Referee report regarding the manuscript:

# Presentation of the EURODELTA III intercomparison exercise - Evaluation of the chemistry transport models performance on criteria pollutants and joint analysis with meteorology

Authors: B. Bessagnet et al.

**We thank a lot the reviewer for a very complete review of our paper. The answers are written here below in bold characters after each comment.**

General comments

In my opinion this paper is not suitable for ACP — it is essentially a technical report from the EURODELTA project, and I think it should be published as such; a (rather confusingly presented) model intercomparison, for a limited set of "standard" atmospheric components, is not interesting enough for publication in ACP. Many similar model intercomparisons have been published and, as written, this one does not contribute anything new.

**As mentioned in the introduction: "Differently to the previous inter-comparison exercises, most of models have been run in EURODELTAIII with the same input data (emissions, meteorology, boundary conditions) and over the same domain (domain extension and resolution) with some rare exceptions. Participating models were applied over four different periods, within a rather limited number of years thus allowing to evaluate the influence of different meteorological conditions on model performances."**

**The joint analysis of meteorology and criteria pollutants is not so frequent in the literature; we think it is the first time in the frame of an intercomparison exercise such an analysis is performed. For instance, the analysis of the boundary layer height and wind speed of each model are particularly interesting and show how the models are dependent on variables that are not so frequently evaluated.**

The paper only documents model results for seven different chemical transport models without enough detail to be able to draw any useful conclusions for the general scientific community. I can not see what this paper contributes to the understanding about the atmospheric chemistry or physics, or any new information that aids in improving modelling of the atmospheric composition.

**See above. Moreover, as mentioned in the introduction we remind here the objectives of the paper : "The objective of this paper is twofold, (i) to introduce the exercise, the input data and the participating models, and (ii) to analyse the behaviour of models in the four campaigns focussing on the criteria pollutants $PM_{10}$, $PM_{2.5}$, $O_3$, $NO_2$ and $SO_2$ and relevant meteorological variables. Complementary analyses of depositions fluxes and PM composition data at high temporal**

**resolution will be discussed in companion papers in order to better understand the behaviour of models. »**

I also think that the authors are fragmenting their research (over several papers). This should be avoided (see the ACP Obligations for authors). In order to better understand model performance you need to take into account all important processes — including deposition and chemistry. If the authors want to publish this material in a scientific journal I think it has to be combined with the information about deposition and chemical composition of particulate matter. Splitting the model evaluation into three different papers is not appropriate.

**The two objectives of the paper (mentioned before) are not fragmented. It is common to break down the results analysis of such projects in several papers.**

The paper could have been acceptable for Geoscientific Model Development (GMD, which accepts model evaluation papers) — if the presentation had been better — but I think that a much more scientific approach is needed to make the material presented in this manuscript interesting enough for publication in a peer-reviewed journal.

**For the first part of the comment, the editor will also have an opinion. However, we see several model evaluation papers in ACP such as these recent ones:**

**Pan, X., Chin, M., Gautam, R., Bian, H., Kim, D., Colarco, P. R., Diehl, T. L., Takemura, T., Pozzoli, L., Tsigaridis, K., Bauer, S., and Bellouin, N.: A multi-model evaluation of aerosols over South Asia: common problems and possible causes, Atmos. Chem. Phys., 15, 5903-5928, doi:10.5194/acp-15-5903-2015, 2015.**

**Prank, M., Sofiev, M., Tsyro, S., Hendriks, C., Semeena, V., Vazhappilly Francis, X., Butler, T., Denier van der Gon, H., Friedrich, R., Hendricks, J., Kong, X., Lawrence, M., Righi, M., Samaras, Z., Sausen, R., Kukkonen, J., and Sokhi, R.: Evaluation of the performance of four chemical transport models in predicting the aerosol chemical composition in Europe in 2005, Atmos. Chem. Phys., 16, 6041-6070, doi:10.5194/acp-16-6041-2016, 2016.**

**For us, GMD should be more dedicated to model development that is not strictly the topic of our paper here.**

**For the second part of the comment, see above our first answers.**

I am sure that there are a number of interesting scientific questions that the EURODELTA project can answer, and I suggest that the authors focus mainly on that, and keep this kind of model intercomparison/evaluation documentation to technical reports. This paper in itself has little significance for the ACP community.

**As we previously mentioned, we think this paper is the first to address both an analysis on meteorology and chemistry. Most of the previous exercises addressed only comparisons on chemical compounds without any analysis on meteorology. Also, this paper is an introduction of the exercise with other papers that are in preparation or submitted.**

The paper has a very long author list with 36 authors! However, the brief statement on page 3, about what seven of the participating institutes (and I think NILU is missing in this list) have contributed to the project, is not motivation enough for the inclusion of so many authors.

**NILU is included in the list of authors but we forgot it in page 3. Finally, we removed the sentence mentioning the role of partners, following the recommendation of the other referee.**

Considering the very long author list, please give a brief explanation of what each individual has contributed to this paper in the reply to this referee comment (the statement of contributions can be added to the supplement of the paper). Please note Point 9 under the General Obligations for Authors for ACP (my highlighting):

"To protect the integrity of authorship, only persons who have significantly contributed to the research and paper preparation should be listed as authors. The corresponding author attests to the fact that any others named as authors have seen the final version of the paper and have agreed to its submission for publication." … "The author who submits a manuscript for publication accepts the responsibility of having included as co-authors all persons that are appropriate and none that are inappropriate."

**Regarding the number of authors: EURODELTA is an on-going project which started in 2001. This project is a very cooperative project involving an important number of organization and researchers to cover all topics : coordination, data management, modelling, emissions, meteorology, boundary conditions, results analysis, manuscript preparation. For a first paper on a project it is normal to put all people who have participated to the project. As an example, for the MACCII project, we count 60 authors for this paper in GMD (same author policy) with the same number of modeling teams :**

**Marécal, V., Peuch, V.-H., Andersson, C., Andersson, S., Arteta, J., Beekmann, M., Benedictow, A., Bergström, R., Bessagnet, B., Cansado, A., Chéroux, F., Colette, A., Coman, A., Curier, R. L., Denier van der Gon, H. A. C., Drouin, A., Elbern, H., Emili, E., Engelen, R. J., Eskes, H. J., Foret, G., Friese, E., Gauss, M., Giannaros, C., Guth, J., Joly, M., Jaumouillé, E., Josse, B., Kadygrov, N., Kaiser, J. W., Krajsek, K., Kuenen, J., Kumar, U., Liora, N., Lopez, E., Malherbe, L., Martinez, I., Melas, D., Meleux, F., Menut, L., Moinat, P., Morales, T., Parmentier, J., Piacentini, A., Plu, M., Poupkou, A., Queguiner, S., Robertson, L., Rouïl, L., Schaap, M., Segers, A., Sofiev, M., Tarasson, L., Thomas, M., Timmermans, R., Valdebenito, Á., van Velthoven, P., van Versendaal, R., Vira, J., and Ung, A.: A regional air quality forecasting system over Europe: the MACC-II daily ensemble production, Geosci. Model Dev., 8, 2777-2813, doi:10.5194/gmd-8-2777-2015, 2015.**

Specific comments

Page 3, lines 34–35: "As a consequence, there were very limited differences in the models set up, representing a sort of sensitivity analysis to several aspects of the modelling chains."

● I do not understand what you mean by this sentence! What do you mean by "a sort of sensitivity analysis" and in what sense is this study a sensitivity analysis? I suggest that this sentence is removed.

**The reviewer is right, we modified the sentence as: "*As a consequence, most of differences in the outputs will be attributed to the simulation of chemical and physical processes*".**

Page 4, lines 3–4: "Complementary analyses of depositions fluxes and PM composition data at high temporal resolution will be discussed in companion papers in order to better understand the behaviour of models."

● In my opinion this is fragmentation of research papers, and the consequence is that the present paper becomes uninteresting. As mentioned in the General Comments, I do not think that splitting the information this way in three different papers is useful. If knowledge about the deposition fluxes and PM composition data are important for understanding the behaviour of the models (which I certainly expect them to be) this information is needed in the present paper!

**This comment on "fragmentation" is discussed in a previous answer.**

Page 4, lines 25–26: "In CMAQ additional anthropogenic dust is calculated as 90% of unspecified PM coarse emissions and attributed to fugitive dust"

● What is the motivation for adding extra anthropogenic dust? Was this just a modeling mistake? Or do you have good reasons to believe that the emission inventory used in the present study lacks a substantial amount of fugitive dust? And if this is the case, why did you not increase the emissions in all models?

**This is exactly what Binkowski and Roselle (2003) wrote :"The emissions inventory used for this contribution estimates that 90% of PM10 is fugitive dust, and that 70% of this dust consists of PM2.5 particles. The paradigm adopted for the CMAQ model is that fugitive dust is a coarse mode phenomenon with a tail that overlaps the PM2.5 range." This fraction would be the resuspension of dust produced by human activities, the same we investigated in Vautard et al., 2005. We did not switch off this additional emission in CMAQ. Such types of emission parameterization are not available in the other models.**

Page 4, line 27: "CAMx did not activate the sea salts parameterisation in this exercise."

● Why not? Was this a modelling mistake? Or are there problems with the sea salt emissions in CAMx?

**Yes they had problems with the use of the sea salt parameterization. Sea salt modeling in general has large uncertainties mainly in generation of sea spray which occurs as the waves break on the surface of the ocean and whitecaps form. Sea-salt pre-processor of CAMx was not available in the lat-lon grid system at the time of the exercise. The initial attempt to adapt it to the required grid to generate sea salt emissions resulted in too high emissions over the north Atlantic. Due to very high uncertainty, we decided not to include it for this exercise.**

Page 4, lines 30–31: Why was CAMx not included in the ENSEMBLE for O3, NO2, and SO2? I would guess that the lack of sea salt would hardly have any impact on these three gaseous species.

**We preferred to have an ENSEMBLE consistent for all species since we also compared the ENSEMBLE between gases and PM, that's why we excluded CAMx.**

Page 8 — Emissions: Are the emissions used in the EURODELTA project available for use by scientists outside the project? If they are, please specify this and where they can be found. If they are not available, more details are needed regarding the emissions in order for others to be able to evaluate/compare this work to other studies using other emission data. Without more detailed information the work presented in this manuscript can not be considered reproducible.

**The emission data are available, we would suggest to put them on ACP if zip files are permitted, but the amount of data is too large. We wrote that data are available on request: "*The full emission dataset is available on request to INERIS*".**

Page 8, lines 16–22: "EMEP national emissions were kept except for..." 14 countries, for which GAINS emissions were used.

● This seems a bit strange - why did you change emissions for these 14 countries and not for the other countries? Please give a motivation. "Additional factors were applied on two Polish regions (x4 or x8) for PM2.5 and PM10 emissions"

● For which Polish regions? They need to be specified in detail to make this work reproducible. It is also unclear if the x4 factor applies to PM2.5 and x8 to PM10 or if the same factors were used for PM2.5 and PM10?

**Yes it is unclear, we have changed it. The same factor was applied for PM2.5 and PM10, but for two different regions. We rephrased it as : "*The country emissions were re-gridded with coefficients based on population density and French bottom-up data, the methodology (Terrenoire et al., 2015) was extrapolated to the whole Europe. For PM2.5 emissions, the annual EMEP national totals were kept except for the countries: Czech Republic, Bosnia and Herzegovina, Belgium, Belarus, Spain, France, Croatia, Ireland, Lithuania, Luxemburg, Moldavia, Republic of Macedonia, Netherland, Turkey. For these countries, PM2.5 emissions from GAINS were used as this database provides higher numbers and certainly more realistic ones since wood burning is known to be underestimated in the EMEP database (Denier van der Gon et al., 2015). Additional factors were applied on two Polish regions for both PM2.5 and PM10 emissions. As a preliminary solution, domestic combustion emissions from provinces with active coal mines were multiplied by a factor of 8, while those in neighbouring provinces were adjusted by a factor of 4 (Kiesewetter et al., 2015).*"**

Page 8, line 27: What do you mean by "artificial area"?

**We mean "built-up" area.**

Page 8, lines 28 and 31: EPER data are only available for the EU-countries + Norway — how did you treat industrial emissions in the other countries?

**They are treated using the "built-up area" proxy**

Page 8, line 31: What is "artificial landuse"?

**We mean "built-up" area. We mentioned it in the revised manuscript.**

Page 8, lines 36–38: Considering the great uncertainties in the residential combustion emissions I suggest that you give some more details about the emissions you have used in EURODELTA. The statement that "Germany, Sweden, and Spain clearly have the lowest (levels of) emissions" is not clear enough. Do you mean the lowest emission per capita? Or per square km? In order for the results from the EURODELTA modelling to be comparable to other studies I suggest that you add a table to the Supplement specifying annual total national residential combustion emissions assumed in the EURODELTA inventory.

**We mean the lowest emission for the whole country, we clarified it. We added a table for the PM emission of sector 2 in the supplementary material S8: "*Residential emissions of particulate matter are dominant in wintertime. In most countries, they come from wood burning or coal uses. Germany, Sweden, Spain clearly have the lowest levels of PM2.5 emissions for this activity sector. Romania, Poland and France have the highest levels of annual total emissions per country (Terrenoire et al., 2015). For this activity sector, the PM2.5 emissions by components are provided in supplementary material S8.*"**

Page 9, line 3: What are "the usual default profiles"?

**We mean the usual <u>vertical</u> profiles, that are used in all models to redistribute the emissions.**

Page 9, lines 4–6: "a PM speciation profile provided by IIASA (Personal Communication from IIASA) was used to estimate the fraction of Non-carbonaceous species, Elemental Carbon and Organic Matter per activity sectors and country"

● This PM speciation profile must be provided with the article. Personal communication with an organisation (IIASA) is not a reference that makes it possible for readers to find the relevant information to be able to reproduce the work. A table specifying the three PM2.5 and coarse PM fractions for each emission sector and country should be added to the Supplement of the paper.

**In the revised manuscript we added the reference Klimont et al. (2013) that is the most appropriate and often cited in ACP for the PM emissions and split into EC/OM/Other:**

**Klimont, Z., Kupiainen, K., Heyes, C., Cofala, J., Rafaj, P., Höglund-Isaksson, L., Borken, J., Schöpp, W., Winiwarter, W., Purohit, P., Bertok, I., and Sander, R.: ECLIPSE V4a: Global emission data set developed with the GAINS model for the period 2005 to 2050: key features and principal data sources, International Institute for Applied Systems Analysis (IIASA), Schlossplatz 1, 2361 Laxenburg, Austria, 8 pp., available at: http: //eccad.sedoo.fr/eccad_extract_interface/JSF/page_login.jsf, 2013.**

Page 9, lines 22–23: There is no reference to a description of the SMOKE system.

**We added the web site address; there is no publication, only a user manual.**

Page 10, lines 10–15, Wildfire emissions: Which emitted species were included for wildfires?

What gases and which particulate species were included (include information about how the PM-emissions were split between organics, BC, and other PM-components)?

**We added the species in the paper. The following compounds have been selected: CO, CH4, NOx, SO2, PM2.5, TPM, OC, BC. We did not include VOC as the split.**

Page 10, lines 22–23: Why were the agricultural and road dust PM sources not activated in the LOTOS-EUROS model?

**This was a decision of the TNO modeling team. This kind of parameterization can be considered not robust enough and too dependent on the meteorological driver. The same opinion is shared by the CHIMERE team, the dust resuspension scheme was fitted with the MM5 soil moisture, this variable was differently diagnosed in WRF or IFS for instance.**

Page 15, lines 4–6, Regarding the PBL and the LOTOS-EUROS and EMEP models: "LOTOSEUROS and EMEP that should adopt IFS PBL too, show partially different performance, suggesting that the latter models partially recomputed boundary layer height."

● This is too unclear! You have to be able to describe how these models handle the PBL! In what way do they "partially recompute" the BL height?

**We clarified it in the revised version. Some variation can occur due to differences in the interpolation processes. For EMEP, as explained in page 6 , line 12, a minimum PBL is assigned, explaining the differences on the diurnal cycle Fig. 3.**

Page 16, lines 5–6: "The large positive bias in 2007 and negative in 2009 are largely explained by the boundary conditions that are biased respectively of +8 and -20 μg m-3"

● I agree about the negative bias in the 2009 campaign but the bias of +8 μg m-3 in the 2007 campaign can hardly be considered to "largely explain" the very large positive bias (21–23 μg m-3) for CAMx, CMAQ and Chimere — I guess there must be other factors that are more important than the boundary conditions to explain the poor performance of these three models?

**Yes for this group of models in 2007 this bias on boundary conditions partly explains the overestimation. In winter these models give the highest values, the chemistry processes are certainly the main reasons. We modified the comment.**

Page 16, lines 7–8: "For the summertime campaign 2006 CHIMERE and CMAQ display the lowest correlation for daily averaged concentrations"

● Can you explain the very poor correlation for Chimere and CMAQ for this summer period?

**The poor correlation is associated to both low spatial and temporal correlations. These models have troubles to estimate the background concentration over the Alpine regions. We improved the comment in the revised manuscript as follows: "*The low correlation for CMAQ and CHIMERE is due to the difficulties to reproduce both spatial patterns and day to day variations.*"**

Page 16, lines 11–13: "All models simulate high ozone concentrations over the Mediterranean sea, most of them behaves satisfactorily in Malta and Cyprus stations confirming the ozone concentrations pattern over the seas for the "ensemble" shown in Fig. 6."

● What do you mean by "confirming the ozone pattern over the seas"? Do you mean that a "satisfactory" behaviour at two sites in the Mediterranean region proves that the model ensemble

gives good ozone concentrations over all sea areas? Also, in Fig. 6 I see no observation data from Cyprus so for this summer period it is really only one site you base your statement on?

**Yes there are two sites even for the summer period (the site is difficult to see in the Figure over Cyprus). But you are right two sites are certainly not sufficient for a "confirmation", we rephrased the comment and added this reference : "*Nolle, N., Ellul, R., Heinrich, G., Güsten, H. (2002) A long-term study of background ozone concentrations in the central Mediterranean—diurnal and seasonal variations on the island of Gozo, Atmospheric Environment, Volume 36, Issue 8, March 2002, Pages 1391-1402*"**

Page 16, lines 20–21: "This result confirms that during stable conditions the pollutant concentration is influenced not only by the PBL height, but also by the overall reconstruction of vertical dispersion."

● What do you mean by "the overall reconstruction of vertical dispersion"? And could the differences of the results not also be due to differences in dry deposition and chemistry?

**The reviewer is right, certainly this point must be addressed for a pollutant like NO2 less influenced by chemistry. We have the diurnal profiles for CO and it is coherent with our comment. Then we removed the sentence.**

Page 16, lines 26–28: "Not only the bias is affected by global boundary conditions, but also this result indicates that biased ozone boundary conditions globally impair the normalized statistics confirming the non linearity of ozone chemistry."

● This sentence hardly makes any sense at all to me. I think it is unclear what you mean and it seems like just speculation to me.

