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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.

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On behalf of all the co-authors I would like to thank the referee #2 for his/her constructive comments on the manuscript. Overall, the referee recognizes the interest of the manuscript in the context of this the paper deals with a relevant subject of chemical transport modelling on a regional scale, with a particular focus on PM. The manuscript has been revised after the referee's comments in order to introduce the suggestions for improving the quality of the paper. A revision of the manuscript has already been sent to the Editorial Office. Please find hereafter an item-by-item response following to all the statements of the referee.

Referee #2: [. . .] 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.

Authors: The year 2004 has been selected because is the year selected under the CALIOPE project (www.bsc.es/caliope) to make the model evaluation studies. The selection of this year is linked to the fact that for the Iberian Peninsula domain the CALIOPE modelling system works with a bottom-up inventory based on 2004. We agree with the reviewer, and currently, we are working on updating the emission inventory for a more recent year, as well as for improved emission disaggregation techniques.

Referee #2: 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 appears 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?

Authors: The domain of simulation of the BSC-DREAM8b model covers North Africa, Europe and Middle East (the box at 0-65°N and 26°W-60°E). The BSC-DREAM8b is run offline and the outputs are then added to the CMAQ-calculated PM (see Jiménez-Guerrero et al., 2008). Since BSC-DREAM8b used a 50 km x 50 km horizontal resolution, its outputs are interpolated to the CMAQ's Lambert conformal conic grid with a 12 km x 12 km horizontal resolution in order to add the corresponding aerosol components. After the interpolation, the total modelled PM2.5 is the sum of the Aitken and Accumulation aerosol species from CMAQ and the corresponding bins with diameter less than and equal to 2.5 μm. In the same way, total modelled PM10 is the sum of the

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Aitken, accumulation and coarse species from CMAQ and the corresponding bins with diameter less than and equal to 10 μm. It is true that the CALIOPE modelling system is the only air quality modelling system for Europe that includes desert dust contributions in the forecast mode (Balk et al, 2010; Menut and Bessagnet, 2010). On the one hand, EMEP model is not run in forecast mode. On the other hand, CHIMERE-Dust provides daily dust forecast, although the PM10 forecast of the photochemical CHIMERE model does not include these dust contribution. This is highlighted in the manuscripts as follows: P. 20578 Line 10: "In contrast to many other European modelling systems, CALIOPE includes a non-climatic representation of Saharan dust transport in its forecast mode."

Referee #2: In the paper, higher correlations of calculated PM10 with observations compared to calculate 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.

Authors: First of all, we want to emphasize that the contribution is obtained from the annual aerosol budget while the correlation is computed considering the variability of the aerosol concentration along the year on an daily basis. The correlation for PM10 improves with the inclusion of desert dust contribution from the BSC-DREAM8b model. The CALIOPE modelling system is capable to reproduce the episodic and marked seasonal dust transport towards Europe. However, as indicated by the referee #2, these improvements are not reflected in the PM2.5 fraction. In the PM2.5 fraction, slightly lower correlation values are obtained when the BSC-DREAM8b model contribution is included in the CALIOPE system although the mean bias is reduced over all the seasons. As we show in Pay et al. (2011a; accepted in Atmos. Environ.), desert dust events are overestimated by BSC-DREAM8b in the PM2.5 fraction in the Iberian Peninsula while the PM10 fraction is well reproduced as presented in the Figure 1R for the Víznar station in the southern Spain. In this figure, PM2.5 model overestimations are

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associated to overestimations of the BSC-DREAM8b. These overestimations penalize in the annual correlation calculations. Therefore, despite the total dust mass is well captured by the BSC-DREAM8b model; its size distribution tends to overestimate the finer fractions.

This evidence has been highlighted in the manuscript as follows: P. 20587 Line 16: "Although the model calculates considerable contribution of dust to both PM2.5 and PM10, higher annual correlation is observed in PM10 than in PM2.5. This is associated to overestimations of PM2.5 (see Fig. 2f) during desert dust outbreaks in southern Europe sites (Pay et al., 2011a). Therefore, despite the total dust mass (i.e. PM10) is well captured by the BSC-DREAM8b model, its size distribution tends to overestimate the finer fractions (i.e. PM2.5)."

Referee #2: 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. (2004a) 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?

