 14-16). Please, explain why inclusion of natural dust improves model results for PM10, but not for PM2.5.

3. The paper finds large discrepancies between CALIOPE calculated and observed concentrations of carbonaceous aerosols. The associated discussion indicates uncertainties in emission and in SOA modelling as main reasons for that. In this regard: (1) Any description of emission data for EC and primary OC and their source is missing in the paper. They are not included in EMEP and as far as I can see not covered by Baldasano et al., 2008. Further, the reasoning about significant model underestimation of EC and OC (p.20594, l. 13-16) is very general and vague. As no information on the source of EC/OC emissions is provided in the paper, the given discussion on emission uncertainties becomes groundless. In addition, Schaap et al. (2004) did discuss BC emission uncertainties, but those of much older inventories. (2) contradictory to what the paper says about typically great underestimation of EC/OC by regional (p. 13), several modelling studies showed quite good agreements with observations at Birkenes and Melpitz (even some overestimation) (e.g. Hallquist et al, ACP, 9, 2009; Simpson et al., JGR, 2007; Tsyro et al., JGR, 2007). For EC, is it possible that the CALIOPE calculates too short life-time? Does the model account for EC ageing and the changes in hygroscopic properties and thus in wet scavenging?

4. The paper estimates that deserts dust causes daily exceedances of the PM10 European air quality threshold (50 um/m3) for more than 75 days in 2004 in the areas south of 45N. How does that compare with observed exceedances?

5. On p. 20595 (l. 8-10) the authors recognize that using one and the same for the whole Europe correction factor to concentration fields is rather crude method. However, it appears from the text that the improvement obtained for PM and AOD in this case justifies the method. Would not it be more appropriate to apply spatially variable (though annual mean) correction, taking in to account the geographical differences in model performance?

Other comments

Abstract: p. 20576 (3) – explain what “1h” is; and also on p. 20582 (20) line 8 (and through the paper) – I think it is more correct to talk about aerosol components than PM chemical composition (as the measurements were not necessarily done at the same sites, and the samples for chemical analyses were most likely collected with filter-packs without any defined cut-off size). lines 10-12 (and other places) : . . . the correlation between model calculated and observed PM10 and PM2.5 lines 15-16: underestimation of measured concentrations by the model (not overestimation of the modelled ..); “particularly” is redundunt; line 20: should be either PM10 or PM2.5 mass instead of “aerosol budget” line 21: “aerosol concentrations” meaning all of the individual aerosol components? Line 22: “High values” of what?; Line 23-24: should be “particles which contribute”; What is “total aerosol mass”? Total suspended matter? Line 24: maximum seasonal dust concentrations Line 27: “reaching up to more than 75 days” is not a good language

Introduction: p. 20577, 1: Do you mean by “inhomogeneous” something else than “variable in space and time”? Explain then, please. Line 10: Why cursive? Line 11: “may have been transposed” ? Line 12: introduced; Line 20-21: what about windblown dust from European semi-arid areas and bare fields? Should be “ contribute to occurrence of (or cause) those enhanced levels” Line 22-23: “Air quality models are useful .. to manage air quality” - not a good statement; p. 20578, line 2: Please give a reference about “23 modelling systems...” line 13: should be “confidence in” line 20: Should be “underestimated by a factor of”

p. 20579, 19-22: Please explain what is meant by the last sentence, starting with “As CALIOPE is a fundamental system..”. Some thin else than just the same version of the model was used in Pat et al. and the present work? It is also confusing as calculation results are indeed “bias” corrected later in the paper. Line 10: dust aerosol instead of soil; Line 18: correct to “SIA are generated by... processes and include/consist of nitrate ...” p. 20581, the explanation to eq. 2 says that both Organic Mass and Unspecified portion of PM2.5 are included, whereas p. 20580 (11-12) says that the unspecified PM2.5 includes non-carbon atoms associated with Organic aerosols. p. 20582: eq. 3: Should it be 3/(4*PI*Ro*r3) ? p. 20583: please explain more clear about meteorological driver and boundary conditions, for which meteorological parameters/ chemical species the boundary conditions are used.