● What do you mean by "globally impairing normalized statistics" and how does this "confirm the non linearity of ozone chemistry"?

● As mentioned above I do not think that you have shown that the global boundary conditions is the main reason for the model problems for the 2007 campaign! Of the four ENSEMBLE models Chimere performs very poorly for 2007 (or at least very differently than the other three models) and this can not be explained by the global boundary conditions.

**Yes we agreed and removed this sentence that was unclear.**

Page 17, lines 8–9: "This underestimation of NO2 concentrations is certainly related to rather high ozone concentrations."

● Can you explain why CAMx behaves differently than the other models (e.g. CMAQ also has high ozone concentrations)?

**The reviewer is right, this comment has to be complemented by the previous remark of the reviewer on ozone (Page 16, lines 20–21). Looking at elemental carbon (primary species) in Bessagnet et al. (2014) confirms the hypothesis of an impact of vertical mixing that is different and the minimum Kz quite high in CAMX explain the height dilution of primary compounds. We write in the revised version: "*Bessagnet et al. (2014) showed rather low concentrations of elemental carbon compared to other models, this inert species is particularly sensitive to vertical mixing and***

***CAMx presents the highest minimum diffusion coefficient that is of major importance during stable conditions and partly explaining the low NO2 concentrations.*"**

Page 17, lines 16–17: "Over lands the NO2 chemistry and the different biogenic NO emissions explain a large part of the differences far from urban areas."

● How does this explain the differences between the models — be specific.

**Far from the anthropogenic sources, the chemical processes and the biogenic emissions have more impact with respect to anthropogenic emissions. We changed to "*Over land the NO2 chemistry and the different biogenic NO emission modules in the models are believed to explain a large part of the differences on NO2 concentrations far from urban areas*".**

Page 17, lines 19–20: "It should be pointed out that the observed NO2 concentrations can be slightly overestimated because of sampling artefact (evaporation of nitric acid)."

● What do you mean by slightly? Give some number/estimate! How large overestimation of NO2 could you possibly get from the evaporation of HNO3?

● Provide a reference for this sampling artefact.

**We added this explanation with a correction:"*For some types of analyzers, NO2 is catalytically converted to NO on a heated molybdenum surface and subsequently measured by chemiluminescence after reaction with ozone. The drawback of this technique is that other oxidized nitrogen compounds such as peroxyacetyl nitrate and nitric acid are also partly converted to NO (Steinbacher et al., 2007)*". The reference is given below:**

**Steinbacher, M., C. Zellweger, B. Schwarzenbach, S. Bugmann, B. Buchmann, C. Ordonez, A. S. H. Prevot,and C. Hueglin (2007), Nitrogen oxide measurements at rural sites in Switzerland: Bias of conventional measurement techniques, J. Geophys. Res., 112, D11307, doi:10.1029/2006JD007971.**

Page 17, lines 32–33: "Differently, differences in diurnal temperature between CMAQ and other models seem less relevant with respect to pollutant concentration."

● How do you know that the temperature differences are less relevant? And does this statement only refer to the NO2-concentrations or to all pollutants?

**The reviewer is right, actually, this statement is not relevant, because CMAQ uses a very different meteorology compared to the others. We therefore removed this sentence and focused on the other models particularly those which reported CO concentrations.**

Page 18, Sect 6.3 Sulphur dioxide

● General comment: This section is very short and essentially only states that the model results for SO2 are quite poor with hardly any explanation why. I think a much more detailed investigation of the differences in deposition and chemistry are needed here.

**This part will be more detailed in a companion paper submitted in Atmospheric Environment in June 2016: "Garcia Vivanco et al., Joint analysis of deposition fluxes and atmospheric**

**concentrations predicted by six chemistry transport models in the frame of the EURODELTAIII project".**

Page 18, lines 6–7: "The overestimation of the first group of models could be explained as follows for MINNI which has the lowest PBL and RCG having the lowest wind speed."

● The sentence is strangely formulated — perhaps it could have been written something like: "The overestimation in the MINNI model could possibly be partially explained by the low model PBL height"

**Yes, thanks, we modified it as suggested.**

● However, I do not think that the "explanations" are very satisfying — in my opinion they are not really explanations at all:

○ For 2006 the EMEP model also severely underestimate the PBL height without overestimating SO2.

○ The wind speed in CMAQ is as low as in the RCG model, without overestimation of SO2, and these models actually have the smallest bias for U10 for the 2009 period.

**We agree that for SO2 it is much more difficult to interpret the model outputs without information on chemistry and deposition fluxes. The impact of the PBL for MINNI is discussed latter in section 7.1 and we can see the "partial" impact. We rephrased the section on SO2 as :**

**"*The correlations are rather low for all models in the range 0.2-0.4 for the 2006 campaign to 0.5-0.6 for the 2007 campaign (Fig. 4 and supplementary material S1 for all statistics). Two groups of models are identified CAMx, MINNI and RCG that largely overestimate the concentrations and CHIMERE, CMAQ, EMEP and LOTOS-EUROS which are closer to the observations on average with the best performances on the RMSE. The overestimation in the MINNI model could be partially explained by the low model PBL height. For CAMx, the possible reasons such as the vertical distribution of SO2 emissions near the harbours and coastal areas, insufficient conversion to sulfate and too low deposition were discussed in Ciarelli et al. (2016). This leads to a positive bias of the "ensemble" as shown in Fig. 10 (supplementary material S4) particularly in Western Europe; the normalized RMSE is frequently above 100% in most part of Europe. The main hot spots are located in the Eastern Europe in addition with high concentrations along the shipping routes. The coefficient of variation is the lowest over emission areas but very high in remote areas like over the oceans far from shipping tracks and over mountain areas. This behaviour, very different from a primary species like CO, is a first indication of the very different way to simulate the SO2 chemistry and deposition processes in the models.*

*The diurnal cycles presented in Fig. 11 show a peak at about 10:00 – 12:00. This peak is coherent with the hourly emission profiles of the industrial sector showing an emission peak at the same hours; however, most of models predict a larger decrease in the afternoon. Only CMAQ for the 2007 campaign captures satisfactorily the diurnal profile*."

**Ciarelli, G., Aksoyoglu, S., Crippa, M., Jimenez, J. L., Nemitz, E., Sellegri, K., Äijälä, M., Carbone, S., Mohr, C., O'Dowd, C., Poulain, L., Baltensperger, U., and Prévôt, A. S. H.: Evaluation of European air**

**quality modelled by CAMx including the volatility basis set scheme, Atmos. Chem. Phys., 16, 10313-10332, 10.5194/acp-16-10313-2016, 2016.**

Page 18, lines 20–21, Regarding the CMAQ-results:

● I do not think that the CMAQ results are very different for "at least three campaigns" — it strongly deviates for 2006 and deviates somewhat for 2008 but for the other two campaigns the CMAQ results look "similar" to the other models (at least for the RMSE, which is what was discussed here).

**In fact the CMAQ is very different in 2006 and 2008, we modified the text accordingly.**

Page 22, lines 6–8, Regarding the NO2 results at the German sites; only meteorological aspects are discussed here, but other things can also lead to modelling problems:

**Yes, we agree with the referee, but the aim of this section was indeed to focus only on the relationship between NO2 and meteorology, that –of course- is not the only possible reason of discrepancy, but surely one of the most relevant ones.**

● How do the model results for ozone look at the same sites?

**As shown in the supporting material, ozone for CAMx during the winter campaign is on average in line with observations.**

● Could NO2 emissions be underestimated?

**What is important here is to compare the different behaviour during day time and night time as well as from day to day. A global underestimation or overestimation could be attributed to the emissions but it is not the subject here. However, for traffic emissions we have added references: "*However, underestimation of NOx emissions cannot be ruled out as depicted in Vaughan et al. (2016) or Chen and Borken-Kleefeld (2016), these works highlight the potential underestimation of NOx traffic emissions*".**

**Vaughan, A. R., Lee, J. D., Misztal, P. K., Metzger, S., Shaw, M. D., Lewis, A. C., Purvis, R. M., Carslaw, D. C., Goldstein, A. H., Hewitt, C., N., Davison, B. D., Beevers, S. D., Karl, T. G. Spatially resolved flux measurements of NOx from London suggest significantly higher emissions than predicted by inventories. Faraday Discussions, DOI: 10.1039/c5fd00170f, 2016.**

**Chen Y. and Borken Kleefeld J.: NOx Emissions from Diesel Passenger Cars Worsen with Age. Environmental Science & Technology, 50 (1). pp. 3327-5851, 2016.**

Page 22, lines 8–14, regarding the NO2 results in the Po Valley

● Are you sure that you are not having problems with underestimated NOx emissions in this region?

**Same comments of before, we analyse here the evolution of the bias, not the bias itself. However, we added the previous comment regarding a possible underestimation.**

Page 22, lines 26–30, the discussion about the correlation between the performances of the ensemble (RMSE) with the variability of the models is a bit confusing.

● What values are you correlating?

**We calculate the correlation of the ensemble RMSE and the coefficient of variation of the ensemble.**

● Can low correlation coefficients (-0.2 to -0.3) for only three of four campaigns and only two species be considered significant? What the correlation coefficients for the other species?

**On average the correlations are very low but this slight negative value for these two compounds is significant.**

● Providing a table with the correlation coefficients for the different species and seasons may could probably make this easier to understand.

**In fact, for the other compounds the correlation is close to zero, we mentioned it, we are not sure it is necessary to create a table.**

Page 23, line 21: What do you mean by "a relevant spatial variability"?

**This sentence was removed, it was unclear.**

Page 23, lines 25–26: "Such spread can be considered as a measure of the uncertainty related do vertical mixing and qualitatively correspond to 80-100% of the observed mean concentration."

● I do not understand how the model spread can be considered a measure of the uncertainty related to vertical mixing. Could there not be other differences between the models that are important?

**We admit that this argumentation is a bit short and valid only for primary species. Here we have to remind the results on CO concentrations that show this high variability over the emission zones. Since all models share the raw meteorological variables and since far from emissions area this variability is low, the only explanation comes from the vertical dispersion (Kz) that is differently diagnosed by the models. Particularly in the first layer this will be crucial. We added a sentence on the role of the first layer height that is connected to the vertical diffusion. We have modified the sentence focusing only on primary species.**

**We also wrote in the revised version :"** ***Such spread for primary species and particularly for CO can be considered as a measure of the uncertainty related to vertical mixing and qualitatively corresponds to 80-100% of the observed mean concentration. The height of the first level is also very important for the mixing and deposition processes, it ranges from 20 m for CAMx and CHIMERE to 90 m for EMEP. To be more representative of surface concentrations a correction is implemented for models having a coarse first surface layer (LOTOS-EUROS and EMEP). "*****.**

Page 23, lines 31–32: As pointed out above I do not think that you have shown that the "lower PBL heights (for MINNI) and wind speed (for RCG)" really explain the errors. Also the CMAQ wind speed seems to be as low as the RCG wind speed (according to S0).

**Ok we have removed the sentence, it partly explains but it is too uncertain**

Page 24, line 12: "while EMEP seems more able to capture the evolution of the single PM compound."

● Which single PM compound?

**The PM compounds are the inorganic species : sulfate, nitrate and ammonium, we clarified. This refers to the paper Bessagnet et al. (2014): "*The analysis of each PM compound for the 2009 period (Bessagnet et al., 2014) revealed that MINNI and EMEP were characterized by rather different scores, suggesting that their overall performance is influenced in a different way by both chemistry and meteorology*".**

Page 24, lines 21–22: "The analysis of individual compounds of PM will bring more detailed, it will be investigated in a companion paper."

● Excluding this detailed information from the present paper makes the whole discussion of PM totally uninteresting.

**As we explained at the beginning, the goal of this paper is twofold (i) to present the EURODELTA exercise, the input data and the participating models, and (ii) to analyse the behaviour of models in the four campaigns focusing on the criteria pollutants PM10, PM2.5, O3, NO2 and SO2 and relevant meteorological variables, to our knowledge this has never been addressed in previous papers in a multi model exercise.**

## Language

The manuscript is not very well written, which makes it tedious to read. Large parts of the manuscript needs language editing/corrections. It is not the job of the referees of a paper to correct the language — so I only give some examples below, in the Technical corrections section. Some of the 36 authors of the paper are likely very good at English and, since all authors must have seen the manuscript before submission (according to the obligations for authors), I am surprised that they have accepted the submission without helping to improve the language before the paper was submitted. Please make sure that the whole manuscript is checked carefully if it is resubmitted.

**We thank the reviewer for his help in improving the quality of the manuscript. We agreed and accepted all the comments here below. Sometimes we added a remark in bold character.**

Technical corrections

Page 1 line 37: "period" → "periods"

Page 1 line 38: "allowing evaluating the influence" → "allowing evaluation of the influence"

Page 2 line 5: "good very similar" do you mean "good and very similar"? **Yes we do**

Page 2 line 18: replace "modelling, techniques" by "modelling techniques"

Page 2 line 19: “calculation uncertainty” do you mean “model (or perhaps modelling) uncertainty”? **yes we do**

Page 3 line 7: “exercise” → “exercises”

Page 3 lines 23–24: I guess the list of “non-model” institutes should include NILU as well (since W. Aas is included in the author list)? **Actually we follow the comment of the second reviewer we decided to remove this sentence.**

Page 3 line 28: replace “join analysis” by “joint analysis”

Page 8, line 36: replace “most of countries” with “most countries” or “most of the countries”

Page 9, line 32: The first sentence of the “Sea salt emissions” paragraph is strange. As formulated it does not make sense. **We have corrected** it

Page 11, lines 1–2: “was diagnosed in ECMWF was made available” should probably be “as diagnosed in the IFS-ECMWF model was made available” **Yes**

Page 12, line 12: “most of species” → “most of the species”

Page 12, line 19: “at some EMEP.” → “at some EMEP sites.”

Page 12, line 27: “converted in m/s” → “converted to m/s”

Page 13, line 1: “Being the boundary layer height a concept valid only for convective” → “Since the boundary layer height is a concept valid only for convective”

Page 13, line 21: “compare” → “compared”

Page 13, line 22: “is” → “was”

Page 13, line 22: “characterized by windy conditions in Europe with cool temperature above average everywhere in Europe” — strange formulation; what do you mean by “cool temperature above average”? **Yes we have removed “cool”**

Page 13, line 24–25: “Precipitation were low over the Mediterranean basin but above the climatic average compare to 1961-1990 base period in the rest of Europe.” could be changed to “Precipitation was small over the Mediterranean basin but above the climate average, compared to the 1961-1990 period, in the rest of Europe.” **Yes**

Page 13, line 28: “spells end” → “spells in the end”

Page 13, line 28–29: “After some cold spells end of February, March 2009 turned cooler with on average warmer temperatures compare to the 1961-1990 base period” — strange formulation; did March 2009 turn cooler than the cold spells in the end of February but it was still warmer than the climate average? **Yes “milder” is more appropriated**

Page 14, line 3: “whatever the model” → “for all models”

Page 14, line 6: “this bias exceed” → “this bias exceeds” (or “these biases exceed”) and “whatever the campaign” → “for all campaigns”

Page 14, lines 25–26: “In the IFS only 10m winds are used from ships over the oceans for data assimilation (problem of station representativeness for inland stations).” — awkward formulation — I would suggest something like: “In the IFS only 10m winds from ocean going ships are used in the data assimilation due to problems with station representativity for inland sites.” **Yes it is a better formulation**

Page 14, lines 27–29: “For the lowest winds generally observed during nightime the comparison of the predicted diurnal cycle with observations show a largest positive bias at night than during the afternoon (Fig. 2), this behaviour could lead to an overestimation of the advection process.”

This is a very strange sentence that I do not understand. It needs to be reformulated.

**We changed it as :“For the lowest winds, the comparison of the predicted diurnal cycle with observations shows a larger positive bias at night than during the afternoon (Fig. 1), this behaviour could lead to an overestimation of the advection process in the chemistry transport models”**

Page 15, line 13: “convention” → “convection”

Page 15, line 17: “use the PBL from ECMWF PBL” → “use the PBL from IFS”

Page 15, line 22: “the negative bias of MINNI has the same order of magnitude as the other models” → “the negative bias of MINNI is of the same order of magnitude as those of the other models”

Page 15, line 23: “are still lower” → “are somewhat lower”

Page 15, line 25: “model” → “models”

Page 15, line 28: “on emission areas” → “in emission areas” and “Besides of urban areas” → “Besides in urban areas” (or perhaps “Besides urban areas”)

Page 15, line 29: “that are related to the differences of PBL predicted” → “which is related to the differences in the PBL predicted”

Page 17, lines 14–15: “the mixing of close to emissions is responsible for model output differences” — I think the whole sentence is a bit awkwardly formulated, perhaps this part could be changed to something like: “variations in the PBL height between different models may lead to large differences in modelled concentrations in high-emission areas”

**In the revised versions we replaced by “*…the differences of mixing in models over emission areas lead to large differences in modelled concentrations…*”**

Page 17, lines 32–33: “Differently, differences in diurnal temperature...” — strangely formulated sentence. **The sentence has been removed based on a previous comment**.

Page 18, line 8: “in-deep” → “in-depth”

Page 18, line 9: “This involves a positive bias” → “This leads to a positive bias”

Page 19, line 2: “of the seas” → “over the seas”

Page 19, line 16: “and a few” → “and a little” (or perhaps “and some”)

Page 19, line 27: “all models underestimate” → “all other models tend to underestimate”

Page 19, line 31: “Whatever the campaign” → “For all campaigns”

Page 20, line 13: “are coherent with the completeness of our inventory” — I think a better formulation could be “are consistent with our incomplete inventory”. **We would say better : “*are consistent with the level of the completeness of our inventory*”**

Page 21, line 22: “smaller areas” → “limited areas”

Page 21, lines 25–26: Remove the sentence: “Finally, as already mentioned, PBL heights derived at SIRTA site has been included too.” — this manuscript is too long to state this twice within the same paragraph.

Page 22, line 27: “close between” → “close to”

Page 23, line 8: “mainly driven by a relevant underestimation” → “at least partly driven by a major underestimation”

Page 23, line 9: “CTMs are affordable in reproducing ozone” → “CTMs are able to reproduce ozone”

Page 23, line 11: “nigh-time” → “night-time”

Page 23, line 22: “Likewise ozone” → “Similar to ozone” or “As for ozone”

Page 24, line 1: “rely in chemistry” → “be due to chemistry”

Page 24, line 7: “Differently, the RMSE rises up 15 µg m-3, representing more than 80% of the observed mean.” — incomplete sentence; I guess you mean “rises up to 15 µg m-3 for the campaign XXXX...”? **Ok, we accept this comment.**

Page 24, line 27: “are still missing in state of art CTM” → “are still missing in some state of the art CTMs”

Interactive comment on "Presentation of theEURODELTA III inter-comparison exercise – Evaluation of the chemistry transport models performance on criteria pollutants and joint analysis with meteorology" by B. Bessagnet et al.