Authors: As the reviewer suggests here we explain more in detail how particulate

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matter (emissions and atmospheric chemistry) are treated in the CALIOPE system, with special focus on OC and EC in order to understand their significant model underestimation. The CALIOPE system considers the anthropogenic emissions of SO2, NOx, NMVOC (Non-Methanic Volatile Organic Compounds), CO, PM10, PM2.5, and NH3 derived from the 2004 annual EMEP emission database (EMEP, 2007). Raw emission data are processed by HERMES-EMEP model in order to provide a comprehensive description of the emissions to the air quality model. The inventory distinguishes the source categories following the Selected Nomenclature Air Pollution (SNAP). Since the gas-phase chemistry is resolved with the CB-IV mechanism, PM2.5 emissions are distributed to elemental carbon, organic aerosol, nitrate, sulphate, and unspecified fine PM based on the emission profiles from SPECIATE 3.2 (http://www.epa.gov/ttn/chief/emch/speciation/cbiv-profiles_mar_4_2002.xls) which are function of the emission sector. Traffic emissions have the highest contribution of OC and EC (80% of PM2.5), followed by combustion in energy and transformation industries (60% of PM2.5). Natural PM emissions and primary biogenic emissions (such as pollen, bacteria, fungal, and fern spores, viruses, fragments of animals and plants, virus, etc.) containing organic compounds, are not included in the emission model, since it is one of the most uncertain aspects in current aerosol research (Monsk et al., 2009; Menut and Bessagnet, 2010). This absence contributes to OC underestimation in particulate matter. Furthermore, the current version of the CALIOPE modelling system does not include wildfire emissions. Wildfire emissions are significantly important in southern Europe during summer (European Commission, 2005). In Table 1R we present the number of fire and burnt area in southern Europe for 2004 in the last 25 years (European Commission, 2005). During 2004, fires in southern Europe burned a total of 346.766 hectares, which is below the average for the last 25 years. The most affect countries are Spain, in the northeastern (Galicia and Castilla Léon) and the southwestern part (Andalucía and Extremadura) and Portugal. Wildfires are an important source of atmospheric pollutants (NOx, VOC, EC and OC) in Southern Europe during the dry season, especially in summertime (European Com-

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mission, 2005). The inclusion of the aforementioned natural emissions could be an important point to improve in order to better reproduce EC and OC in southern Europe during summer. Secondary formation of organic aerosol from biogenic sources is another source of uncertainty which contributes to OC underestimation. The traditional 2-product SOA model adopted from CMAQv4.5 (Binkowski and Roselle, 2003) does not include SOA formation from isoprene and sesquiterpenes. The absence of the isoprene-SOA route on SOA may impact significantly Europe during summer where the predominant vegetation types favour isoprene as the main biogenic VOC component (Keenan et al., 2009). Due to this absence, possible underestimations of SOA can be detected during summer, and consequently underestimation of OC, PM2.5 and PM10 concentrations. As opposite to the results presented in our study, the analysis presented in Tsyro et al. (2007) showed the EMEP model tends to overestimate EC for Birkenes and underestimate EC for more southern sites for 2002-2004. These overestimations were likely linked to EC emissions from residential combustion, in particular from wood burning. In Simpson et al. (2007), which also used the EMEP model for 2002-2003, showed a signiïňĄcant (factor 3–5) underprediction of SOA levels for sites in central-southern Europe. However, the same model showed a very good agreement with total carbon levels at Birkenes site. Differences are linked to differences in the speciation of the EC/OC emissions done in the CALIOPE modelling system as well as wildfires are not included in our modelling system. As the reviewer suggest, we have extended the discussion of the discrepancies between CALIOPE calculated and observed concentrations of carbonaceous aerosols as follows: P. 20590 Line 22: “In the anthropogenic emissions used in the CALIOPE system the primary traffic emissions have the highest contribution of OC and EC (80% of PM2.5), followed by combustion in energy and transformation industries (60% of PM2.5). Additionally, the absence of some natural PM sources (such as wildfire emissions) and primary biogenic emissions (such as pollen, bacteria, fungal, and fern spores, viruses, fragments of animals and plants, virus, etc) contributes to OC underestimation in PM. Wildfire emissions during 2004 were significantly important in southern Europe during summer (Euro-

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pean Commission, 2005). Additionally, the traditional 2-product SOA model adopted from CMAQv4.5 (Binkowski and Roselle, 2003) does not include SOA formation from isoprene and sesquiterpenes. The absence of the isoprene-SOA route on SOA may impact significantly in southern Europe during summer where the predominant vegetation types favour isoprene as the main biogenic VOC (Keenan et al., 2009)." As the reviewer points out Schaap et al. (2004a) used an old inventory (year 1995) and it is not comparable with the present work. In this sense, we have removed the reference to this work in the reviewed manuscript. CMAQv4.5 model implemented in the CALIOPE system is based on the two-product model of SOA formation which does not take into account the aging process which has been observed in the atmosphere and in the laboratory (Jiménez et al., 2009). An assumption of the CMAQv4.5 model is that organics influence neither the water content nor the ionic strength of the aerosol particles. Therefore, changes in hygroscopic properties and thus in removal process are not taken into account (Binkowski and Roselle, 2003). This assumption may be responsible for the errors in modelled OC and EC aerosols. This evidence has been included in the manuscript as follows: P. 20580 Line 21: "Furthermore, an assumption of the CMAQv4.5 model is that organics influence neither the water content nor the ionic strength of the aerosol particles (Binkowski and Roselle, 2003)." Unfortunately, a detailed investigation of the organic aerosol cannot be done for the present study since there are only very few measurements available that cover a longer time, therefore OC and EC could only be evaluated at Birkenes (NO01) and Melpitz (DE44) in 2004 on an annual basis. Here, the modelling system underestimates the measurements by a factor of 4 (DE44: measured mean (OC+EC) 3.21 μg m-3, modelled 0.66 μg m-3; NO01: measured mean (OC+EC) 0.97 μg m-3, modelled 0.23 μg m-3). This cannot be said more precisely because measurements were missing at the other sites. As indicate in P. 20590, l 14, Matthias (2008) also found that modelled OC+EC concentration were underestimated by a factor of 3 at Birkenes using CMAQ over Europe. This factor is lower than that obtained with the CALIOPE system partly because carbonaceous aerosols from biomass burning emissions were taken into account in the