P. 20583, l. 22-23: “model calculated PM concentrations are compared” or “model output is compared”; P. 20584, l. 2-3: The last sentence “Details on the location...” with reference to Pay et al. needs clarification. Does not Table A1 present sites’ details. Also the analysis of results is presented in this paper. l. 4: Should it be “Modelled aerosol concentrations of aerosol species ..”? l. 6: What is meant by “aerosol mass”? Which aerosol component? Or PM? l. 9-10: Positive measurements artefacts should also be mentioned (gas condensation on filters) l. 11-12: I suggest to re-write the sentence “Inorganic species may be ...” as e.g. SIA components can be measured with uncertainty of about +/- 10% (Putaud et al., 2004). Also, it should be made consistent with text and reference on l. 16-17, as EMEP measurements are also based Ion chromatography method. l. 18: Explain again “aerosol concentrations and aerosol mass are available...” l. 21-23: consider re-writing as “.. 53 for sulphate, 27 for nitrate, etc.”

P. 20585, l. 6-8: Please, re-write more clearly; l. 14-15: move “in 2004” after “30 h” l. 17: “localizations” ?? l. 20: check on “data from all of the 440-870nm wavelength range..”

p. 20586, l. 4-6: could the authors please explain the difference between “model performance goal” and “model performance criterion” l. 14: as before “aerosol concen-

trations and aerosol mass (?) ...” l. 20: suggested “Because the differences between simulations and observations are small. . .”

p. 20587, l. 1: what is meant by “general trends”? l. 4: suggested “daily time series” instead of “evolution of time series” l. 5-9: remove “Although”; suggested to be more precise instead of using “moderately well” (also on p. 20592 line 15) and also “to a slightly larger extent underestimated” l. 22-28: Total particulate mass, discussed here, was not considered in the present evaluation at all. What about SIA? Could the authors point the main reasons for the model’s underestimation of SIA? The explanations to model’s PM underestimation are rather general. Could the authors outline the specific deficiencies and uncertainties associated with model results in the present work?

p. 20588, l. 12-14: recommended to re-write about the results for correlation, dropping “as much as PM2.5”; l. 15-16: are not characteristics “low air renovation” and “favoring the regional mixing” somewhat contradictory?

p. 20589, l. 3-5: The emissions of ammonia, a gaseous precursor of ammonium nitrate, are very uncertain. This problem is definitely worth discussing; l. 16-17: recommended re-writing the sentence (poor language) l. 22: suggested “For sea salt components..”

p. 20590, l. 6-7: please explain “Transfer from PM10 to PM2.5 is not considered in AERO4 EITHER”; l. 8: According to observations, EC can contribute significantly to PM2.5 at kerbsides (with 17% on average as in Putaud et. al), but it is not “the major component” of PM10 and of PM2.5 otherwise.

p. 20591, l. 7-8: repetition l. 19-20: Do the authors really mean that the current model performance for AOD is satisfactory?

p. 20592, l. 13: correct “being” to “were” or “are”..

p. 20583, l. 17: consider to change “the timing of sudden increases” by something like “reproduce the occurrence of enhanced (or peak) concentrations (or episodes)” l. 23:

Please explain what exactly “challenges models”. Do the authors refer to any specific process, which the CALIOPE fails to model soundly?

p.20594, l. 13-16: Could the authors suggest any specific type of local or natural emissions responsible for the EC underestimation presented here? One of the main sources of EC emission underestimation which is often discussed is associated with uncertainties in residential combustion, especially wood burning. However, this cannot explain large EC underestimation in the summer of 2004. Neither can “cold start” of motor vehicles. l. 25: what are “different aerosol fractions”?

p. 20596, l.9: suggested to change “aerosol fractions” to “ PM concentrations and AOD”; l. 13: remove “To a lesser extent”

p.20597, l.13: correct Romania l. 27-29: Not a very good statement. Indeed, calculations of EC and OC are still associated with considerable uncertainties; they are not just the same for both components and therefore should be explained individually. Also, there has been an increasing amount of EC/OC observations available for model evaluation in the last decade.

p.20598, l. 13: should be “non-climatological basis”

p. 20600, l. 1-2: very imprecise “missing aerosol sources, which affect these large fractions”. Do you mean some missing sources of coarse PM? l. 25: “ contributors to the aerosol mass budget”? or to PM2.5?

l. 12-13: the SOA formation is repetition of the on line 10. Could you please explain concerning “the dynamic interactions between fine and coarse aerosol”.

p. 20618, Table B1: check the formulas for MNBE (1/n is missing), MFB (should be [c+o]/2 in denominator), and MFE (should be |c-o| in numerator).

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