Anonymous Referee #4

Received and published: 16 June 2016

**We thank a lot the reviewer for this review of our paper. The answers are written here below in bold characters after each comment.**

General comments

This manuscript is a thorough description of an international model inter-comparison exercise. It can in my view be published in ACP, provided that the comments and concerns below will be taken into account.

The article contains interesting and useful results. However, in my view the discussion of results should focus much more on the results that have some general interest, and on the more general insights and conclusions, and the amount of small details should be substantially reduced. By small details I mean e.g. discussion on how each individual model has performed for each pollutant and each campaign. The amount of figures and tables is also very large; I would advise the authors to reduce these.

**In the revised manuscript we have separated the discussion and the conclusion. The conclusion section is then more general and shorter. We understand the concern of the reviewer on the number of figures, however, we did an important effort to keep the most essential figures, we prefer to keep all of the current figures in the manuscript that bring a lot in the discussion, except Fig. 1 we have removed.**

However, the figures that that make it possible to draw general conclusions should be included. I suggest that the authors would add to conclusions a discussion on the most important improvements of the models, and areas of improvement for the CTM's in general in the future, based on their findings. The terminology also should be more precise, and some of the conclusion more cautious, taking into account the limitations of the data; details are discussed below.

**We have modified the last section of the conclusions as follows : « *The study stresses the importance of emission sources particularly in wintertime, wood burning emissions are likely the most underestimated source, through the missing species called semi-volatile organic compounds. Road traffic emissions could also be underestimated, gasoline and diesel vehicles are both concerned, and more generally all activity sectors involving combustion processes can be concerned. In this study, the importance of meteorological data is highlighted, the difficulties for meteorological models to simulate meteorological variables like wind speed and PBL height during stable conditions can lead to dramatic consequences on air quality modelling. Developments in air quality modelling have not only to focus on processes but also on emissions and meteorological***

***input data. To complement the analysis, companion papers will focus on depositions of sulphur/nitrogen compounds and on the behaviour of models for particulate matter species. This ensemble of analyses will help to prioritize the improvement of air quality models used in the frame of the CLRTAP*»**

Specific comments

Abstract. Explain which experimental datasets were used, and how many stations were included, please. 'Background stations', specify which background; probably regional background, not urban or global background. The discussion would be in my view more clear, if the evaluation of met parameters would be presented first, then evaluation of concentrations. 'performances were good', specify what is meant with 'performance', do you mean e.g. bias or correlations, or both ? PM, specify which PM fraction.

**Here is the new abstract :**

**" The EURODELTA III exercise has facilitated a comprehensive inter-comparison and evaluation of chemistry transport model performance. Participating models performed calculations for four one-month periods in different seasons in the years 2006 to 2009, allowing the influence of different meteorological conditions on model performances to be evaluated. The exercise was performed with strict requirements for the input data, with few exceptions. As a consequence, most of differences in the outputs will be attributed to the differences in model formulations of chemical and physical processes. The models were evaluated mainly for background rural stations in Europe. The performance was assessed in terms of bias, root mean square error and correlation with respect to the concentrations of air pollutants ($NO_2$, $O_3$, $SO_2$, PM10 and PM2.5), as well as key meteorological variables. Though most of meteorological parameters were prescribed, some variables like the planetary boundary layer (PBL) height and the vertical diffusion coefficient were derived in the model pre-processors and can partly explain the spread in model results. In general the day time PBL height is underestimated by all models. The largest variability of predicted PBL is observed over the ocean and seas. For ozone, this study shows the importance of proper boundary conditions for accurate model calculations and then on the regime of the gas and particle chemistry. The models show similar and quite good performance for nitrogen dioxide, whereas they struggle to accurately reproduce measured sulphur dioxide concentrations (for which the agreement with observations is the poorest). In general, the models provide a close-to-observations map of particulate matter (PM2.5 and PM10) concentrations over Europe with rather correlations in the range 0.4 – 0.7 and a systematic underestimation reaching -10 μg $m^{-3}$ for PM10. The highest concentrations are much more underestimated particularly in wintertime. Further evaluation of the mean diurnal cycles of PM reveals a general model tendency to overestimate the effect of the PBL height rise on PM levels in the morning, while the intensity of afternoon chemistry leading to formation of secondary species to be underestimated. This results in larger modelled PM diurnal variations than the observations show and this is so for all seasons. The models tend to be too sensitive to the daily variation of the PBL. All in all, in most cases model performances are more influenced by the model set-up than the season. The good representation of temporal evolution of wind speed is most responsible for models' skillfulness in reproducing the daily variability of pollutant concentrations (e.g. the development of peak episodes), while the reconstruction of the PBL diurnal cycle seems to play a larger role in driving the corresponding**

**pollutant diurnal cycle and hence determine the presence of systematic positive and negative biases detectable on daily basis."**

Introduction. In discussing model inter-comparisons, refer also to the most recent relevant ones, especially Prank et al, 2016, ACP (16, 6041–6070). ". . . showed better performance but higher uncertainty. . .' define what is meant with 'performance' and what you mean with 'uncertainty'. The institutes participating. . . this sentence should be deleted; not scientifically relevant information. 'criteria pollutants': define concept (which criteria ? defined by whom ?); probably the authors refer to the latest EU directives or limit values (?); but that should then be specified.

**Yes, we have included the references and taken into account all the remarks. Criteria pollutants refer to the Air quality directives, we have modified accordingly. Uncertainties are related to the model formulation (parameterization) and input data. We modified it as :"** ***The objective of this paper is twofold, (i) to present the exercise, the input data and the participating models, and (ii) to analyse the behaviour of models in the four campaigns focussing on the criteria pollutants PM10, PM2.5, O3, NO2 and SO2 as defined in the EU directive on air quality 2008/50/EC (EC, 2008), and relevant meteorological variables.*****"**

Methods. p 8 'lowest levels of emissions': emissions of which pollutant ?

**They refer to $PM_{2.5}$ for the residential sector, we clarified it in the revised version.**

Discussion. p 23: 'model formulation and setup . . . more influencing than met conditions'.

Define what is meant with 'model formulation and set-up' (is it the setup of input data, which ones ? set-up of model parameters and submodels, which ones ?;or selection of CTM's themselves ?). This statement is also over-interpretation; it has only been shown to be valid for the range of met parameters that were included in the selected conditions, which was not especially wide. Please re-write this, allowing for the limitations of the data used.

**We replaced this statement by «** ***This confirms once again that on average and for the limited dataset used in this exercise, the model formulation (parameterization of chemical / physical processes, calculation of meteorological diagnosed variables) and set-up (number of vertical levels, value of key parameters, etc...) are more influencing than raw meteorological conditions on model performance.*** ».

p. 23. 'highest errors': which stat. model evaluation parameter is meant by 'error' ?

**We have replaced by RMSE instead of error.**

Technical corrections

I would also suggest that the whole text and the language will be checked, and the fairly numerous misprints and language mistakes will be corrected.

**Yes, we have revised the language; there were several misprints and mistakes.**

# Presentation of the EURODELTA III inter-comparison exercise - Evaluation of the chemistry transport models performance on criteria pollutants and joint analysis with meteorology

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**Abstract.**

The EURODELTA III exercise has facilitated a comprehensive inter-comparison and evaluation of chemistry transport model performances. Participating models performed calculations for four one-month periods in different seasons in the years 2006 to 2009, ~~thus~~ allowing ~~evaluation of~~ the influence of different meteorological conditions on model performances to be evaluated. The exercise was performed ~~under~~with strict requirements ~~concerning~~for the input data, ~~despite some~~with few exceptions. As a consequence, most of differences in the outputs will be attributed to the differences in model

formulations of chemical and physical processes. The models were evaluated mainly for background rural stations in Europe. The performance~~s~~ was ~~are~~ assessed in terms of bias, root mean square error~~s~~ and correlation~~s,~~ with respect to the concentrations of air pollutants ($NO_2$, $O_3$, $SO_2$, PM10 and PM2.5), as well as key meteorological variables. Though most of meteorological parameters were prescribed, some variables like the planetary boundary layer (PBL) height and the vertical diffusion coefficient were ~~diagnosed~~derived in the model pre-processors and can partly explain the spread in model~~s~~ results. In general the day time PBL height is underestimated by all models. The~~, the~~ largest variability of predicted PBL is observed over the ocean and seas. For ozone, this study shows the importance of proper boundary conditions for accurate model calculations and then on the regime of the gas and particle chemistry. The models show similar and quite good performance for nitrogen dioxide, whereas they struggle to accurately reproduce measured sulphur dioxide concentrations (for which the agreement with observations is the poorest). In general, the models provide a close--to--observations map of particulate matter (PM2.5 and PM10) concentrations over Europe with rather correlations in the range 0.4 – 0.7 and a systematic underestimation reaching -10 µg $m^{-3}$ for PM10. The highest concentrations are much more underestimated particularly in wintertime. Further evaluation of the mean diurnal cycles of PM reveals a general model~~s~~' tendency to overestimate the effect of the PBL height rise on PM levels in the morning, while the intensity of afternoon chemistry leading to formation of secondary species to be underestimated ~~by the models~~. This results in larger modelled PM diurnal variations than~~, whereas~~ the observations show ~~rather flat PM diurnal profiles~~ and this is so for all seasons. The models tend to be too sensitive to the daily variation of the PBL. All in all, in most cases model performances are more influenced by the model set-up than the season. The good representation of temporal evolution of wind speed is most responsible for models' skillfulness in reproducing the daily variability of pollutant concentrations (e.g. the development of peak episodes), while the reconstruction of the PBL diurnal cycle seems to play a larger role in driving the corresponding pollutant diurnal cycle and hence determine the presence of systematic positive and negative biases detectable on daily basis.

~~The EURODELTA III exercise allows a very comprehensive inter-comparison and evaluation of chemistry transport models performance. Participating models were applied over four different one month period, within a rather limited number of years (from June 2006 to March 2009) thus allowing evaluating the influence of different meteorological conditions on model performance. The exercise was performed under strict requirements concerning the input data. As a consequence, there were very limited differences in the models set up, representing a sort of sensitivity analysis to several aspects of the modelling chains. The models were evaluated mainly on background stations. Even if the meteorology was prescribed, some variables like the planetary boundary layer (PBL) height, the vertical diffusion coefficient are diagnosed in the model pre-processors and explain the spread of models results. For ozone, this study shows the importance of boundary conditions on model calculations and then on the regime of the gas and particle chemistry. The worst performances are observed for sulphur dioxide concentrations that are poorly captured by the models. The performances of models are rather good very similar for the nitrogen dioxide. On average, the models provide a rather good picture of the particulate matter (PM) concentrations over Europe even if the highest concentrations are underestimated. For the PM, the mean diurnal cycles show a general tendency to overestimate the effect of the PBL height rise while the afternoon chemistry (formation of secondary~~

~~species) is certainly underestimated, PM observations show very flat diurnal profiles whatever the season. In general the day time PBL height is underestimated by all models, the largest variability of predicted PBL is observed over the ocean and seas. More generally, in most cases model performances are more influenced by the model setup than the season. The temporal evolution of wind speed is most responsible of model skilfulness in reproducing the daily variability of pollutant concentrations (*e.g.* the development of peak episodes), while the reconstruction of the PBL diurnal cycle seems more influencing in driving the corresponding pollutant diurnal cycle and hence the presence of systematic positive and negative biases detectable on daily basis.~~

## 1 Introduction

The ongoing project EURODELTA has very successfully extended the European Air Quality Modelling capability by providing a forum in which modelling teams could share experiences in simulating technically interesting and policy relevant problems. The joint exercises contribute to further improve modelling~~,~~ techniques as well as to quantify and understand the sources of model~~calculation~~ uncertainties~~y~~ related to the parameterization of processes and the quality of input data. EURODELTA is now an activity contributing to the scientific work of the UNECE (United Nations Economic Commission for Europe) Task Force on Measurement and Modelling (TFMM) under the Convention on Long-range Transboundary Air Pollution (CLRTAP). The TFMM was established in 2000 to ~~offer~~ provide a forum to the Parties, the EMEP (European Monitoring and Evaluation Programme) centres and other international organizations for scientific discussions to evaluate measurements and modelling and to further develop working methods and tools. These are used for policy studies in support of ~~.~~ ~~In that context,~~ the Gothenburg Protocol signed in 1999 which is a multi-pollutant protocol of the Convention designed to reduce acidification, eutrophication and ground-level ozone by setting emissions ceilings for sulphur dioxide, nitrogen oxides, volatile organic compounds, fine particulate matter and ammonia.

In 2004, EURODELTA I (van Loon *et al.*, 2007) examined the common performance of the chemistry transport models (CTM) in predicting recent (2000) and future (2020) air quality in Europe using the concept of a model ensemble to measure robustness of predictions. The spread of model predictions about the *ensemble* mean gave a measure of uncertainty for each predicted value. In a 2020 world the effect of making emission reductions for key pollutants in specific geographic areas was investigated. The pollutants were ~~of~~ NOx (nitrogen dioxide), $SO_2$ (sulphur dioxide), VOC (Volatile Organic Compound), PM (particulate ~~matter, as~~matter as PM10 and PM2.5 ~~respectively~~ for particle diameters below 10 µm and 2.5 µm respectively) and $NH_3$ (ammonia). The countries were ~~independently in~~ France, Germany and Italy. ~~and,~~the effect of reducing ~~and of~~ NOx and SOx in sea areas~~,~~ was also investigated. Source-receptor relationships used in integrated assessment (IA) modelling were derived for all the models and compared to assess how model choice might affect this key input. EURODELTA II (Thunis *et al.*, 2008) ~~was~~ built on this project by taking a closer look at how the different models represent the effect on pollutant impacts on a European scale by applying emission reductions to individual emission sectors.

In the recent literature, several inter-comparison and evaluation exercises of regional-scale chemistry transport models for PM ~~are~~ have been reported ~~in the literatur~~for PM models : McKeen *et al.* (2007), van Loon *et al.*, (2007), Vautard *et al.* (2007), Hayami *et al.* (2008), Stern *et al.* (2008), Smyth *et al.* (2009), Vautard *et al.* (2009), Solazzo *et al.* (2012), Pernigotti, *et al.* (2013) and Prank *et al.* (2016). ~~Most of these model inter-comparison exercises were performed at the regional scale with chemistry transport models~~. In one of the most recent exercises, AQMEII (Solazzo *et al.*, 2012), models clearly tend to underestimate ~~$PM_{10}$~~PM10 background concentrations in US and EU regions. Model results for ~~$PM_{2.5}$~~PM2.5 concentrations showed better performances but large uncertainty remained certainly due to the simulation of secondary organic aerosols. Prank *et al.* (2016) stressed the problems of emission underestimates to explain the model discrepancies.

The new EURODELTA III (ED-III) exercise was ~~promoted~~ designed to exploit and interpret intensive measurement campaigns carried out by~~the~~ EMEP (Aas *et al.*, 2012)~~intensive measurements~~ As far as possible ~~Differently to the previous inter-comparison exercises, most of~~the models have been run in ED-III with the same input data (emissions, meteorology, boundary conditions) and over the same domain (domain extension and resolution)~~, with some rare exceptions~~ This distinguishes the study from other model inter-comparisons. ~~, by making modelling analysis of the campaigns to re-examine the performance on usual error statistics (bias, root mean square error, correlation) of chemistry transport models. The~~ ED-III ~~exercise has~~ focussed on four EMEP intensive measurement periods ~~(Aas *et al.*, 2012)~~:

- 1 Jun - 30 Jun 2006
- 8 Jan - 4 Feb 2007
- 17 Sep - 15 Oct 2008
- 25 Feb - 26 Mar 2009

~~Differently to the previous inter-comparison exercises, most of models have been run in ED-III with the same input data (emissions, meteorology, boundary conditions) and over the same domain (domain extension and resolution) with some rare exceptions.~~ The ~~Participating models were applied over~~ four different periods, within a rather limited number of years, ~~thus~~ allowed ~~ing to evaluate~~ the influence of different meteorological conditions on model performance to be evaluated. ~~All models except RCG have run the four periods~~. The ~~institutes/laboratories~~list of modelling teams participating in the ED-III ~~with their models are~~ is reported in Table 1~~Table 1~~. FUB ran two of the four periods ~~The other participants JRC, BSC, CIEMAT, CONCAWE, AERIS EUROPE, LMD/IPSL and University of Brescia contributed to the project bringing their expertise in air quality modelling, model evaluation and management of observational data.~~ The ED-III framework (emissions, model configurations) was also used to assess the impact of the horizontal resolution on the performance of air quality models (Schaap *et al.*, 2015).

The ED-III exercise allowed~~s~~ a very comprehensive inter-comparison and evaluation of chemistry transport model~~s~~ performance with a joint analysis of some meteorological variables to be made. A first evaluation on the 2009 campaign with an interim version of models was ~~already~~ published in Bessagnet *et al.* (2014). Moreover, the selected periods coincide with EMEP intensive measurement periods so that an extended set of observational data were available. Therefore, in addition to EMEP operational monitoring data, ~~also~~ size disaggregated (in ~~$PM_{2.5}$~~PM2.5 and ~~$PM_{10}$~~PM10) aerosol data~~s~~ and hourly measurements for studying diurnal cycles have been used~~employed~~. Additional AirBase data (Mol and de Leeuw, 2005)

were used to evaluate the impact of meteorology on air pollutant concentrations. Finally, the exercise was performed under strict requirements (with some exceptions) concerning the input data. As a consequence, most of differences in the outputs will be attributed to the simulation of chemical and physical processes~~As a consequence, there were very limited differences in how the models were set up, representing a sort of sensitivity analysis to several aspects of the modelling chains~~. The objective of this paper is twofold, (i) to present the exercise, the input data and the participating models, and (ii) to analyse the behaviour of models in the four campaigns focussing on the criteria pollutants ~~$PM_{10}$~~PM10, ~~$PM_{2.5}$~~PM2.5, $O_3$, $NO_2$ and $SO_2$ as defined in the EU directive on air quality 2008/50/EC (EC, 2008), and relevant meteorological variables. Complementary analyses of depositions~~ fluxes and PM composition data at high temporal resolution will be discussed in companion papers in order to better understand the behaviour of models.

## 2 Description of models

### 2.1 Overall description of models

The models are synthetically described in Table 2~~Table 2~~ and Table 3~~Table 3~~. All the models were run on the same domain at 0.25°x0.25° resolution in longitude and latitude~~, shown in Fig. 1,~~ except CMAQ. CMAQ simulations were performed on a Lambert-conformal conic projection with the standard parallels at 30 and 60 degrees and a grid of 112 by 106 cells of size 24km x 24km. The results of the CMAQ simulations were interpolated to the prescribed EURODELTA grid.