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aforementioned study.

Referee #2: The paper estimates that deserts dust causes daily exceedances of the PM10 European air quality threshold (50 μg/m3) for more than 75 days in 2004 in the areas south of 45N. How does that compare with observed exceedances?

Authors: Next, in Table 2R we include the number of exceedances of the daily PM10 air quality limit value established by the European Commission (i.e. 50 μg m-3) for each EMEP station included in the present analysis for 2004 as well as the number of exceedances obtained from the simulated values. As you can see, the CALIOPE system (CMAQ + BSC-DREAM8b) underestimates the number of exceedances for the year 2004. Particularly for the Spanish stations; most of these exceedances are caused for the desert dust contributions obtained from the BSC-DREAM8b mode (see Table 2R). It is difficult to find temporal series of desert dust observations over Europe. However, the recently work of Pay et al. (2011a; accepted to Atmos. Environ.) includes an experimental data set of African PM10-dust for Spain obtained with the methodology described in Escudero et al. (2007) for the evaluation of the BSC-DREAM8b model. Pay et al. (2011a; accepted to Atmos. Environ.) conclude that the BSC-DREAM8b model is able to reproduce the daily variability of the observed levels of desert dust and most the outbreaks affecting southern Spain. Table 2R has been included in the manuscript as Table 5 as well as the following lines: P. 20599 Line 18: "Table 5 shows the number of exceedances of the daily PM10 air quality limit value established by the European Commission (i.e. 50 μg m-3) for each EMEP station included in the present analysis for 2004 as well as the number of exceedances obtained from the simulated values. As a result, the number of exceedances of the daily PM10 threshold obtained from the 25 EMEP stations for 2004 is underestimated by the CALIOPE modelling system (241 observed days versus 72 simulated days that exceed the daily PM10 threshold). In those stations mostly affected by desert dust outbreaks like the Spanish EMEP sites, these differences are reduced (with 80 observed versus 37 simulated days that exceed the daily PM10 threshold) indicating the improvement that represents the

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inclusion of desert dust contribution in model simulations."

Referee #2: 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?

Authors: The seasonal variability has stronger impact than the geographical differences in the formation of the secondary atmospheric aerosols. As discussed during the model evaluation, some chemical species show a marked seasonal behaviour. This is shown in Table 3 of the manuscript through the estimated factors for each secondary inorganic aerosol component. This is associated to the dependency of some atmospheric reactions with the meteorology. On the other side, a geographically dependent correction factor would involve a more complex approach and is beyond the scope of the paper. This has been included in the manuscript as follows: P. 20595 Line 8: "However, as shown in the model evaluation results, the seasonal variability has stronger impact than the geographical differences in the formation of the secondary atmospheric aerosols. As described below the modelled bulk parameters: PM levels and AOD, significantly improve after correcting the bias of each aerosol species individually."

Referee #2: 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 redundant; line 20: should be either PM10 or PM2.5 mass instead of "aerosol budget" line 21: "aerosol concentrations" meaning all of the

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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.

Authors: The term “1h” is related to the frequency when the model outputs are written in the outputs files. This is modified in the manuscript as follows: P. 20576 Line 1: “The CALIOPE air quality modelling system is developed and applied to Europe with high spatial resolution and output frequency (12 km$\times$12 km and 1 h, respectively).” About the comment related to “line 21: “aerosol concentrations” meaning all of the individual aerosol components?”, aerosol concentrations means all of the individual simulated aerosol components (i.e. carbonaceous matter, secondary inorganic aerosols, sea salt and desert dust) and not only on surface level but at the column-load. This comment and the rest of the suggestions of the reviewer have been introduced in the abstract as follows: P. 20576 Lines 19: “The simulated PM10 and AOD present maximum values over the industrialized and populated areas of the Po Valley and the Benelux regions. SIA are dominant in the fine fractions representing up to 80% of the aerosol budget in latitudes beyond 40°N. In southern Europe, high PM10 and AOD are linked to the desert dust transport from Sahara which contributes up to 40% of the aerosol budget. Close to the surface, maximum seasonal concentrations (PM10 >30 μg m$-$3) are found between spring and early autumn. We estimate that desert dust causes daily exceedances of the PM10 European air quality threshold (50 μg m$-$3) in large areas south of 45°N achieving more than 75 days per year in the southernmost regions.”