Participants delivered both air concentrations and meteorological parameters. Most of variables were delivered on an hourly basis, while dry and wet deposition fluxes were provided on a daily basis. The output species include, among others: $O_3$, $NO_2$ and $SO_2$, total PM mass concentrations both in 2.5 and 10 µm fractions (~~$PM_{10}$~~PM10 and ~~$PM_{2.5}$~~PM2.5)~~,~~ Secondary ~~I~~inorganic a~~A~~erosols such as ~~(~~ammonium $(NH_4^+)$, sulphate $(SO_4^{2-})$ and nitrate $(NO_3^-)$~~)~~ and other PM components relevant for the analysis as well as wet deposition of sulphur and nitrogen compounds were also collected and will be used in companion papers. The delivered air concentrations should approximately correspond to the standard measurement height (typically 3 m) and were directly derived from the first model layer, except for LOTOS-EUROS and EMEP that corrected the concentrations from the first layer to be representative of 3-m concentrations. The ~~$PM_{2.5}$~~PM2.5 and ~~$PM_{10}$~~PM10 concentration are calculated as follows in each model:

$$PM_{xx} = PPM_{xx} + |SO_4^{2-}|_{xx} + |NO_3^-|_{xx} + |NH_4^+|_{xx} + |SOA|_{xx} + |Dust|_{xx} + |Sea\ Salts|_{xx}$$

~~$PM_{xx}$ = PPM + $SO_4^{2-}$ + $NO_3^-$ + $NH_4^+$ + Sea Salt + SOA + Dust~~

where xx=2.5 µm or 10 µm, PPM stands for Primary Particulate Matter and includes Elemental carbon, Primary organic aerosol and primary non-carbonaceous aerosol, SOA represents Secondary Organic Aerosol, *Sea Salt* and *Dust* represent the contribution of the corresponding natural processes mainly controlled by the wind speed.

The participating models differ in the availability of PM components and formation routes. For instance, EMEP, LOTOS-EUROS and RCG contain coarse mode nitrate formation (produced by reaction of nitric acid with sea salts and dust),

whereas the others do not. In CMAQ additional anthropogenic dust is calculated as 90% of unspecified PM coarse emissions and attributed to fugitive dust (Binkowsky and Roselle, 2003). ~~CAMx~~ CAMx did not activate the parameterisation of sea salts ~~parameterisation~~ in this exercise.

Based on the set-up of models and completeness of datasets, an "ENSEMBLE" called **ENS** has been built based on mean values of model outputs. To compare the behaviour of models for all pollutants and campaigns, only CHIMERE, MINNI, LOTOS-EUROS and EMEP constitute the "ENSEMBLE". ~~CAMx~~ CAMx, CMAQ and RCG were not included in the ensemble for three reasons: (i) ~~CAMx~~ CAMx did not account for sea salts ~~giving rise~~leading to very different PM patterns over the oceans and seas, (ii) CMAQ used a different meteorology and (iii) RCG did not cover the four campaigns.

### 2.2 PBL height and mixing in models

**~~CAMx~~ CAMx**

In ED-III the Planetary Boundary Layer was directly taken from the IFS-ECMWF data (Integrated Forecast System of the European Centre for Medium-Range Weather Forecasts). The PBL height was then used by ~~CAMx~~ CAMx pre-processor to derive $K_z$ profiles. For ED-III the O'Brien scheme (1970) has been used to derive $K_z$ profiles as Eq.1:

$$K_z = K_A + \frac{(z-z_A)^2}{(z_A-z_B)^2}\left\{K_B - K_A + (z-z_B)\left(K'_B + 2\frac{K_B-K_A}{z_A-z_B}\right)\right\} \quad \text{(Eq. 1)}$$

Where $K_z$ is a value of $K_A$ at the height of the atmospheric boundary layer, $z_A$, and $K_B$ at the height of the surface layer $z_B$, the so-called constant-flux layer. Minimum $K_z$ values have been set to 1 $m^2\ s^{-1}$. Any values of $K_z$ calculated below, will be set to this value. By default, ~~CAMx~~ CAMx employs a standard "K-theory" approach for vertical diffusion to account for sub-grid scale mixing layer-to-layer.

**CHIMERE**

In this study, the Planetary Boundary Layer is directly taken from the IFS ECMWF data. Horizontal turbulent fluxes were not considered. Vertical turbulent mixing takes place only in the boundary layer. The formulation uses K-diffusion following the parameterization of (Troen and Mahrt, 1986), without a counter-gradient term. In each model column, diffusivity $K_z$ is calculated as Eq. 2:

$$K_z = \mathrm{k}\,\mathrm{w_s}\,\mathrm{z}\left(1-\frac{\mathrm{z}}{\mathrm{h}}\right)^{1/3} \quad \text{(Eq. 2)}$$

where $w_s$ is a vertical velocity scale given by similarity formulae.

- In the stable case (surface sensible heat flux < 0): $w_s = u_*/(1 + 4.7\,z/L)$
- In the unstable case: $w_s = (u_*{}^3 + 2.8 e w_*{}^3)^{1/3}$

where $e$ = *max(0.1,z/h)*, $L$ is the Monin-Obukhov Length, $w_*$ is the convective velocity scale, $u_*$ the friction velocity and $h$ the boundary layer height. The~~A~~ minimum value of~~al~~ $K_z$ is assumed to ~~have~~,be ~~with a value of~~ 0.01 $m^2\ s^{-1}$.

$K_z$ and the wind speed were corrected in urban zones according Terrenoire *et al.* (2015) by applying a correction factor to limit the diffusion within the urban canopy, but this correction has very little effect at this resolution.

**CMAQ**

The boundary layer height in COSMO is calculated with the turbulent kinetic energy (TKE) method (Doms *et al.* 2011). CMAQ directly used the PBL fields from COSMO.

In CMAQ the vertical turbulent mixing is estimated using the Asymmetric Convective Model scheme version 2 (ACM2, Pleim, 2007a,b). The ACM2 replaces the simple eddy viscosity (K-theory) scheme. ACM2 scheme allows the non-local mixing, which means upward turbulent mixing from the surface across non-adjacent layers through the convective boundary layer. Pleim (2006) compared the eddy viscosity and the ACM2 schemes in CMAQ, finding that the ACM2 schemes tends to predict larger concentrations of secondary pollutants and smaller concentrations of primary pollutants at the surface, and has a more well-mixed profile in the PBL than the eddy viscosity scheme.

CMAQv5 has also an improved version of the minimum allowable vertical eddy diffusivity scheme. The new version interpolates between urban and nonurban land cover, allowing a larger minimum vertical diffusivity value for grid cells that are primarily urban. Moreover, the minimum eddy diffusivity values were reduced from 0.5 $m^2$ $s^{-1}$ to 0.01 $m^2$ $s^{-1}$, and from 2.0 $m^2$ $s^{-1}$ to 1.0 $m^2$ $s^{-1}$ for urban areas.

**EMEP**

The mixing height is calculated using a slightly modified Richardson number ($Ri_B$) following Jeričevič *et al.* (2010) and defined as the lowest height at which the $Ri_B$>0.25. Finally, the PBL is smoothed with a second order Shapiro filter in space. The PBL height is not allowed to be less than 100 m or exceed 3000 m.

The initial calculation of the vertical exchange coefficients is done using the Ri number and wind speed vertical gradient for the whole domain. Then, $K_z$ values within the PBL are recalculated based on Jeričevič *et al.* (2010) for stable and neutral conditions. For unstable situations $K_z$ is calculated based on the similarity theory of Monin-Obukhov for the surface layer, whereas $K_z$ profiles from O'Brian (1970) are used for the PBL above the surface layer. For more detail see Simpson *et al.* (2012).

**LOTOS-EUROS**

The first model layer is by definition the mixing layer, with height equal to the boundary layer height as given by ECMWF. Horizontal diffusion is not used, but for vertical mixing the vertical diffusion coefficient is calculated according to Eq. 3:

$$K_z = \frac{\kappa u^*}{\Phi(z/L)} \qquad \text{(Eq. 3)}$$

where $\kappa$ is the von Karman constant, $u^*$ the friction velocity, $\Phi$ the functions proposed by Businger (1971) for stable, neutral or unstable atmosphere, $z$ the height and $L$ the Monin-Obhukov length. The friction velocity is calculated depending on the wind at reference height (10 m), the Businger functions and the roughness length per land use class. The vertical structure of LOTOS-EUROS is determined by the mixing layer height, with a shallow surface layer (25 m) to avoid too fast mixing of near-surface emissions and a second layer equal to the mixing layer as given by ECMWF.

**MINNI**

In MINNI, the ~~F~~friction velocity and Monin-Obukhov length are determined by using the~~:~~ Holtslag and van Ulden (1983) iterative scheme for unstable conditions and the Venkatram (1980) iterative method for stable conditions. Micro-meteorological parameters over water are derived with the profile method, using air-sea temperature difference (Hanna *et al.*, 1985), with the needed roughness length, depending on wind speed, supplied by the Hosker (1974) parameterization.
During daytime both convective and mechanical heights are determined, keeping then the maximum value between the two parameters. The convective height is calculated following the Maul (1980) version of Carson (1973) algorithm, essentially based on heat conservation equation. The~~;~~ mechanical mixing height is ~~instead~~ estimated by using the Venkatram (1980) algorithm. During night-time, the Bulk Richardson number method is applied (Sorensen, 1998), in which the height of the boundary layer is given by the smallest height at which the bulk Richardson number reaches the critical value fixed to 0.25.

**RCG**

The mixing layer depth in the model is the height of the layer closest to the input boundary layer height taken from the IFS ECMWF data. Vertical diffusion parameters for stable and unstable conditions are derived using the Monin-Obukhov similarity theory for ~~the description of the structure of~~ the diabatic surface layer. The friction velocity and Monin-Obukhov-length are calculated iteratively depending on the 10 m wind, the stability correction~~s~~ factors and the roughness length determined from land use. ~~dependent on roughness length.~~

# 3 Input data

## 3.1 Anthropogenic emissions

The first step in the emission preparation was to calculate the spatial pattern of emissions for the reference year 2007, that was selected because it was a key year for the TNO-MACC inventory (Kuenen *et al.*, 2011). The anthropogenic emission input was harmonized following the methodology described in Terrenoire *et al.* (2015). The total emissions per sector and country were then scaled to the ~~year~~ corresponding year ~~to~~of the campaigns: 2006, 2007, 2008 and 2009. Emission categories are ~~numbered~~ broken down into~~in~~ 11 classes called SNAP (Selected Nomenclature for Air Pollutants): (1) Public Power stations, (2) Residential and Comm./inst. Combustion, (3) Industrial combustion, (4) Production processes, (5)

Extraction and distribution fossil fuel, (6) Solvents use, (7) Road traffic, (8) Other mobile sources (trains, shipping, aircrafts, ...), (9) Waste treatment, and (10) Agriculture. and , (11), Natural emissions (11) were calculated by the (models as set out in section 3.2 used their own natural emissions in this exercise).

The gridded distribution of anthropogenic emissions was provided by INERIS and it was based on a merging of different databases from:

- TNO 0.125°×0.0625° emissions for 2007 from MACC (Kuenen *et al.*, 2011)
- EMEP 0.5°×0.5° emission inventory for 2009 (Vestreng *et al.*, 2007)
- Emission data from the GAINS database (http://gains.iiasa.ac.at/gains).

Emission re-gridding was based on INERIS expertise and performed by means of various proxies:

- population data coming from the EEA database merged with global data (from the Socioeconomic Data and Applications Center http://sedac.ciesin.columbia.edu) to fill gaps in Europe.;
- the US Geophysical Survey land use at 1 km resolution (http://www.usgs.gov/).
- French bottom-up emission data for wood combustion to derive a proxy based on population density;;
- EPER data for industries; the EPER Decision is based on Article 15(3) of Council Directive 96/61/EC (EC, 1996) concerning integrated pollution prevention and control. EPER is a web-based register, which enables the public to view data on emissions to water and air of 50 key pollutants from large and medium-sized industrial point sources in the European Union.

The TNO-MACC dataset provides two distinct datasets (i) large point sources (LPS) with the coordinates of stacks and (ii) surface emissions on a fine grid (0.125°×0.0625°). In the gridding process, the first step consisted in summing up LPS emissions from the TNO-MACC emissions inventory for 2007 with surface emissions to obtain total emissions as in the EMEP inventory. LPS were aggregated with surface emissions because no data were available to calculate plume rise heights for point sources emissions. For the various SNAP sectors, the processing steps were the following:

- SNAP 2: The country emissions were re-gridded with coefficients based on population density and French bottom-up data, the methodology (Terrenoire *et al.*, 2015) was extrapolated to the whole Europe. For PM2.5 emissions, the annual EMEP national totals were kept except for the countries: Czech Republic, Bosnia and Herzegovina, Belgium, Belarus, Spain, France, Croatia, Ireland, Lithuania, Luxemburg, Moldavia, Republic of Macedonia, Netherland, Turkey. For these countries, PM2.5 emissions from GAINS were used as this database provides higher numbers and certainly more realistic since wood burning is known to be underestimated in the EMEP database (Denier van der Gon *et al.*, 2015). Additional factors were applied on two Polish regions for both PM2.5 and PM10 emissions. As a preliminary solution, domestic combustion emissions from provinces with active coal mines were multiplied by a factor of 8, while those in neighbouring provinces were adjusted by a factor of 4 (Kiesewetter *et al.*, 2015).~~The country emissions were re-gridded with coefficients based on population density and French bottom-up data, the methodology (Terrenoire *et al.*, 2015) was extrapolated to the whole Europe. For $PM_{2.5}$ emissions, the annual EMEP national totals were kept except for the countries: Czech Republic, Bosnia and Herzegovina, Belgium, Belarus, Spain, France, Croatia, Ireland, Lithuania, Luxemburg, Moldavia, Republic of Macedonia, Netherland, Turkey. For these countries, $PM_{2.5}$ emissions from GAINS were used. Additional factors were applied on two Polish regions (×4 or ×8) for $PM_{2.5}$ and $PM_{10}$ emissions (Kiesewetter *et al.*, 2015)~~. The former activity in coal mine regions still leads to high emissions of PM due to domestic uses of coal.

- SNAP 3,7,8,9,10: TNO-MACC emission spatial distribution was used as proxy to regrid EMEP 0.5°x0.5° annual totals into the finer modelling grid.

- SNAP 1,4,5,6: EMEP 0.5°x0.5° emissions were regridded by using "artificial area" (or built-up area), except for industries where EPER data were used.

For countries where TNO-MACC emissions were not available, the EMEP 0.5°×0.5° emissions were used (Iceland, Liechtenstein, Malta and Asian countries) and regridded with adequate proxies ("artificial land use", EPER data for industries).

The following emitted species were used in the models: methane (this species comes from the TNO-MACC inventory), carbon monoxide, ammonia, sulphur oxides, non-methane volatile organic compounds (NMVOC), nitrogen oxides, primary particulate matter.

Residential emissions of particulate matter are dominant in wintertime. In ~~in~~ most ~~of~~ countries~~;~~ they come from wood burning or coal uses. Germany, Sweden, Spain clearly have the lowest levels of PM2.5 emissions for this activity sector. Romania, Poland and France have the highest levels of annual total emissions per country (Terrenoire *et al.*, 2015). For this activity sector, the PM2.5 emissions by components are provided in supplementary material S8.

The time profiles are those used in Thunis *et al.* (2008). Three types of profiles were provided:

- Seasonal factors : one value per species, month, activity sector and country
- Weekly factors : one value per species, day type (Monday – Sunday), activity sector and country
- Hourly factors : one value per hour (local time), species and activity sector

The vertical injection profile in CTMs was prescribed according to Bieser *et al.* (2011) where industrial sectors and residential heating were assigned in lower levels compared to the lower vertical levels than other literature default profiles ~~usual default profiles~~ (Mailler *et al.*, 2013).

Since only ~~$PM_{2.5}$~~PM2.5 and coarse PM emissions were provided by ~~the~~ EMEP, a PM speciation profile provided by IIASA (~~Personal Communication from IIASA~~based on Klimont *et al.*, 2013) was used to estimate the fraction of n~~N~~on-carbonaceous species, Elemental Carbon and Organic Matter per activity sectors and country. Models used their own split for NOx, SOx and NMVOC emissions. This emission inventories did not account to recent changes in the way to account for Semi Volatile Organic Compounds from wood burning emissions as discussed in Denier van der Gon *et al.* (2015).

The full emission dataset is available on request to INERIS.

### 3.2 Natural emissions

***Biogenic VOC emissions from vegetation***

CHIMERE and MINNI used the version 2.04 of the MEGAN model while ~~CAMx~~ CAMx use~~d~~s the 2.1 version (Guenther *et al.*, 2006, 2012). The Model of Emissions of Gases and Aerosols from Nature (MEGAN) is a modelling framework for estimating fluxes of biogenic compounds between terrestrial ecosystems and the atmosphere using simple mechanistic

algorithms to account for the major known processes controlling biogenic emissions. It is available as an offline code and has also been coupled into land surface and atmospheric chemistry models.

EMEP, LOTOS-EUROS and RCG used parameterizations derived from Simpson *et al.* (1999) for the temporal variations according to temperature and light, with maps of tree species from Koeble and Seufert (2001).

CMAQ used the BEIS (Biogenic Emission Inventory System: Vukovich and Pierce, 2002) module developed by the US EPA. BEIS estimates volatile organic compound (VOC) emissions from vegetation and nitric oxide (NO) and carbon monoxide (CO) emissions from soils. Because of resource limitations, recent BEIS development has been incorporated into the Sparse Matrix Operational Kernel Emissions (SMOKE) system (available at https://www.cmascenter.org/smoke), so that the native version of BEIS is built within the SMOKE architecture.

***Soil ~~N~~Nitrogen Monoxide (NO)~~O~~ emissions***

CHIMERE and MINNI used the version 2.04 and CAMX used version 2.1 of the MEGAN model to calculate the NO~~CHIMERE, CAMx and MINNI used the version 2.04 of the MEGAN model to calculate the NO~~ emissions. RCG used a parameterization of NO emissions described in Simpson *et al.* (1999). LOTOS-EUROS did not include NO emissions in this simulation. CMAQ used the BEIS (Biogenic Emission Inventory System) module developed by the US EPA. ~~NO~~ The ~~s~~Ssoil NO emission parameterization ~~s~~ for EMEP ~~are~~ is described in Simpson *et al.* (2012)

***Sea salt emissions***

All models host very different schemes based on Monahan (1986) ~~for CHIMERE and~~ with some updates from Martensson *et al.* (2003) for LOTOS-EUROS, and Gong *et al.* (1997) for RCG. CMAQ and MINNI used the Zhang *et al.* (2005) parameterization and ~~CAMx~~ CAMx had no sea salts for this exercise due to too high uncertainty in sea salt parameterization. EMEP used parameterisation from Monahan (1986) for larger sizes of sea spray and Martensson *et al.* (2003) for smaller sizes.

CMAQ emits also sea salts sulphate using a fraction of 7.76% of emitted sea salts split into the accumulation and coarse modes.