Referee #2: 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

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a reference about “23 modelling systems...” line 13: should be “confidence in” line 20: Should be “underestimated by a factor of”

Authors: About the comment related to “Do you mean by “inhomogeneous” something else than “variable in space and time”?”, this is related to the composition of aerosols and their chemical and physical properties. This is introduced in the manuscript as follows P. 20577 Line 1: “inhomogeneous chemical and physical properties” About the comment related to “p. 20578, line 2: Please give a reference about “23 modelling systems...”, the reference is (Menut and Bessagnet, 2010) and it is found in Line 3. The rest of the suggestions have been amended.

Referee #2: 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 Pay 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.

Authors: The CALIOPE modelling system version used for the present analysis is the same than that used in Pay et al. (2010). With the sentence starting with “As CALIOPE is a fundamental system [. . .]”, we would want to emphasize that the original code of the model used in the simulation has not been modified. However, the results of the model evaluation highlighted the necessity to include some corrections in the model outputs in order to give a more better estimation of the spatial and seasonal distribution of the different aerosol components over Europe. These lines have been removed in the manuscript. In relation about the reviewer comments “Line 10: dust aerosol

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instead of soil. Line 10: dust aerosol instead of soil"". The AERO4 aerosol module considers soil as coarse mode soil-derived mass. In the case of the present model simulation, the emissions do not include this specie since the windblown dust emissions are not considered yet. Therefore, the reference to the soil specie is removed in the reviewed manuscript. In the Eq. (2), organic mass (OM) is taken as the sum of all non-light absorbing organic species, i.e., the organic carbon mass from the CMAQ outputs. Therefore, OM has been replaced by OC in the Eq. (2). Details about the meteorological driver and boundary conditions have been included in the manuscript as follows: P. 20583 Line 1: "The Advanced Research Weather Research and Forecasting (WRF-ARW) Model v3.0.1.1 (Michalakes et al., 2004; Skamarock and Klemp, 2008) is used to provide the meteorology to the chemical transport model." P. 20583 Line 4: "WRF-ARW initial and boundary conditions (at intervals of 6 h) are based on the Final Analyses of the National Centers for Environmental Prediction (FNL/NCEP; at $1^\circ \times 1^\circ$) at 12 UTC." P. 20583 Line 8: CMAQ boundary conditions are based on the global climate chemistry model LMDz-INCA2 (Szopa et al., 2009) developed by the Laboratoire des Sciences du Climat et l'Environnement (LSCE). Monthly mean data for the year 2004 are interpolated in the horizontal and vertical dimensions to force the major chemical concentrations (i.e. O3, NO, NO2, HNO3, HCHO, H2O2, PAN and CO) at the boundaries of the domain (Piot et al., 2008). A detailed description of the INteractive Chemistry and Aerosol (INCA) model is presented in Hauglustaine et al. (2004) and Folberth et al. (2006). P. 20582 Eq. 3: the equation included in the manuscript is correct. The extinction efficiency Qext for the interaction of radiation with a scattering sphere of radius r are cross sections ïĄşi normalised to the particle cross section, ïĄřr2, and follows the expression: Q_ext= σ_ext/(πrˆ2) The rest of the comments have been amended.

Referee #2: 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

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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.”

Authors: P. 20584 Line 2: the sentence has been modified as follows: “Details on the location of the EMEP stations used for this comparison can be found in Table A1.” P. 20584 Line 6: as the referee #2 indicates, in this case aerosol mass is related to PM. This has been corrected in the manuscript. As the referee #2 indicates, the following sentence has been included in P. 20584 Line 11: “as well as gas condensation on filters which could introduce positive measurements artefacts”. P. 20584 Line 18: “measured aerosol surface concentrations and aerosol mass” has been replaced by “measured PM10, PM2.5 and their chemical aerosol components (i.e. sulphates, nitrates, ammonium, sea salt and carbonaceous matter)”. The rest of the comments have been amended.

Referee #2: 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..”

Authors: P. 20585 Line 20: “from all of” has been replaced by “in”. P. 20585 Line 8-9: “from which the fraction of fine mode (FMF) to total AOD can be computed obtaining the fine and coarse fraction of the total AOD” has been removed. P. 20585 Line 17: “localizations” has been replaced by “sites”.

Referee #2: p. 20586, l. 4-6: could the authors please explain the difference between

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"model performance goal" and "model performance criterion" l. 14: as before "aerosol concentrations and aerosol mass (?) ..." l. 20: suggested "Because the differences between simulations and observations are small. . ."

Authors: According to Boyland and Russell (2006), the performance goal is the level of accuracy that is considered to be close to the best a model can be expected to achieve. The performance criteria are the level of accuracy that is considered to be acceptable for modelling applications. The rest of suggestions have been amended. This has been clarified in the manuscript as follows: P. 20586 Line 4: "Additionally, they propose model performance goals (the level of accuracy that is considered to be close to the best a model can be expected to achieve) and criteria (the level of accuracy that is considered to be acceptable for modelling applications) that vary as a function of concentration and extinction." The rest of the comments have been amended.

Referee #2: 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?

Authors: P. 20587 Line 1: "General trends" is related to the spatial patterns and seasonal evolutions. This is emphasized in the manuscript as follows: "[. . .] the modelling system is able to reproduce daily variations of gas phase pollutants (SO2, NO2 and O3) as well as their spatial distribution and seasonal evolution." P. 20587 Line 6: "moderately well" has been replaced in the sentence as follows: "It is able to reproduce the daily evolution through the year with annual correlations between PM2.5model calculated and observed of 0.47 (Fig. 2a)." P. 20592 Line 15: "The model reproduces mod-

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erately well [. . .]" has been replaced by "In general, the CALIOPE system reproduces [. . .]" P. 20587 line 9: "PM10 is to a slightly larger extent underestimated" has been replaced by "PM10 is larger underestimated (annual MB = -10.30 μg m-3) than for PM2.5". P. 20587 Line 22: "total particulate mass" has been replaced by "PM10". Concerning SIA, the CALIOPE system overall underestimates measured aerosol species. Although the present manuscript mainly focuses on quantifying those underestimations, a more detail discussion about discrepancies between model and observations is provided in a companion paper submitted in Atmospheric Environment (Pay et al., 2011b: Assessing sensitivity regimes of secondary inorganic aerosol formation in Europe with the CALIOPE-EU modelling system) which evaluate temporal and spatial modelled SIA and their gas precursors, as well as SIA formation regimens in Europe. First of all, one source of model underestimation of measured SIA is related to the size fraction of aerosol components in both measurements and the model. The observed concentrations are available in total mass fraction, without any discrimination of size since EMEP sites typically used filter-packs to measure SO42-, NO3-, NH4+ and captured particles are approximately PM10 fraction. In contrast, the CMAQv4.5 model considers speciated inorganic aerosol only in the PM2.5 fraction. One exception is marine sulphate aerosol which is present also in the coarse fraction. Model evaluation of gas precursors SO2 in the framework of the CALIOPE system over Europe (Pay et al., 2010; Pay et al. 2011b) shows a positive mean bias for SO2 which suggests that SO42- formation in the system is often limited by oxidant availability and not always by SO2 availability. Winter underestimation of SO42- is a common issue in most models integrated in Europe which represent a direct couplet of sulfur chemistry with photochemistry, even detected with CMAQv4.5 over Europe (Matthias, 2008). This feature can be probably explained by a lack of model calculated oxidants or missing reactions (Kasibhatla et al., 1997). In this context, besides the gas-phase reaction of SO2 by OH, Tarrasón and Iversen (1998) and Schaap et al. (2004b) included additional oxidation pathways in clouds under cool and humid conditions that improve modeled SO42- performance. Concerning nitrate, one source of underestimation is related to the fact that

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CMAQv4.5 (AERO4) does not consider the formation of Ca(NO3)2 and NaNO3 salts in the coarse fraction which are account to be significant in Spain from mid-spring to mid-autumn (Rodríguez et al., 2002; Querol et al., 2004, Querol et al., 2009). NH4NO3 is usually dominant in winter whereas calcium- and/or sodium-nitrate dominate in summer (Rodríguez et al., 2002; Querol et al., 2004, Querol et al., 2009). NH4NO3 mostly occurs in PM2.5, whereas calcium- and/or sodium-nitrate nitrate mostly occur in the coarse PM2.5-10 mode (Rodríguez et al., 2002; Querol et al., 2004, Querol et al., 2009). This could be related with the underestimations observed in Eastern Spain, particularly in summer (Fig. 3). Moreover, correlations between the simulated and the observed values are rather low in summer in Spain (Fig. 3). A better knowledge on NH3 emission (sources and temporal and spatial dessagregations) and concentrations is needed in order to improve SIA modelling. de Meij et al. (2006) shows that high temporal and spatial resolution in NH3 emissions is crucial when SIA concentrations are modelled. Another source of the underestimation of aerosol species is related to the fact that long-range transport is not taken into account. We include chemical lateral boundary conditions from LMDz-INCA2 global model for gas-phase species, but we could not provide similar boundary conditions for aerosols species. Realistic lateral boundary conditions for aerosol species are needed. A summary of thie discussion is included in the reviewed version of the manuscript as well as in Pay et al. (2011b). The rest of the comments have been amended.