***NO emissions from lightning***

The only model to describe ~~Climatologies of~~ NO emissions from lightning is the EMEP model, following ~~based on~~ Köhler *et al.* (1995) ~~in EMEP. The other models do not account for this kind of emissions.~~

***Wildfire emissions***

Fire emissions were provided by the GFASv1.0 database (Kaiser *et al.*, 2012) only for the 2006 campaign. The Global Fire Assimilation System (GFASv1.0) calculates biomass burning emissions by assimilating Fire Radiative Power (FRP) observations from the MODIS instruments onboard the Terra and Aqua satellites. It corrects for gaps in the observations,

which are mostly due to cloud cover, and filters spurious FRP observations of volcanoes, gas flares and other industrial activities. For all models the wildfire emissions were assigned in the whole PBL layer. Only the following species were taken into account: CO, $CH_4$, NOx, $SO_2$, PM2.5, TPM (Total Primary Matter), OC (Organic Carbon) and EC (Elemental Carbon).

***Dust emissions***

For CAMx CAMx, CHIMERE and CMAQ, no natural dust module is activated for this exercise. For these three models, natural dust only comes from the boundary conditions. For EMEP, windblown dust parameterisation is documented in Simpson *et al.* (2012), road dust calculations are included in the calculations from Denier van der Gon *et al.* (2009). LOTOS-EUROS contains emission parameterizations for several sources of mineral dust (Schaap *et al* 2009). Only wind-blown dust, resulting from wind erosion of bare soils, was taken into account here, together with dust from boundary conditions. Other sources (agricultural activities, road dust resuspension) were not activated in ED-III. In MINNI, dust emissions from local erosion and particle resuspension (Vautard *et al.*, 2005) with attenuation in the presence of vegetation from Zender *et al.* (2003) is activated in this exercise. RCG considers resuspension of mineral aerosol as a function of friction velocity and the nature of soils. Two mechanisms are treated: direct release of small dust particles by the wind (Loosmore and Hunt, 2000), and indirect release by collisions with bigger soil grains, that are lifted by the wind but return to the surface because of their weightby sedimentation (saltation process from Claiborn *et al.*, 1998).

### 3.3 Meteorology

All models except CMAQ and RCG shared the same meteorological dataset at 0.2° resolution based on ECMWF IFS (Integrated Forecast System) calculations.

Because of its importance for applications (*e.g.* in air pollution modelling), the boundary layer height, as diagnosed in the IFS-ECMWF model, was made availablewas diagnosed in ECMWF was made available. The parameterization of the mixed layer (and entrainment) uses a boundary layer height from an entraining parcel model. But, in order to get a continuous field, also in neutral and stable situations the bulk Richardson method proposed by Troen and Mahrt (1986) is used as a diagnostic, independently of the turbulence parameterization. Boundary layer height is defined as the level where the bulk Richardson number, based on the difference between quantities of energy at that level and the lowest model level, reaches the critical value $Ri_{cr} = 0.25$.

For RCG, a different meteorological data set was used. The 3D-data for wind, temperature, humidity and density were produced employing a diagnostic meteorological analysis system developed at Freie Universität (Berlin, Germany) and based on an optimum interpolation procedure on isentropic surfaces. The system takes into account all available observed synoptic surface and upper air data as well as topographical and land use information (Reimer and Scherer, 1992). Rain data,

cloud data and boundary layer heights were retrieved from the IFS data set. Boundary layer parameters as friction velocity and Monin-Obukhov- ~~lengh~~length~~th~~ were calculated on-the-fly by applying standard boundary layer theory.

The CMAQ model used meteorological variables calculated with the COSMO model in CLimate Mode (COSMO-CLM) version 4.8 clm 11. The COSMO model is the non-hydrostatic operational weather prediction model applied and further developed by the national weather services joined in the COnsortium for SMall scale MOdeling (COSMO) described in Bettems *et al.*~~,~~ (2015).

### 3.4 Boundary conditions

In this study, the MACC reanalysis ~~were~~ was used as input data for the boundary conditions (Inness *et al.*, 2013; Benedetti *et al.*, 2009). The MACC II project (Modelling Atmospheric Composition and Climate) ~~is establishing~~established the core global and regional atmospheric environmental service delivered as a component of the COPERNICUS initiative (http://copernicus.eu/). The reanalysis production stream provides analyses and 1-day forecasts of global fields of $O_3$, CO, $NO_2$, $SO_2$, HCHO, $CO_2$, $CH_4$, and aerosols. Other reactive gases are available from the coupled chemistry transport model. The reanalysis covers the period 2003 – 2011 with a one~~-~~month spin-up. It runs at approximately 78 km by 78 km horizontal resolution over 60 levels. The coupled chemistry transport model has the same 60 vertical levels and a horizontal resolution of 1.125 ~~degrees~~ ° x 1.125 ~~degrees~~°. For aerosols only elemental carbon, organic carbon, dust and sulphate were used.

Stratospheric ozone fields from the MACC reanalysis agree with ozone sondes and ACE-FTS ~~data~~ (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) data within ±10% in most seasons and regions. In the troposphere, the reanalysis shows biases of −5% to +10% with respect to ozone sondes and aircraft data in the extratropics, while larger negative biases are shown in the tropics. Area-averaged total column ozone agrees with ozone fields from a multi-sensor reanalysis data set within a few percent. For aerosols, the observed Aerosol Optical Depth (AOD) is assimilated in the model with a feedback on individual PM species (sea salts, dust, elemental carbon, organic carbon and sulphate). When available, the MACC reanalysis is compared with observations, the model acronym in the supporting material is MACCA.

## 4 Observation dataset and statistics

### 4.1 Air pollutant concentrations

The evaluation was carried out ~~on~~ with the available EMEP standard monitoring (Tørseth *et al.*, 2012) and intensive period observations for 2006, 2007, 2008 and 2009 (Aas *et al.*, 2012) on hourly and daily bases (see supplementary material S8 for the description of background sites). Elevated sites above 1500 m in altitude have been excluded from the analysis. The

measurements were downloaded from the EBAS database. (http://ebas.nilu.no/). Additional AirBase data (Mol and de Leeuw, 2005) were used to evaluate the impact of meteorology on air pollutant concentrations in section 7.2.

It is important to note that daily measurements for a day, N, is the averaged value between day N HH:00 and day N+1 HH:00, with HH usually varying in the range [00, 09] in GMT. For most of the species, measurements on daily and hourly bases are not necessarily performed for the same set of stations. Deposition and the PM composition are also available; the dataset will be detailed in the companion papers.

### 4.2 Meteorology

**Temperature and wind speed**

The temperature, wind speed and precipitation measurements come from 2016 synoptic stations in Europe reported by the European meteorological centres. The data are provided on an hourly basis. The temperature is measured at 2 m and the wind speed at 10 m. Some meteorological data are also reported at some EMEP sites. At EMEP sites, daily accumulated measurements (*e.g.* precipitation) for a day N represent the integral between day N HH:00 to day N+1 HH:00, with HH usually varying in the range [00, 09] in GMT.

**Planetary Boundary Layer (PBL) height**

The soundings data were extracted from the University of Wyoming database (http://weather.uwyo.edu/). For each site and for each day, two soundings are available at 00:00 and 12:00 GMT. The provided meteorological parameters are: pressure (hPa), the corresponding height above ground level (m), dew point temperature (°C), relative humidity (%), mixing ratio (g $kg^{-1}$), wind direction (degrees) and wind speed (expressed in knot and converted ~~in~~ to ~~m/s~~m $s^{-1}$ by applying the conversion factor 0.514), potential and virtual potential temperature (K). For the present study, data were extracted over 77 stations in Europe. The boundary layer height is estimated using the calculation of the Bulk Richardson number profile and searching for the altitude where the critical value of $Ri_{cr}=0.25$ is reached. The analysis was limited to the first 25 vertical points, roughly corresponding to an altitude of 5000m above ground level. Since the boundary layer height is a concept valid only for convective~~Being the boundary layer height a concept valid only for convective~~ periods, only the soundings of 12:00 GMT~~Z~~ were analyzed and used for the model~~s~~ evaluation.

In addition to the previous PBL data, hourly heights of the atmospheric boundary layer were calculated from LIDAR measurements in a background site near Paris (SIRTA in Palaiseau, France). A new objective method for the determination of the atmospheric boundary layer depths using routine LIDAR measurements ~~have~~ has been used (Pal *et al.*, 2013).

### 4.3 Error statistics for the evaluation of model performances

The errors statistics considered in this report are presented in Table 4~~Table 4~~. In supplementary material S0-S1 the ~~statistic~~ performances of all models for the four campaigns are reported. For a given pollutant or meteorological variable, model performance is computed for a common set of stations (over the same common geographic area). All maps of pollutant

concentrations and meteorological variables concerning individual models and ensemble are provided in supplementary material (S2-S6).

For the analysis of the "ensemble" a coefficient of variation *VAR* is defined as follows in Eq. 4:

$$VAR = \frac{1}{C_{ENS}} \sqrt{\frac{1}{M} \sum_m (C_m - C_{ENS})^2} \qquad \text{(Eq. 4)}$$

With $C_m$ the concentration of individual model *m* included in the *ensemble* (CHIMERE, LOTOS-EUROS, MINNI and EMEP), M is the number of models, and $C_{ENS}$ is the *ensemble* mean concentration.

## 5 Evaluation of the meteorology

Some general features for each campaign can be provided, they ~~are issued from~~follow~~are taken from~~ the NOAA (National Oceanic and Atmospheric Administration) global analysis (https://www.ncdc.noaa.gov/sotc/global/).

June 2006 temperatures were above average everywhere in Europe with low precipitation except in Balkan countries and Spain compared to the 1961-1990 base period.

January 2007 wa~~i~~s characterized by windy conditions in Europe with ~~cool~~ temperatures above the average everywhere ~~in Europe~~ except in Spain where temperatures were close to the average values. In the beginning of February temperatures ~~decreased~~ were particularly low in Scandinavia. Precipitations were low over the Mediterranean basin but above the climate average, compared to the 1961-1990 period~~,~~ in the rest of Europe~~Precipitation were low over the Mediterranean basin but above the climatic average compare to 1961-1990 base period in the rest of Europe~~.

In September- October 2008, no ~~strong~~ clear general characteristics were recorded; this transition period was characterized by slight negative temperature anomalies ~~in~~ over the western part of Europe, mainly France, United Kingdom and north of Spain.

After some cold spells in the end of February, March 2009 turned ~~cooler~~ milder with on average warmer temperatures compare to the 1961-1990 base period. Precipitation ~~anomalies were negative~~was below average in the west part of Europe and ~~positive~~ above average in the central and east part of Europe.

### 2-m t~~T~~emperature

As summarized in supplementary material S0, the models using ECMWF data show comparable high temporal correlation coefficients based on hourly values over the whole domain ($0.88 < R < 0.94$), with highest correlations values in northern Germany and France when looking on a daily basis. Correlations are lower ~~whatever the~~for all models over north of Italy and Austria. On average for the considered period, the bias is negative for all models in the range [-0.3 K, -0.7 K] for CAMx~~CAMx~~, CHIMERE, EMEP and LOTOS-EUROS. The negative bias for this group of models is more important for the two wintertime campaigns, however in Switzerland and Austria this bias exceeds -2K ~~whatever the~~for all campaigns. Since this group of models shares the same meteorology, the error statistics are very similar; the discrepancies are due to the different interpolation methods ~~from~~ used to regrid the 3D and 2D ECMWF ~~data~~ variables to the final CTM grid.

RCG displays a very low absolute bias close to zero for the 2009 campaign, and CMAQ displays the lowest negative bias up to -2K for the 2009 campaign. CMAQ has lower correlation coefficient particularly in Germany and Poland for the 2008 and 2009 campaigns.

As displayed in Fig. 1~~Fig. 2~~, the negative bias is driven by afternoon temperatures that are underestimated by all models, this statement is valid for all campaigns. The night-time temperatures are more in line with the observations. The RCG diurnal cycle is rather different with a flatter profile but for the other models using ECMWF or COSMO data, the general pattern is well captured.

**10-m w~~W~~ind speed**

All the models using ECMWF data overestimate the wind speed from +0.1 to +0.9 m $s^{-1}$, while CMAQ, driven by COSMO, showed on average the lowest absolute bias. The biases are the highest for the two winter (2009) and fall (2008) campaigns, while for the summer campaign (2006) the biases are lower. It is worth noting that the 2007 campaign was the most windy period, showing a mean observed wind speed of 4.77 (m/s).

The b~~B~~ias is generally higher in eastern and northern Europe than in western and Mediterranean areas. In Europe, the spatial pattern of biases shows high positive bias in several coastal areas and negative bias in mountainous areas (Alps). This clearly points out a problem in some regions for the calculation of some emissions directly relying on IFS U10 fields. According to Ingleby *et al.* (2013) ECMWF 10 m wind speeds are slightly overestimated especially at night. In the IFS only 10m winds from ocean going ships are used in the data assimilation due to problems with station ~~representativity~~representativeness for inland sites~~In the IFS only 10m winds are used from ships over the oceans for data assimilation (problem of station representativeness for inland stations)~~. Moreover, errors on wind speed measurements are ~~stronger~~ higher for low winds. For the lowest winds the comparison of the predicted diurnal cycle with observations shows a larger positive bias at night than during the afternoon (Fig. 1), this behaviour could lead to an overestimation of the advection process in the chemistry transport models~~For the lowest winds generally observed during nightime the comparison of the predicted diurnal cycle with observations show a largest positive bias at night than during the afternoon (), this behaviour could lead to an overestimation of the advection process~~.

Time correlations are better for models using ECMWF data but all models exhibit low correlations over the Alpine~~s~~ region~~s~~ (North of Italy, South East of France, Switzerland and Austria). The RCG model shows higher correlation coefficients over northern Europe (Finland and Sweden) for the 2009 campaign.

**Planetary boundary layer (PBL) and mixing**

As explained in section 4.2, the observed PBL height was calculated at 12:00 because of methodology hypotheses, except at the SIRTA site where hourly measurements are available for 2008 and 2009. All models have a negative bias, the lowest RMSE are ~~displayed~~ shown for CAMx~~CAMx~~ CAMx and CHIMERE which use the ECMWF PBL, the biases are in the range -237 m and -100 m for these two models. It is worth noting that CAMx~~CAMx~~ CAMx and CHIMERE exhibit~~s~~ exactly

the same performance, while LOTOS-EUROS and EMEP that ~~should~~ adopted IFS PBL too, show partially different performances., ~~suggesting that the latter models partially recomputed boundary layer height~~Some differences are attributed to different interpolation schemes and the use of minimum PBL values during night-times as for EMEP. The largest underestimation of the PBL height is usually found for MINNI particularly for the 2006 campaign (up to -616 m) and EMEP (up to -451 m) and the correlation coefficients for these models are lower compared to the others. CMAQ has the lowest bias for most of campaigns. Models using IFS PBL data showed the best performance for temporal correlation (see supplementary material S0)~~The temporal correlations displayed in supplementary material S0 are the best for models using the IFS PBL~~, the main discrepancies are observed for the 2006 campaign with several sites in Europe with negative correlations. The largest negative biases are observed in the south of the domain, in these regions CMAQ performs better. In some regions over the Mediterranean basin, particularly in coastal areas, the MINNI's PBL is sometimes strongly biased up to -1000m. The obtained results suggest that either the Carlson algorithm or the micro-meteorological parameterization implemented by MINNI tends to underestimate the intensity of convec~~n~~tion.

The spatial representation of the PBL for the 2009 campaign shows higher differences between the models mainly over the ocean and seas where the coefficient of variation reaches 40% in some areas (Fig. 2~~Fig. 3~~). While LOTOS-EUROS, CHIMERE, RCG and CAMx~~CAMx~~ CAMx use the PBL from ~~ECMWF PBL~~ IFS with some differences on spatial and time interpolations, the other models use their own parameterizations discussed in section 2.2. The diurnal cycles displayed in Fig. 2~~Fig. 3~~ show that MINNI simulates a higher PBL at night and a lower PBL during daytime compared to ECMWF.~~,~~ ~~t~~The difference in ~~s of~~ the afternoon PBL is quite important over countries influenced by the ocean like the Great Britain. CMAQ and EMEP simulate over France and Great Britain the highest PBL at night. The hourly times series at the SIRTA site confirm the underestimation of the ECMWF PBL but at this station, the negative bias of MINNI is of the same order of magnitude as those of the other models~~the negative bias of MINNI has the same order of magnitude as the other models~~. The correlations based on hourly values are ~~still~~ somewhat lower for CMAQ, EMEP, MINNI (below 0.50) compared to the models using ECMWF data.

The differences ~~of~~ in treatment~~s~~ of ~~the~~ advection and mixing as reported in section 2.2 lead to differences in the reconstruction of pollutant dispersion~~of the dispersion~~. Fig. 3~~Fig. 4~~ shows the mean coefficient of variation of CO concentrations predicted by the models sharing the same raw meteorology (IFS) for the 2006 campaign. This pollutant can be considered as a tracer with low influences of deposition and chemistry processes, most of the differences on concentrations are related to transport and mixing. The figure clearly shows that mixing ~~on~~ in emission~~s~~ areas, such as big cities, produces the highest differences exceeding 20% of variations. ~~Besides of in urban areas, the~~The next highest coefficients of variation are observed over the seas and ocean ~~that~~ which are related to the differences of PBL predicted by the models (Fig. 2~~Fig. 3~~), elsewhere this coefficient remains below 10%.

## 6 Overall model performance evaluation on criteria pollutants

### 6.1 Ozone

The models' performances (supplementary material S1) are very different from campaign to campaign. Most of the models overestimate ozone concentrations in 2006, 2007 and 2008 (Fig. 4Fig. 5). Only the 2009 campaign show a systematic underestimation of observed ozone concentrations from -5 to -16 µg $m^{-3}$. The large positive bias in 2007 and negative in 2009 are largely explained by the boundary conditions that are biased respectively of +8 and -20 µg $m^{-3}$ (Supplementary material S1). For the positive bias in 2007 the boundary conditions cannot be the sole reason, chemical processes play an important role. Correlations are similar for all models in the range 0.5-0.6, only CMAQ has lower correlations on average. For the summertime campaign 2006 CHIMERE and CMAQ display the lowest correlation for daily averaged concentrations but CHIMERE has the lowest bias with EMEP. The low correlation for CMAQ and CHIMERE is due to the difficulties to reproduce both spatial patterns and day to day variations. For this campaign most models underestimate concentrations in the mountainous regions in Spain and over the Alps (Fig. 5Fig. 6). The models tend to over predict ozone concentrations on background stations influenced by large urban areas like GR01 station in Greece and IT01 close to Romae. All models simulate high ozone concentrations over the Mediterranean seaMediterranean Sea, most of them behaves satisfactorily in Malta and Cyprus stations confirming in agreement with the ozone concentrations pattern over the seas for the "ensemble" shown in Fig. 5Fig. 6 and particularly in Malta (Nolle *et al.*, 2002). The diurnal cycles in Fig. 6Fig. 7 reflect the overall performances depicted previously. All models fairly simulate the timing of the daily peak. For campaign 2007, except MINNI the models overshoot during nighttimenight-time and daytime. For campaign 2008, the very good shape of the LOTOS-EUROS diurnal cycle is remarkable. For the summertime campaign 2006, CHIMERE and EMEP provide on average the best diurnal cycles. Focussing on 2006 and 2008 campaigns, the two campaigns which are not biased by the boundary conditions, LOTOS-EUROS show the best performances regarding the bias. For these two campaigns, CAMx CAMx has a strong positive bias particularly at night. CAMx CAMx shares and exactly the same PBL height of CHIMERE use exactly the same PBL height of, but night-time performances of the two models are rather different. This result confirms that during stable conditions the pollutant concentration is influenced not only by the PBL height, but also by the overall reconstruction of vertical dispersion.