Referee #2: 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 “favouring the regional mixing” somewhat contradictory?

Authors: As the reviewer indicates, “low air renovation” and “favouring the regional mixing” seem contradictory terms. However, in summer the stable anticyclonic weather conditions are associated to the larger soil temperature, and as a consequence, larger heat and momentum ïňĆuxes. These conditions favour the growth of planetary boundary layer (PBL) height. Thus, “low air renovation” is referring to processes at synoptic

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scale meanwhile “favouring the regional mixing” is associated to a mesoscale processes. This has been emphasized in the manuscript. The rest of the comments have been amended.

Referee #2: 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.”

Authors: As the reviewer indicates NH3 is a direct gas precursors of NH3NH4 aerosols: atmospheric NH3 is first neutralized by H2SO4 to form (NH4)2SO4. Remaining NH3 may then combine with HNO3 to form NH4NO3. de Meij et al. (2006) shows that high temporal and spatial resolution in NH3 emissions is crucial when modelling aerosol concentrations, and especially in modelling policy related reduction strategies (Reis et al., 2009) 94% of NH3 total emissions are attributed to agriculture and livestock (EMEP, 2007). Domestic animals contribute most to total emissions, followed by fertilizers, crops and others. The fact that agricultural activities contribute most to ammonia emissions implies that densely populated regions tend to have the highest ammonia emissions. Livestock sources vary during the year since volatilization of NH3 from the animal waste is a function of temperature (Gilliland et al, 2003). Seasonality in NH3 emission is expected since field application of fertilizers occurs during specific seasons (Asman, 2001). In the CALIOPE system over Europe, NH3 emissions are derived from the 2004 annual EMEP emission database (EMEP, 2007). The spatial top-down disaggregation applies high-resolution land use map (EEA, 2000). In the time dimension, data are mapping from annual to an hourly basis using the temporal factors of EMEP/MSC-W. Annual emissions are temporally distributed applying fixed seasonal variations. This methodology is widely used in chemical transport models like the EMEP model (Fargely and Aas, 2008), CHIMERE (de Meij et al., 2009), TM5 (de Meij et al., 2006), MATCH (Langner et al., 2009). This methodology is simple because detailed agricultural registers are not generally available in many countries. In

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the framework of COST ES0602, Menut and Bessagnet (2010) indicate that none of the 27 air pollution forecasting system, intercompared in the European domain, contains an accurate temporal profile for ammonia. The main limitation for such NH3 emission model is the lack of reliable input data. These evidences have been included in the manuscript as follows: P. 20589 Line 16: "94% of NH3 total emissions are attributed to agriculture and livestock (EMEP, 2007). Livestock sources vary during the year since volatilization of NH3 from the animal waste is a function of temperature (Gilliland et al, 2003). Seasonality in NH3 emission is expected since field application of fertilizers occurs during specific seasons (Asman, 2001). In the CALIOPE system over Europe, annual emissions of NH3 are derived from the 2004 annual EMEP emission database (EMEP, 2007). However, detailed agricultural registers are not generally available in many countries (e.g. Menut and Bessagnet, 2010). The evaluation of modelled ammonium concentrations with measurements shows the annual trend is correctly reproduced (Fig. 2e) with a high correlation coefficient (r=0.62, see Table 1 and Fig. 3). Although, the modelled ammonium annual mean underestimates observed mean by a 36%, with the highest bias in winter (MB=-0.8μg m-3)". The rest of the comments have been amended.

Referee #2: 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.

Authors: As the reviewer suggests we explain more in detail the sentence in P. 20590 Line 6-7: The coarse mode in the AERO4 module implemented in CMAQv4.5 (Binkowski and Roselle, 2003) is implemented in a non-interactive way. That is, fine particles do not coagulate with coarse particles, nor do coarse particles coagulate with each other. This also means that dry deposition of chemical species that are emitted into the fine modes may be underestimated because a fractional amount of these species may have been moved to the coarse model by intermodal coagulation where

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removal by particle dry deposition may be stronger. Therefore, line 6-7 has been rewritten as follows: “The coarse mode in the AERO4 module in CMAQv4.5 (Binkowski and Roselle, 2003) is implemented in a non-interactive way. That is, fine particles do not coagulate with coarse particles, nor do coarse particles coagulate with each other.” P. 20590 Line 8: According to Putaud et al. (2004), that sentence has been rewritten in the reviewed manuscript as follows: “Both EC and OC can contribute significantly to PM2.5 and PM10 in urban and kerb sites, and their mass fraction might be higher than the sum of the inorganic components.”