In Fig. 5Fig. 6, the right side is the gridded coefficient of variation that is a standardized measure of the dispersion of model results. It is defined as the ratio of the standard deviation to the mean of models. This coefficient is very low for the 2006 campaign, below 10%, the models have different responses along the ship tracks. The coefficients of variation are the highest for the 2007 campaign (supplementary material S2) associated with low performances of the "ensemble" (high normalized root mean square errors). Not only the bias is affected by global boundary conditions, but also this result indicates that biased ozone boundary conditions globally impair the normalized statistics confirming the non linearity of ozone chemistry. France, Spain and Norway show the lowest coefficient of variation indicating a more coherent behaviour among the models, but not necessarily corresponding to better model performance than other areas.

At Mace Head (IE31) located on the west part of the domain the time series of model results versus ozone observations show flat shape for the two winter campaigns with very low time correlations in 2009 (Fig. 7~~Fig. 8~~). The best correlation coefficients are observed for 2006 and 2008, the models are able to capture the peaks. At this station the negative bias mentioned in 2009 is roughly the same for LOTOS-EUROS, MINNI and RCG and comparable to the MACC analysis (-20 µg $m^{-3}$), the other models ~~CAMx~~ CAMx, EMEP, CHIMERE and CMAQ have a lower absolute bias (about -10 µg $m^{-3}$). This behaviour shows that concentrations close to boundary conditions are quickly modified certainly because the regional models restor~~e~~ing their own chemical equilibrium in relation with dynamical processes like deposition and vertical dispersion.

### 6.2 Nitrogen dioxide

For $NO_2$, all models perform similarly in terms of correlation with value in the range 0.6-0.7 (Fig. 4~~Fig. 5~~ and supplementary material S1). The spatial correlation is much higher in the range 0.7-0.9 for all models. Only CMAQ strongly overestimates the mean concentrations and ~~CAMx~~ CAMx underestimates the concentrations for all campaigns. Bessagnet et al. (2014) showed rather low concentrations of elemental carbon compared to other models, this inert species is particularly sensitive to vertical mixing and CAMx presents the highest minimum diffusion coefficient that is of major importance during stable conditions and partly explaining the lower $NO_2$ concentrations. For CAMx, the enhanced mixing influences also $O_3$ concentrations that are higher than other models~~This underestimation of $NO_2$ concentrations is certainly related to rather high ozone concentrations~~.

The spatial pattern~~s~~ of the "ensemble" shown for 2009 (Fig. 8~~Fig. 9~~) display~~s~~ high concentrations over the Benelux, North Italy, the biggest cities and over the shipping tracks. The bias of the "ensemble" is rather good except for one station in Serbia (RS05) with high observed values, probably due ~~relevant~~ to local sources. The gridded coefficients of variation provided in Fig. 8~~Fig. 9~~ show that most of differences between models are observed over remote areas f~~a~~or from emission regions even if errors are expected to occur more frequently for low values. As shown for a ~~non reactive~~less reactive species like CO, the differences of mixing in models ~~of~~ over emission areas ~~close to emissions is responsible~~ lead to large differences in modelled concentrations~~for model output differences~~, this effect can be clearly seen over the East Mediterranean for maritime emissions where the PBL is different from model to model. Over land~~s~~ the $NO_2$ chemistry and the different biogenic NO emission~~s~~ modules in the models ~~explain account for~~are believed to explain a large part of the differences on $NO_2$ concentrations far from urban areas. As shown in Fig. 8~~Fig. 9~~, the root mean square errors of the models are the highest for the stations close to the emission areas. The diurnal cycles in Fig. 9~~Fig. 10~~ show a general underestimation during the afternoon. It should be pointed out that the observed $NO_2$ concentrations can be slightly overestimated. ~~-~~For some types of analyzers, $NO_2$ is catalytically converted to NO on a heated molybdenum surface and subsequently measured by chemiluminescence after reaction with ozone. The drawback of this technique is that other oxidized nitrogen compounds such as peroxyacetyl nitrate and nitric acid are also partly converted to NO (Steinbacher et al.,

2007)~~because of sampling artefact (evaporation of nitric acid)~~. In the observations, the presence of two peaks on $NO_2$ concentrations is related to the traffic emissions peaks occurring in the morning and the evening. The timing of the peak occurrences is also modulated by the meteorology, for the 2006 and 2008 campaigns performed with identical summer time shift we clearly see a time shift of +1 and -1 hour respectively for the morning and evening peaks corresponding to a later rise and earlier fall of the PBL. Thus, as expected, the narrowest time lag between the two peaks is observed for the 2007 campaign. Most of the models predict the first peak too early, particularly CHIMERE and CMAQ for the 2006 campaign, and the second peak generally occurs too late.

CMAQ shows the strongest night-time bias, that ~~is the cause of~~contributes to explain the overall overestimation shown by the model in all campaigns. CMAQ was driven by a different meteorology that was characterized by very good performance with respect to both wind speed and PBL height mean bias. Conversely IFS-driven models overestimated night-time wind speed. As night-time vertical mixing is mainly driven by mechanical forces, the ~~obtained~~ model results ~~suggest~~ that models tend to underestimate mixing during stable conditions and, as a consequence, that IFS-driven models show better results ~~for the wrong reason~~suggesting compensation processes. ~~Differently, differences in diurnal temperature between CMAQ and other models seem less relevant with respect to pollutant concentration.~~

### 6.3 Sulphur dioxide

The correlations are rather low for all models in the range 0.2-0.4 for the 2006 campaign to 0.5-0.6 for the 2007 campaign (Fig. 4~~Fig. 5~~ and supplementary material S1 for all statistics). Two groups of models are identified ~~CAMx~~ CAMx, MINNI and RCG that largely overestimate the concentrations and CHIMERE, CMAQ, EMEP and LOTOS-EUROS which are closer to the observations on average with the best performances on the RMSE. The overestimation in the MINNI model could be partially explained by the low model PBL height~~The overestimation of the first group of models could be explained as follows for MINNI which has the lowest PBL and RCG having the lowest wind speed~~. For CAMx, the possible reasons such as the vertical distribution of $SO_2$ emissions near the harbours and coastal areas, insufficient conversion to sulphate and too low deposition were discussed in Ciarelli *et al.* (2016).~~For CAMx, it is not explainable at this stage, an in-deepdepth analysis with deposition and chemistry is necessary to understand this behaviour, this will be done in a companion paper~~. This ~~involves~~ leads to a positive bias of the "ensemble" as shown in Fig. 10~~Fig. 11~~ (supplementary material S4) particularly in Western Europe; the normalized RMSE is frequently above 100% in most part of Europe. The main hot spots are located in the Eastern Europe in addition with high concentrations along the shipping routes. The coefficient of variation is the lowest over emission areas but very high in remote areas like over the oceans far from shipping tracks and over mountain areas. This behaviour, very different from a primary species like CO, is a first indication of the very different way to simulate the $SO_2$ chemistry and deposition processes in the models.

The diurnal cycles presented in Fig. 11~~Fig. 12~~ show a peak at about 10:00 – 12:00. This peak is coherent with the hourly emission profiles of the industrial sector showing an emission peak at the same hours; however, most of models predict a larger decrease in the afternoon. Only CMAQ for the 2007 campaign captures satisfactorily the diurnal profile.

### 6.4 ~~$PM_{10}$~~PM10

~~Looking at~~Concerning the RMSE, on average the performances of the models are similar except CMAQ which has the highest values driven by low correlations and high negative biases particularly for the 2006 campaign~~for at least three campaigns~~ (Fig. 4~~Fig. 5~~). All models underestimate the concentrations generally in the range -3 to -10 µg $m^{-3}$. Except CMAQ, the correlations are in the range 0.4 – 0.6, but CHIMERE and EMEP reach 0.7 for the 2006 campaign. MINNI has the lowest absolute biases for the 2007, 2008 and 2009 campaigns. The "ensemble" provides a good picture of the ~~$PM_{10}$~~PM10 concentrations in Europe (Fig. 12~~Fig. 13~~ and supplementary material S5) except for two stations IT01 in Italy and CY02 in Cyprus with high recorded values. For CY02, high ~~$PM_{10}$~~PM10 concentrations are linked to high calcium concentrations (Bessagnet et al., 2014) due to dust events issued from North Africa. This dust event can be clearly observed for EMEP in Fig. 14~~Fig. 15~~. The spatial patterns show low concentrations below 5 µg $m^{-3}$ in remote Scandinavia and three hot spots in the Po valley, Benelux and South Poland. The coefficient of variation~~s~~ of model results is rather high over the seas and arid areas as well as over areas influenced by biogenic emissions as in Scandinavia,~~ over the seas and arid areas~~. This coefficient is generally the lowest over the Western Europe. The best RMSE of the "ensemble" are observed for the summer campaign 2006 with values below 50% of the observations data.

EMEP has higher concentrations over ~~the~~ North Africa because the model generates dust in this part of the domain and sea salt concentrations are generally higher ~~of~~ over the seas. EMEP and CHIMERE perform well for the spatial correlations (Table 5~~Table 5~~), EMEP captures better the high concentrations in the south of the domains whereas CHIMERE performs better over the Benelux (supplementary material S5). In 2008, RCG has particularly good spatial correlation compared to the other models. The missing sea salt emission for ~~CAMx~~ CAMx is clearly observed over the ocean with very low ~~$PM_{10}$~~PM10 concentrations impairing the spatial correlations.

As shown in supplementary material S5, most of models underestimate the highest PM10 concentrations observed in 2008 and 2009 by a factor of 2.~~for the highest concentrations observed in 2008 and 2009, most of models underestimate the $PM_{10}$ by a factor of 2~~. For the 10% highest ~~$PM_{10}$~~PM10 concentrations, MINNI has the lowest underestimations for these two campaigns whereas EMEP behaves rather well for the 2006 campaign regarding the bias and the correlation. As shown in Bessagnet *et al.* (2014) the large underestimation in 2009 ~~are~~ is ~~related to~~driven by the underestimation of organic~~s~~ species.

The observed diurnal cycles of ~~$PM_{10}$~~PM10 are very flat ~~whatever~~ for all ~~the~~ campaigns with a small peak in the evening (Fig. 13~~Fig. 14~~). The systematic underestimation of ~~$PM_{10}$~~PM10 can be clearly observed but the shape of cycle is not very well captured, the evening peak is not reproduced. The models simulate low concentrations in the afternoon mainly driven by the

elevation of the PBL. For the 2009 campaign, MINNI reproduces very well the diurnal cycle until 16:00. As shown in Fig. 14~~Fig. 15~~, dust concentrations are higher for MINNI in the ~~center~~centre of the domain. MINNI uses a parameterisation for wind blown dust very productive over any land cover types (Vautard *et al.*, 2005). ~~In comparison~~ EMEP mainly produces dust by traffic resuspension and a ~~few~~ little over arable land~~s~~. This higher production of dust by MINNI in Europe certainly improves the ~~PM~~ negative bias for PM usually observed in chemistry transport models, ~~and~~ particularly in the afternoon when the wind speed is ~~are~~ higher and the soil moisture content lower.

Most of the underestimation of PM10 by the ~~s of~~ models ~~is~~are is driven by too low day time $~~PM_{10}~~$PM10 concentrations. It is noteworthy that MINNI calculate the lowest PBL that could explain its relatively higher PM10 concentrations~~this specific behaviour~~. For the summer campaign 2006, the $~~PM_{10}~~$PM10 observations show an increase of concentrations in the afternoon while all other models tend to ~~all models~~ predict a decrease, indicating that all models are too sensitive to dynamical processes (meteorology) and not sufficiently to the chemical formation.

### 6.5 ~~$PM_{2.5}$~~PM2.5

Performances on ~~$PM_{2.5}$~~PM2.5 concentrations are rather different compared to ~~$PM_{10}$~~PM10 (Fig. 4~~Fig. 5~~). MINNI generally shows a slight positive bias while all models underestimate the averaged concentrations, with CMAQ ~~having~~ showing the highest negative bias. The performances of CHIMERE on the correlation are very good for all campaigns, its RMSE being the lowest for three campaigns. As for ~~$PM_{10}$~~PM10, the "ensemble" captures rather well the spatial patterns of ~~$PM_{2.5}$~~PM2.5. The concentrations in the south of Europe (Fig. 15~~Fig. 16~~ and supplementary material S6) are not specifically underestimated except in Cyprus where dust events also contribute to increase the ~~$PM_{2.5}$~~PM2.5 concentrations. ~~Whatever~~ For all ~~the~~ campaigns the coefficient of variation for ~~$PM_{2.5}$~~PM2.5 is the lowest in Spain but the RMSE of the "ensemble" is not particularly low in this region. The coefficient of variation is generally high over the north east part of the domain. For all campaigns the models simulate a hot spot over the north of Italy. As shown in the supplementary material S6, CMAQ captures the PM2.5 concentrations in Ispra (IT04) for 2007 and 2008 campaigns better than the other models.~~CMAQ better than the other models captures the $PM_{2.5}$PM2.5 concentrations in Ispra (IT04) for 2007 and 2008 campaigns.~~, t~~This station located at the border of the Po valley hot spot is usually underestimated by the models due to the very stablye ~~meteorology~~ stratified meteorological conditions in this region. The spatial correlations are usually better for ~~$PM_{2.5}$~~PM2.5 for all models except for the summer campaign (Table 5~~Table 5~~).

As for the ~~$PM_{10}$~~PM10 concentrations, the diurnal cycle of ~~$PM_{2.5}$~~PM2.5 is rather flat with very small morning and evening peaks (Fig. 16~~Fig. 17~~). The models have a different behaviour; they simulate a sharp decrease of concentrations in the afternoon consistent with ~~$PM_{10}$~~PM10 diurnal cycles. This confirms the lack of secondary production during daytime. The chemical schemes for the production of organic matter are still incomplete for one main reason. As suggested by Jathar *et al.* (2014) a large part of the "unspeciated" fraction of organic species react and produces secondary organic matter and gasoline vehicles could be an important contributor, as well as wood burning emissions according Denier van der Gon *et al.* (2015).

This unspeciated fraction is not included in our emission inventory explaining a part of the negative bias of models observed either in winter and summer campaigns particularly during the afternoon. This suggests that models with negative biases on ~~$PM_{2.5}$~~PM2.5 concentrations are consistent with the level of the completeness of our inventory ~~are coherent with the completeness of our inventory~~ and the state-of-the-art of knowledge on SOA modelling.

## 7 Impact of meteorology on pollutant concentrations

### 7.1 Impact of the PBL parameterization with MINNI results for the 2009 campaign

As shown in the previous section, MINNI underestimates the PBL heights calculated at 12:00 from measurements but it is in a better agreement with hourly data available at SIRTA (Fig. 2~~Fig. 3~~). In order to test the effect of PBL heights on air quality predictions, the MINNI model has been run using the PBL from IFS instead of its own parameterization for PBL heights. As Shown by Curci *et al.* (2015), processes in the PBL can greatly affect the ~~$PM_{2.5}$~~PM2.5 ground concentrations ~~at the ground~~, for instance temperature and relative humidity can favour the production of ammonium nitrate in the upper PBL.

Fig. 17~~Fig. 18~~ shows the average PBL heights and the average concentrations of $O_3$, $NO_2$ and ~~$PM_{10}$~~PM10 using MINNI's parameterizations (left graphs) and the percentage difference between the average concentrations calculated with PBL heights given by IFS ($PBL_{IFS}$) and by MINNI's parameterizations ($PBL_{MINNI}$) (right graphs) using the following formula: $(PBL_{IFS}-PBL_{MINNI})/PBL_{MINNI}$.

It can be seen that over the seas, on average, PBL heights calculated with MINNI's parameterizations ($PBL_{MINNI}$) are lower than PBL heights given by IFS ($PBL_{IFS}$) but over the lands $PBL_{MINNI}$ is higher than $PBL_{IFS}$ in coastal areas, North Africa, Scandinavian mountains and middle of Russian plains, and lower over the rest. Over the sea, $PBL_{IFS}$ are higher than $PBL_{MINNI}$ more than 50% while over the land the differences are between -30 and +30%.

Fig. 17~~Fig. 18~~ also shows that the $O_3$ concentrations increase in correspondence of the increase of PBL heights up to 10% and more, and decrease where the PBL heights decrease. This behaviour is explained by the fact that with a higher PBL more $O_3$ is entrained from high altitudes where $O_3$ concentrations are higher than at surface. Since the $NO_2$ sources are mainly at surface, the $NO_2$ concentrations generally decrease with the increase of PBL heights and increase with the decrease of PBL heights as a consequence of more or, respectively, less effective dilution. Over most of Europe, the $NO_2$ concentrations decrease up to 8% when $PBL_{IFS}$ heights are used. The ~~$PM_{10}$~~PM10 concentrations respond to PBL heights variation in the same way as $NO_2$. The use of $PBL_{IFS}$ heights produces a 4 % decrease of ~~$PM_{10}$~~PM10 concentrations in most parts of Europe but an increase of 6-8% in coastal areas and Russian plains.

In terms of statistics, the use of the PBL from IFS in MINNI slightly improves the correlations mainly driven by an improvement of time correlations. ~~$PM_{10}$~~PM10, ~~$PM_{2.5}$~~PM2.5 and $NO_2$ concentrations are decreased by less than 0.5 µg $m^{-3}$, improving all error statistics reported in Fig. 4~~Fig. 5~~ ~~for MINNI~~. An increase of 2.75 µg $m^{-3}$ is observed for $O_3$ concentrations. It is also worth to mention that the variations in pollutant concentrations are small (over the land below 10%

generally) in comparison to the variations of PBL height, therefore other factors such as emissions spatial distribution, meteorology (e.g. advection and vertical dispersion, especially in low-wind areas), gas phase chemistry, aerosol physics and chemistry have to be investigated for improving model performances.

These results clearly show the importance of having good estimates of PBL heights but they also demonstrates that more investigations are necessary in order to identify the best parameterization of PBL heights but also vertical diffusivities and vertical advection schemes which improves the simulated concentrations over the whole Europe.