Referee #2: p. 20591, l. 7-8 repetition l. 19-20: Do the authors really mean that the current model performance for AOD is satisfactory?

Authors: In the sentence in P. 20591 Line 7 “AODfine corresponds to sub-micron aerosols and AODcoarse corresponds to super-micron aerosols” has been removed. P. 20591 Line 19-20: As indicated in the manuscript, the annual MFB and MFE obtained in the AOD comparison against AERONET observations meet the model performance criteria defined by Boylan and Russell (2006) indicating acceptable performances for the modelled AOD. Typically vertically integrated values are better reproduced by models because among other reasons deficiencies in the vertical distribution do not affect the results as much as for surface concentrations. In the case of the CALIOPE modelling system, annual AOD correlations are in the range of other European modelling systems as shown in the discussion of the manuscript indicating that our modelling system can reproduce reasonably well the daily variability of the main aerosol component in Europe. P. 20591 Lines 18-20 have been removed of the manuscript.

Referee #2: p. 20592, l. 13: correct “being” to “were” or “are”.

Authors: Amended

Referee #2: p. 20593, 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

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refer to any specific process, which the CALIOPE fails to model soundly?

Authors: The Po Valley offers many interesting aspects regarding air pollution problems and reduction strategies (Martilli et al., 2002; Silibello et al., 2008), including (1) frequent episodes with very high photooxidant levels, (2) recirculation of air masses over the Po Valley owing to the topographic situation, (3) rapid changeover from VOC to nitrogen oxide limited O3 production, and (4) large production of secondary aerosol with a major influence from biogenic VOC and NH3 emission due to intensive farming and high emissions of isoprene and terpenes in the pre-Alps. Additionally, in winter, frequent humid and stagnant situations favour the nucleation and growth of aerosols (Mélin and Zibordi, 2005; Rodríguez et al., 2005; Hamed et al., 2007). "Challenges models" refers to the difficulty in implementing these complex processes in regional air quality models which usually work at horizontal resolution of a few tens of kilometers. P. 20593 Line 17: "challenges models" has been replaced by "favours the nucleation and growth of aerosols". The rest of the comments have been amended.

Referee #2: 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"?

Authors: As the reviewer pointed out also in the general comments, the factors of the underestimation of EC and OC in the CALIOPE system was poorly developed. Please, see the discussion about the underestimation of EC and OC in the CALIOPE system in the answer of the general comment number 3. P. 20594 Line 25: "different aerosol fractions" has been replaced by "different aerosol components".

Referee #2: p. 20596, l.9: suggested to change "aerosol fractions" to" PM concentrations and AOD"; l. 13: remove "To a lesser extent"

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Authors: Suggestion has been amended.

Referee #2: 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.

Authors: As the reviewer indicates, these uncertainties are not just the same for EC and OC. However, the model underestimates the measurements by a factor of 4 (DE44: measured mean (OC+EC) 3.21 μg m-3, modelled 0.66 μg m-3; NO01: measured mean (OC+EC) 0.97 μg m-3, modelled 0.23 μg m-3). This cannot be said more precisely because measurements were missing at the other sites. As the referee #2 can see in Table 3R, it is difficult to establish a degree of uncertainties related to each component. For this reason, we have maintained the discussion of both aerosol components together. Figure 2R presents the annual modelled EC and OC surface concentrations with the CALIOPE system. As the referee #2 can see, EC is a primary pollutant so its spatial variability is relatively high. Major sources of elemental carbon include diesel engines, particulate heavy-duty trucks, and combustion process (including biomass and fossil fuel), thus high levels are associated with urban areas and maritime routes (as the Strait of Gibraltar). In contrast, OC is emitted directly or, in a large proportion, formed from the condensation of low-volatility organic compounds. Thus, the spatial variability of OC is between that of purely primary and secondary pollutants. Major primary sources of OC include diesel and gasoline-burning engines, biomass burning and some industrial processes, so OC will be found in urban and rural background environments. These considerations have been included in the discussion of the section 4.2 as follows: P. 20597 Line 27: “EC is a primary pollutant so its spatial variability is relatively high. Major sources of elemental carbon include diesel engines, particulate heavy-duty trucks, and combustion process (including biomass and fossil fuel), thus high levels are associated with urban areas and maritime routes. In contrast,

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OC is emitted directly or, in a large proportion, formed from the condensation of low-volatility organic compounds. Thus, the spatial variability of OC is between that of purely primary and secondary pollutants. Major primary sources of OC include diesel and gasoline-burning engines, biomass burning and some industrial processes, so OC will be found in urban and rural background environments." P. 20598 Line 2: This sentence has been rewritten as follows: "OC+EC estimations should be taken with precaution due to few measurements available over Europe for the model evaluation results as well as the considerable uncertainties associated to the calculations of EC and OC." As indicated previously, the year 2004 has been selected because is the year corresponding to the emission inventory of EMEP that was processed for the present work. Currently, we are working on updating the emission inventory for a more recent year (2008). The rest of the corrections have been amended.