### 7.2 Influence of meteorology on $NO_2$ concentrations with ~~CAMx~~ CAMx results

Pollutant concentrations are strongly influenced by the reconstruction of meteorological fields. In this section a comparison of model~~ling~~ performances in reproducing wind speed and $NO_2$ concentrations is presented and discussed. Furthermore, Planetary Boundary Layer (PBL) height data, collected at SIRTA site (Paris) have been used too. Being mainly related to emission processes, $NO_2$ has been selected as a ~~good~~ tracer of the influence of dispersion on pollutant concentrations. The analysis has been performed over the Paris area since the hourly variation of the PBL is available. Two other ~~smaller~~ limited areas, namely: the whole Germany (DE), the Po Valley (POV) ha~~s~~ves ~~been~~ selected to complement the analysis.

$NO_2$ observed data set has been set up from AirBase database (Mol and de Leeuw, 2005), selecting just background stations, having more than 75% valid data over the whole 2009. ~~Finally, as already mentioned, PBL heights derived at SIRTA site has been included too.~~ Modelled concentrations have been derived from the ~~CAMx~~ CAMx simulation results, while modelled meteorological fields have been derived from IFS.

In the case of the Paris area, the meteorological model showed a very good performance in reproducing the observed wind speed, whose temporal evolution clearly influences the corresponding temporal variability of $NO_2$ concentrations (Fig. 18~~Fig. 19~~). Also the PBL height is quite well reproduced by the model, though the model tends to underestimate the night-time minima and, conversely, to overestimate some diurnal peaks.

Within the Paris area $NO_2$ observations are quite well reproduced by ~~CAMx~~ CAMx, showing a low bias of the median value lower than 2 ppb, corresponding to less than 20% of the observed median concentration (Fig. 18~~Fig. 19~~). The availability of both wind speed and PBL height observations, allow the influence of both processes to be clearly detected. For example 3-4, 10 and 25 of March, the underestimation showed by ~~CAMx~~ CAMx seems well related to a corresponding overestimation of the PBL rather than the wind speed (Fig. 19~~Fig. 20~~). Conversely during night hours of March 5, ~~CAMx~~ CAMx results are more influenced by the wind speed.

The analysis has been completed comparing the diurnal cycle of both $NO_2$ and meteorological variables, reported in Fig. 20~~Fig. 21~~ and Fig. 21~~Fig. 22~~. At German sites $NO_2$ concentrations are slightly overestimated during night-time and underestimated during daytime. This behaviour does not seem strictly related to wind speed, particularly during night-time, thus being probably more related to vertical turbulence. At Po valley sites, $NO_2$ values are systematically underestimated, while wind speed is correctly reproduced, even partially underestimated during daytime hours. $NO_2$ modelled concentrations

show a clear low bias during night-time, probably related to an imprecise reconstruction of the strong stable conditions that characterize this area during the cold season. The ~~difficulty~~ model discrepancies ~~of model is~~ are enhanced during the morning hours, when the model is not able to capture the ~~strength~~ magnitude of the observed peak. The discrepancy is probably caused by a too rapid growth of the PBL during the first daytime hours. Late in the afternoon the $NO_2$ bias tends to decrease, probably thanks to a very quick collapse of PBL height after sunset.

At Paris sites, $NO_2$ modelled concentrations show a behaviour similar to the Po valley area. The availability of both wind speed and PBL height observations, allows most of the previous comments to be confirmed. Particularly it is worth noting that at SIRTA site, PBL height shows a too rapid increase during morning hours followed by a too strong decrease just after sunset. However, underestimation of NOx emissions cannot be ruled out as depicted in Vaughan *et al.* (2016) or Chen and Borken-Kleefeld (2016), these works highlight the potential underestimation of NOx traffic emissions.

## 8 Discussion ~~and Conclusions~~

~~One of the main outcomes of such a multi-seasonal intercomparison is that in most cases model performances are more influenced by the model setup than the season.~~ ~~For example,~~ The results from a mathematical model depend on three main factors: the model formulation (in terms of its assumptions, sub-models, numerical methods and their implementation in computer code); the model input data including boundary conditions; the skill of the model user particularly with respect to use of default values for certain inputs and parameters. When comparing results from a modelling exercise the performance, assessed from comparison between modelling results and data, is influenced by all three of these factors. It is therefore difficult to make judgments on the performance of a model without understanding the importance of configuration and use. In this model intercomparison exercise we have tried to achieve a greater focus on the effects of model formulation by standardising as far as possible the model input data and by running models for specific time periods having different meteorology and season (emissions and meteorology) to test responses over a range of input data. The comparison of results with observations has also been done in a standard way.

~~-~~CMAQ shows the ~~worst~~ largest RMSE between predicted and observed values for $NO_2$ over all campaigns, LOTOS-EUROS shows the lowest RMSE for $SO_2$ over all campaigns, ~~conversely CAMx~~ CAMx always exhibits the highest RMSE for $SO_2$ over all periods. This means that in several cases either the model formulation or the input setup influence the model performances more than specific features of the meteorological season.

~~Whatever the~~For all pollutants and ~~the~~ campaigns, there is not a strong correlation between the performances of the *ensemble* (through the RMSE of the difference between predictions of the ensemble and observed values) with the variability of models (through the coefficient of variation between individual model predictions and the ensemble predictions) ~~of models~~. This means if models are close ~~between~~ to each other (low coefficient of variation), the mean of models can be far or close to the observed values, there are no specific rule. However, for $SO_2$ and ~~$PM_{2.5}$~~PM2.5 a correlation of -0.2 to -0.3 is observed for three campaigns meaning that a large variability tends to improve the performance of the ensemble for these compounds. For

the other compounds, $O_3$, $NO_2$ and PM10, the correlation is close to zero. The coefficient of variation is the lowest for ozone (below 10%) particularly in the afternoon hours (see supplementary material S7) and for the summer period 2006, while for $SO_2$ this coefficient is the highest generally between 30 and 40%. For PM this coefficient is about 10 to 20%, over several countries, the coefficient of variation is higher in the afternoon highlighting the difference between chemical schemes for the aerosol chemistry more active during day times, conversely, the low coefficient of variability for $O_3$ confirms a coherence of ozone chemistry scheme between models.

~~Another general outcome stemming from the whole exercise is that model performances are more different from a pollutant to another than for the same pollutant within the different season. This confirms once again that on average model formulation and setup are more influencing than meteorological conditions on model performance. One of the few exceptions is shown by $O_3$ in 2009 where model results were characterized by RMSE values very similar to the other years, whereas bias was negative instead of positive as in the three previous years. But, as already pointed out, such a result was mainly driven by a relevant underestimation in the ozone boundary concentrations from MACC.~~

The intercomparison proved that CTMs are ~~affordable in~~able to reproduce~~ing~~ ozone concentrations, showing an average RMSE ~~value~~ of individual models corresponding to 30% of the mean observed concentration for daily values. Modelled daily cycles are generally more spread during night-time than daytime hours. This means that, though most models shared the same meteorology, including PBL height, they proved to be very sensitive to vertical dispersion and deposition parameterization, the two key processes governing $O_3$ concentration during night-time. During daytime ~~modelled~~modelled concentrations are more ~~overlapping~~ similar , ~~and showing~~they show a different ranking with respect to night hours. This means, as expected, that during daytime vertical mixing reconstruction is more similar among models and chemical schemes exhibit a different efficiency in ozone production. This behaviour is not detectable in 2007, that was a cold and windy period, hampering the development of photochemical processes.

$NO_2$ performances are less robust than for $O_3$. The RMSE represents about 70% of the observed mean concentration, but the value is even higher in case of CMAQ. Bias is negative for most models, except CMAQ, adopting a different meteorology and MINNI, characterized by lower PBL heights. CHIMERE biases are closer to 0 than other models sharing the same meteorology, such as ~~CAMx~~ CAMx.

~~The Normalized RMSE of the models "ensemble" is characterized by a relevant spatial variability, proving that local emission sources and meteorological conditions strongly influence $NO_2$ performance. Likewise~~ As for ozone, most of the discrepancies among models and with respect to observations take place during night-time, when the atmosphere is more stable. As most models share the same wind fields, the modelled spread in night-time concentrations can be related to vertical dispersion. Such spread for primary species and particularly for CO can be considered as a measure of the uncertainty related t~~d~~o vertical mixing and qualitatively correspond~~s~~ to 80-100% of the observed mean concentration. The height of the first level is also very important for the mixing and deposition processes, it ranges from 20 m for CAMx and CHIMERE to 90 m for EMEP. To be more representative of surface concentrations a correction is implemented for models having a coarse first surface layer (LOTOS-EUROS and EMEP). Daytime modelled concentrations are more similar among

models and generally underestimated, though the modelled PBL field at noon seemed lower than the observed one. As already mentioned such a systematic discrepancy could be related to a measurement artefact, but also to photochemistry that could give rise to an excess of nitric acid. More accurate observations of Nitric acid and Nitrate would be required.

$SO_2$ concentrations show~~s~~ the worst performance, with RMSE values corresponding to 130-160% of the observed mean concentrations. The h~~H~~ighest ~~errors~~ RMSE are shown by ~~CAMx~~ CAMx, MINNI and RCG~~., they were characterized by lower PBL heights (for MINNI) and wind speed (for RCG) than other models, for CAMx~~ CAMx ~~the high errors cannot be explained at this stage~~. It is worth noting that the modelled diurnal cycles show a weak morning peak, more typical of surface sources not observed in ~~measured data~~observations. Conversely, measured data present a diurnal peak, usually related to enhance downward mixing of aloft sources, where most of $SO_2$ is emitted. Discrepancies among models and with respect to observations can also ~~rely on~~be due to chemistry. For example in 2009, Bessagnet *et al.* (2014) report~~ed~~s for CHIMERE an underestimation~~e~~ of $SO_2$ concentrations on an hourly basis, while sulphate ~~is~~ was overestimated; conversely RCG, adopting a more simplified approach for sulphur chemistry than CHIMERE, overestimate~~d~~s $SO_2$, while underestimate~~s~~d ~~sulfates~~sulphates.

~~$PM_{10}$~~PM10 model~~s~~ performances are less homogenous within the four years than other pollutants. ~~Years~~The campaigns 2006 and 2007 that were characterized by a more dispersive atmosphere show a mean RMSE around 10 µg $m^{-3}$, representing 55-65% of the mean observed concentration. Differently, the RMSE rises up to 15 µg $m^{-3}$ for 2008 and 2009 campaigns~~,~~ representing more than 80% of the observed mean. The bias is ~~best~~ better reproduced by EMEP and MINNI, while ~~CAMx~~ CAMx and CMAQ show the strongest underestimation. The analysis of each PM compound for ~~season 2009~~the 2009 period (Bessagnet *et al.*, 2014) revealed that MINNI and EMEP were characterized by rather different scores, suggesting that their overall performance is influenced in a different way by both chemistry and meteorology. Particularly MINNI performance seems more driven by a reduced dispersion often giving rise to higher concentrations than other models, while EMEP seems more able to capture the evolution of the single PM compound as shown in Bessagnet et al. (2014). ~~CAMx~~ CAMx and CMAQ often show the strongest negative bias. As for ~~CAMx~~ CAMx this result is probably driven by the combined effect of meteorology (also $NO_2$ is underestimated by ~~CAMx~~ CAMx) as well as the absence of some key processes such as sea salt and dust resuspension and a PM coarse chemistry. Differently CMAQ model was characterized by very high $NO_2$ concentrations ~~putting in evidence~~stressing a less dispersive ~~atmosphere~~ capability than other models. As for CMAQ, the low ~~$PM_{10}$~~PM10 values ~~can~~ are probably related to deposition processes. Indeed, for 2009 episode (Bessagnet *et al.*, 2014) CMAQ proved to be more efficient than the other models for dry deposition of both $NO_X$ and $SO_X$ compounds.

The observed diurnal cycles of ~~$PM_{10}$~~PM10 are very flat ~~whatever the~~for all campaigns with a small peak in the evening. The ~~$PM_{10}$~~PM10 observations show an increase of concentrations in the afternoon while all models predict a decrease, indicating that all models are too sensitive to dynamical process (meteorology) and not sufficiently to the chemical formation. The analysis of individual compounds of PM will bring more detail~~sed~~, it will be investigated in a companion paper.

Model~~s~~ performance for ~~$PM_{2.5}$~~PM2.5 is on average slightly better than ~~$PM_{10}$~~PM10, both in terms of bias and correlation. ~~$PM_{2.5}$~~PM2.5 concentration is less affected by natural processes, which are more relevant for coarse PM, therefore the

obtained results suggest that modelling natural processes still present some relevant weaknesses (Bessagnet *et al.*, 2014). Modelled diurnal cycles show improved performance in terms of bias, but not with respect to the daily evolution. Firstly, this result confirms that there are processes mainly affecting the coarse fraction that are still missing in some state of art CTMs, highlighted by the different biases between ~~$PM_{10}$~~PM10 and ~~$PM_{2.5}$~~PM2.5. Secondly, the differences in the daily pattern, particularly evident in 2006 where photochemistry is at its maximum, confirm that dilution processes during daytime hours are too efficient with respect to chemical processes, thus preventing the increase of modelled concentrations during afternoon hours.

Even if the meteorology was prescribed in the exercise, some variables related to dispersion modelling such as the vertical diffusion and the PBL height are often diagnosed in the model pre-processing. This step involves important differences in the dispersion as was shown for a tracer species like CO. Although most models used the same PBL from IFS (CHIMERE, ~~CAMx~~ CAMx, LOTOS-EUROS, RCG), the variability of models PBL (including other PBL parameterisation as used in EMEP, CMAQ and MINNI) shows important differences of PBL calculations over the ocean and the Mediterranean sea. IFS wind speeds are overestimated with a bias reaching 1 m $s^{-1}$, which can have a dramatic effect at low wind speed conditions.

The comparison of the meteorological fields pointed out that the reconstruction of the meteorological variables is still affected by relevant uncertainties. Wind speed simulated by IFS and COSMO showed a systematic difference along the whole day, with IFS providing an average wind speed that in 2007 and 2009 was 12% higher than COSMO. PBL reconstruction showed an even higher variability with a spread among the models corresponding to 27-29% of the mean midday PBL value of each campaign.

~~Some additional analyses with respect to meteorology have been carried out. As a first step, a sensitivity analysis with respect to PBL height was performed with MINNI model. Over the sea, $PBL_{IFS}$ are higher than $PBL_{MINNI}$ more than 50% while over the land the differences are between -30 and +30%. As a consequence, $O_3$ concentrations increase in correspondence of the increase of PBL heights up to 10% and more, due to enhanced entrainment and reduced $NO_x$ titration. Over most of Europe, the $NO_2$ concentrations decrease up to 8% when $PBL_{IFS}$ heights are used and the $PM_{10}$PM10 concentration decreases by 4 % but also increases of 6-8% in coastal areas and Russian plains, where IFS PBL were lower than MINNI PBL. The PBL explain only a part of the overestimation of primary species but a complementary study has to be performed including the deposition processes.~~

A comparison of modelling performances in reproducing wind speed and $NO_2$ concentrations was performed too, also including some analysis of the influence of Planetary Boundary Layer (PBL) height estimation. The comparison of modelled concentrations against wind speed and PBL heights confirmed that meteorology strongly influences CTMs performance. Particularly the temporal evolution of wind speed is most responsible of model skilfulness in reproducing the daily variability of pollutant concentrations (*e.g.* the development of peak episodes), while the reconstruction of the PBL diurnal cycle seems more influencing in driving the corresponding pollutant diurnal cycle and hence the presence of systematic positive and negative bias detectable on daily basis.

## 9 Conclusions

One of the main outcomes of such a multi-seasonal intercomparison is that in most cases model performances are more influenced by the model setup than the season. Another general outcome stemming from the whole exercise is that model performances are more different from a pollutant to another than for the same pollutant within the different season. This confirms once again that on average and for the limited dataset used in this exercise, the model formulation (parameterization of chemical / physical processes, calculation of meteorological diagnosed variables) and set-up (number of vertical levels, value of key parameters, etc...) are more influencing than raw meteorological conditions on model performance. One of the few exceptions is shown by $O_3$ in 2009 where model results were characterized by RMSE values ~~st~~ every similar to the other years, whereas bias was negative instead of positive as in the three previous years. But, as already pointed out, such a result was mainly driven by a relevant underestimation in the ozone boundary concentrations from MACC.

Even if the meteorology was prescribed, some variables like the planetary boundary layer (PBL) height, the vertical diffusion coefficient are diagnosed in the model pre-processors and explain the spread of models results. For ozone, this study shows the importance of boundary conditions on model calculations and then on the regime of the gas and particle chemistry. The worst performances are observed for sulphur dioxide concentrations that are poorly captured by the models. The performances of models are rather good and similar for the nitrogen dioxide. On average, the models provide a rather good picture of the particulate matter concentrations over Europe even if the highest concentrations are underestimated. For the PM, the mean diurnal cycles show a general tendency to overestimate the effect of the PBL height rise while the afternoon chemistry (formation of secondary species) is certainly underestimated, PM observations show very flat diurnal profiles for all seasons. In general the day time PBL height is underestimated by all models, the largest variability of predicted PBL is observed over the ocean and seas. The temporal evolution of wind speed is most responsible of model skilfulness in reproducing the daily variability of pollutant concentrations (*e.g.* the development of peak episodes), while the reconstruction of the PBL diurnal cycle seems more influencing in driving the corresponding pollutant diurnal cycle and hence the presence of systematic positive and negative biases detectable on daily basis.

The study stresses the importance of emission sources particularly in wintertime, wood burning emissions are likely the most underestimated source, through the missing species called semi-volatile organic compounds. Road traffic emissions could also be underestimated, gasoline and diesel vehicles are both concerned, and more generally, all activity sectors involving combustion processes can be concerned. In this study, the importance of meteorological data is highlighted, the difficulties for meteorological models to simulate meteorological variables like wind speed and PBL height during stable conditions can lead to dramatic consequences on air quality modelling. Developments in air quality modelling have not only to focus on processes but also on emissions and meteorological input data.

To complement the analysis, companion papers will focus on depositions of sulphur/nitrogen compounds and on the behaviour of models for particulate matter species. This ensemble of analyses will help to prioritize the improvement of air quality models used in the frame of the CLRTAP.