Referee #2: p.20598, l. 13: should be "non-climatological basis"

Authors: Amended.

Referee #2: 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?

Authors: As the reviewer indicates, "missing aerosol sources, which affect these large fractions" are related with those missing sources of coarse PM not considered in the emission inventory as wildfires, windblown dust or resuspension. Furthermore, the model does not estimate the formation of coarse nitrate through reaction of nitric acid with sea salt or dust, as indicated previously. This has been rewritten in the manuscript as follows: P. 20600 Line 1-2: "[. . .] the larger underestimation in PM10 compared with PM2.5 suggests missing sources of coarse PM in the modelling system as wildfires, windblown dust or resuspension. Furthermore, the model does not estimate the formation of coarse nitrate through reaction of nitric acid with sea salt or dust." P. 20600 Line 25: "aerosol mass budget" has been replaced by "PM10 and AOD values".

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Referee #2: p. 20601, 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".

Authors: As the reviewer indicates, P. 20601 Line 13: "the secondary organic aerosol formation and" has been removed. A new version of CMAQ is being tested in the CMAS community, namely CMAQv5.0. It includes a new aerosol module, AERO5, which contains substantial scientific improvements over AERO4 released in CMAQv4.5, especially devoted to improve SOA formation and dynamic interactions of fine and coarse aerosols. CMAQv5.0 allows semi-volatile aerosol components to condense and evaporate from the coarse mode and non-volatile sulphate to condense on the coarse mode. Dynamic mass transfer is simulated for the coarse mode, whereas the fine modes are equilibrated instantaneously with the gas phase. This has been included in last paragraph of the manuscript.

Referee #2: 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).

Authors: Suggestion has been amended.

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Interactive Comment

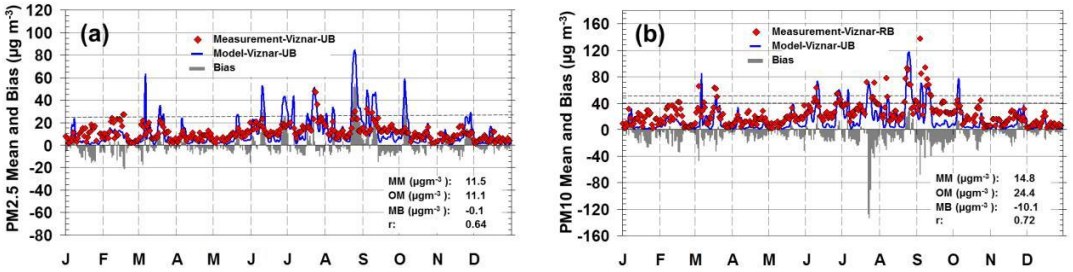

Fig. 1. Figure 1R: Time series of observed (red diamond) and simulated (blue continuous line) daily PM2.5 (left column) and PM10 (right column) for the year 2004: at (a, b) Víznar EMEP station.

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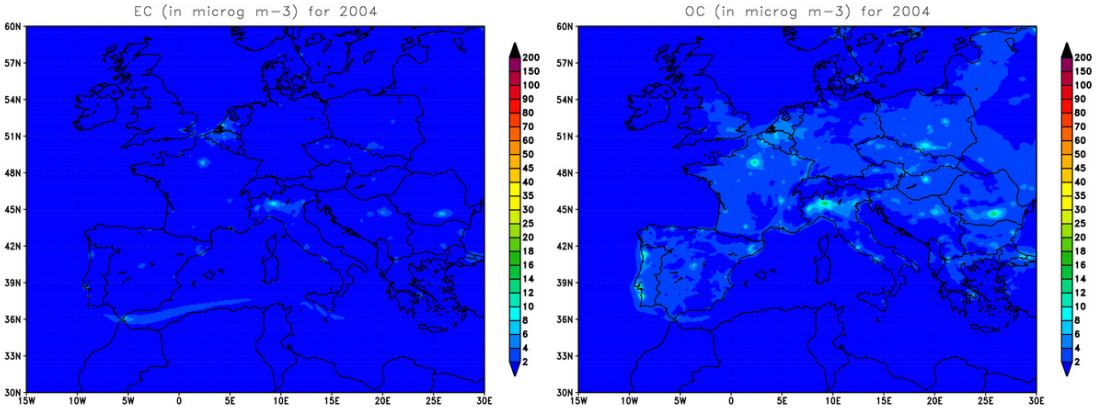

Fig. 2. Figure 2R. Annual average of the EC and OC (in microg m-3) obtained with the corrected CMAQ + BSC-DREAM8b simulations.

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Interactive Comment