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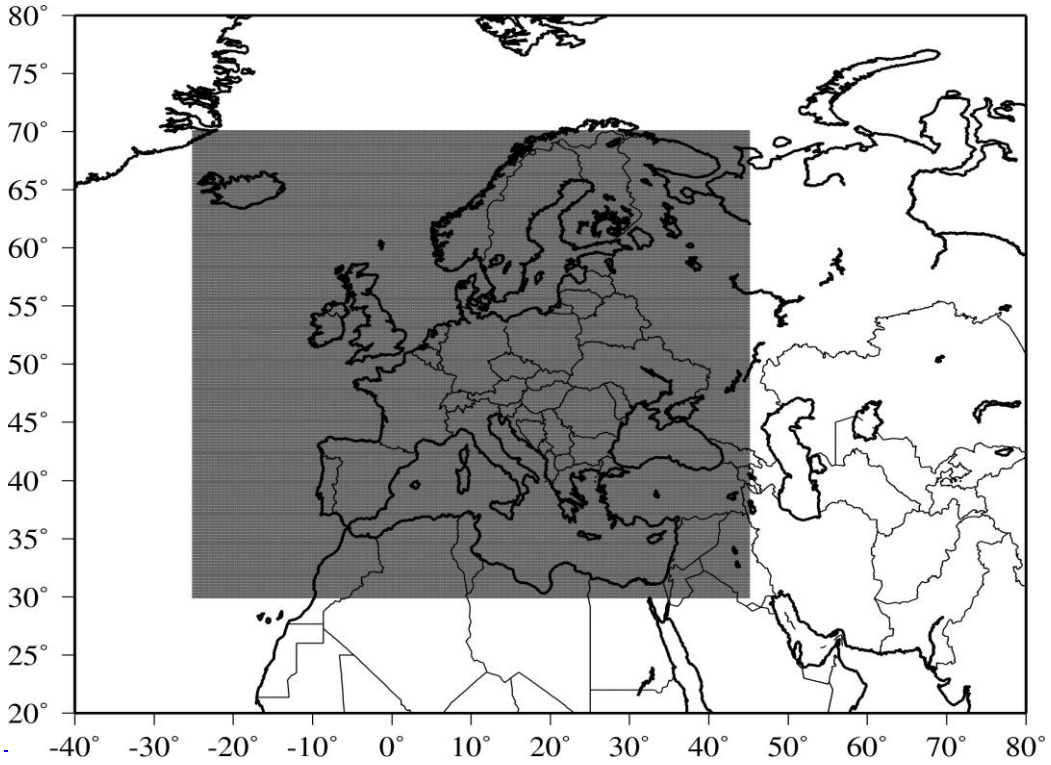
**List of figures**




**~~Fig. 1: The grey zone corresponds to the EURODELTA domain. All model simulations have been performed over this domain except CMAQ.~~**

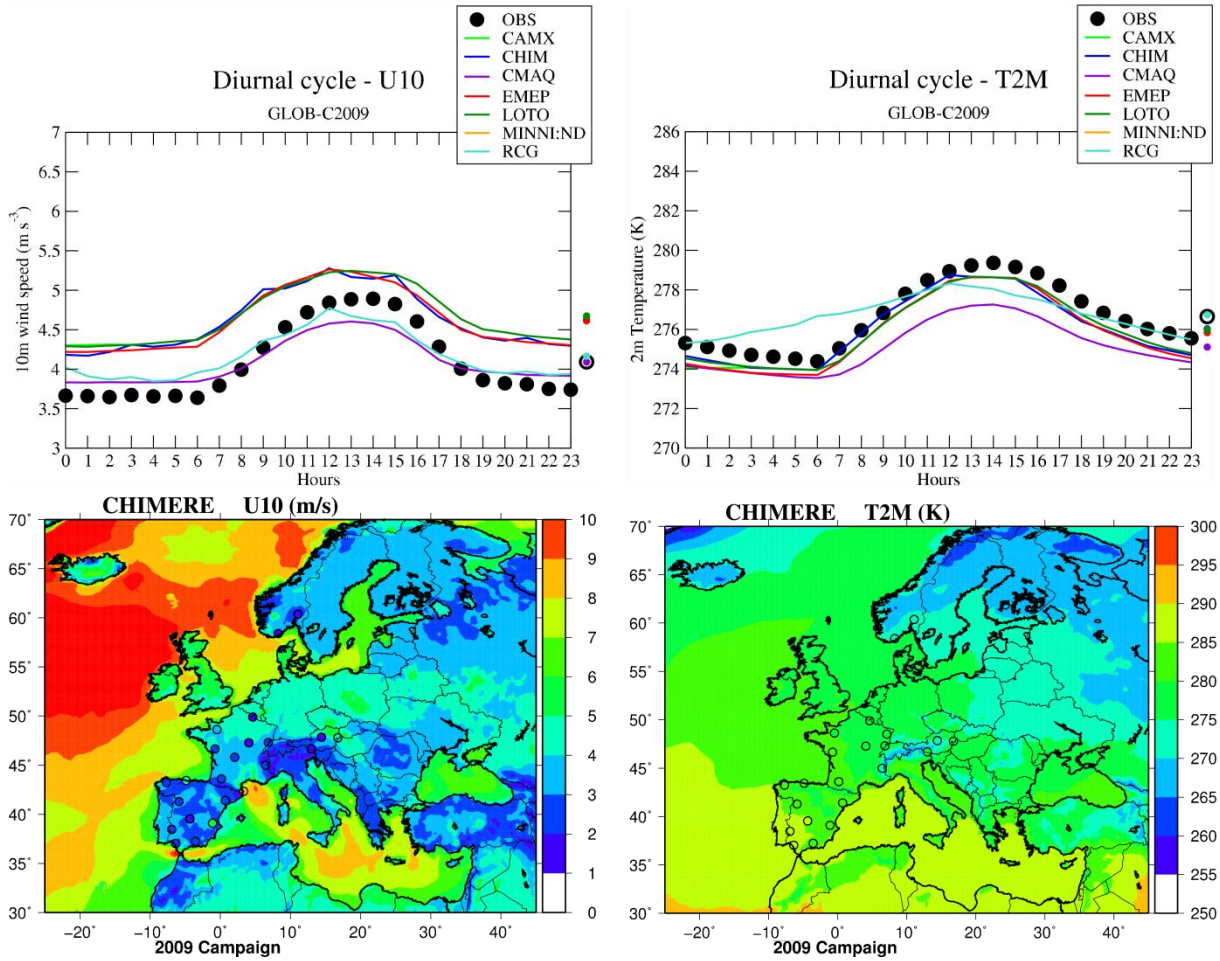



**Fig. 12: Comparisons of observed *versus* predicted meteorological variables (U10, T2M) for the 2009 campaign. *Top left panel*: mean diurnal cycle of the 10 m wind speed, *top right panel*: mean diurnal cycle of the 2 meter temperature, *bottom left panel*: mean 10 meters wind speed for CHIMERE, *bottom right panel*: mean 2 meters temperature for CHIMERE (Some observations at EMEP stations are provided with coloured circles over the maps). Red color is assigned for values exceeding the colour scale.**

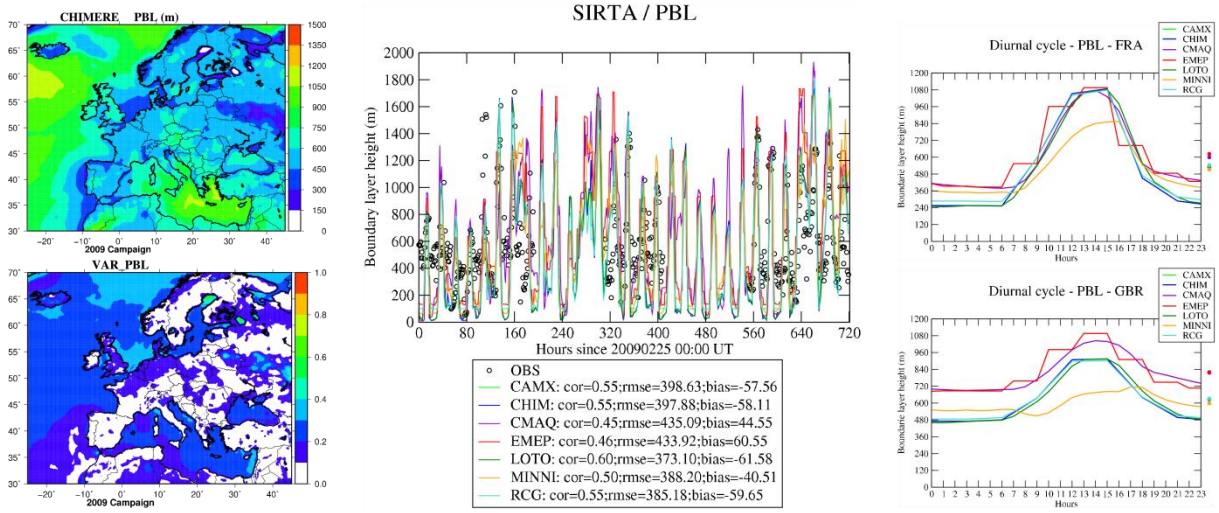



**Fig. 23: Spatial representations and time variations of the PBL height for the 2009 campaign. *Top left panel:* Mean height of the CHIMERE PBL height issued from ECMWF data. *Bottom left panel:* Mean coefficient of variation for the PBL height. *Central panel:* hourly variation of the PBL height at the SIRTA station. *Top right panel*: Average diurnal cycle of the PBL height predicted by the models in France. *Bottom right panel*: Average diurnal cycle of the PBL height predicted by the models in Great Britain.**

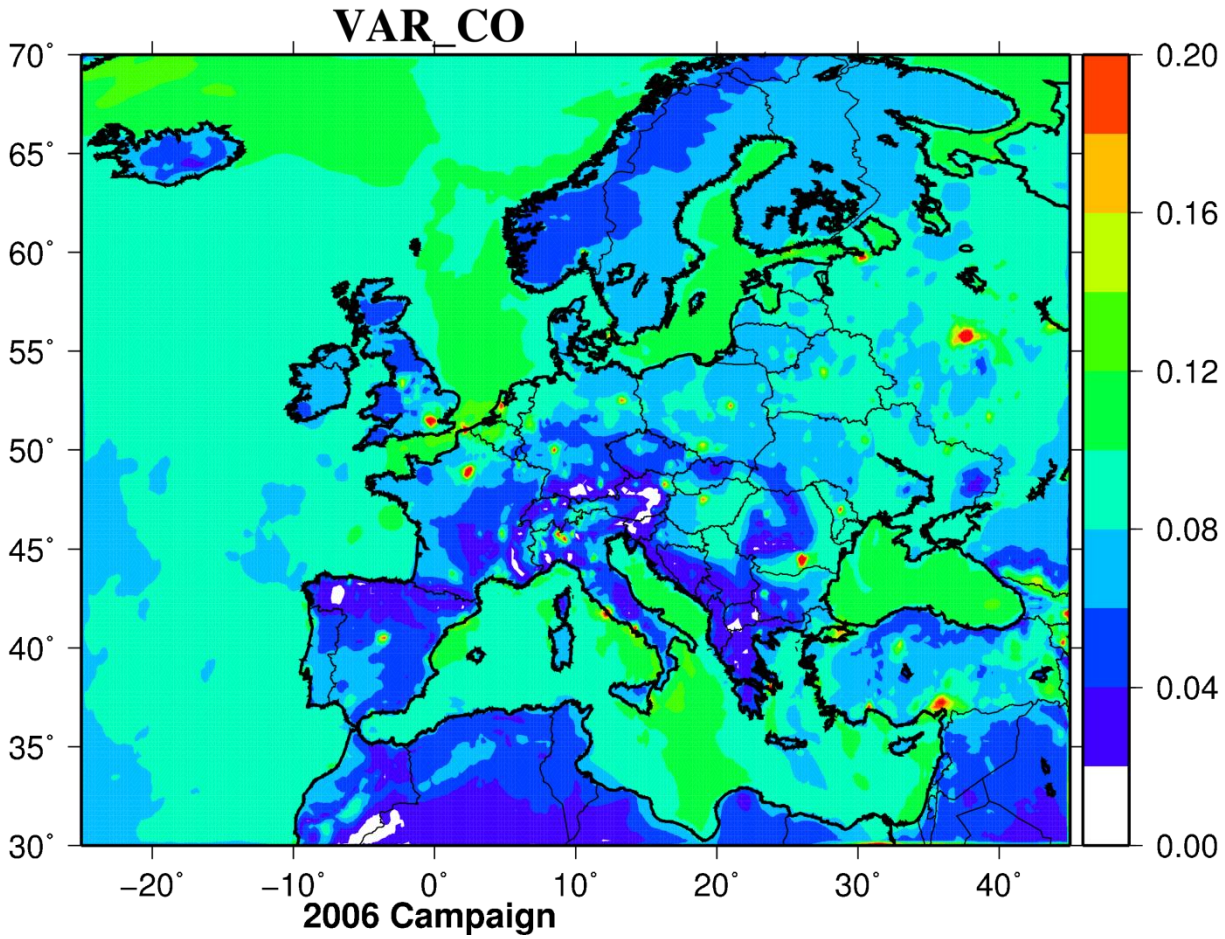



**Fig. 34: Mean coefficient of variation of the CO concentrations predicted by the models for the 2006 campaign (no unit). Red color is assigned for values exceeding the color scale.**

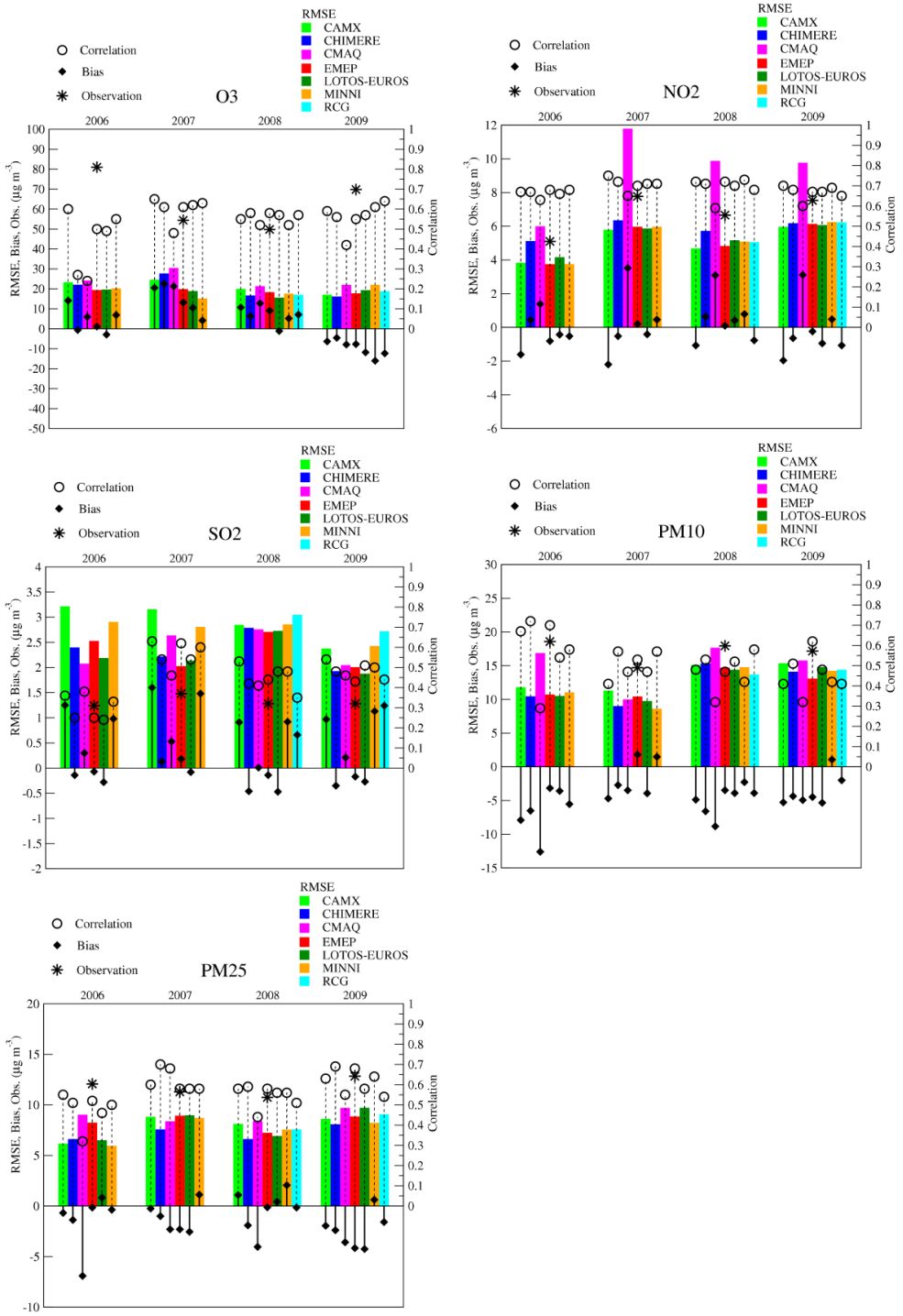



**Fig. ~~45~~: Overall performance of models for Ozone, Nitrogen dioxide, Sulphur dioxide, ~~$PM_{10}$~~PM10 and ~~$PM_{2.5}$~~PM2.5 daily mean concentrations for all campaigns.**

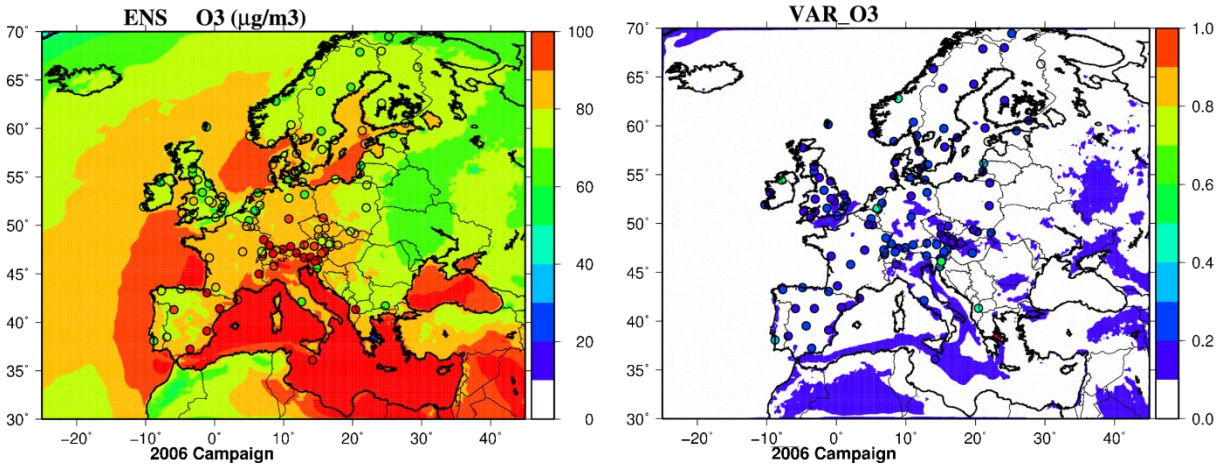



**Fig. 56: *Left column*: Mean ozone concentrations (µg $m^{-3}$) of the "ensemble" (ENS) for the 2006 campaign with corresponding observations (coloured dots). *Right column*: coefficient of variation of models (no unit) constituting the ensemble with corresponding normalized root mean square errors of the "ensemble" (coloured dots). Red color is assigned for values exceeding the color scale.**

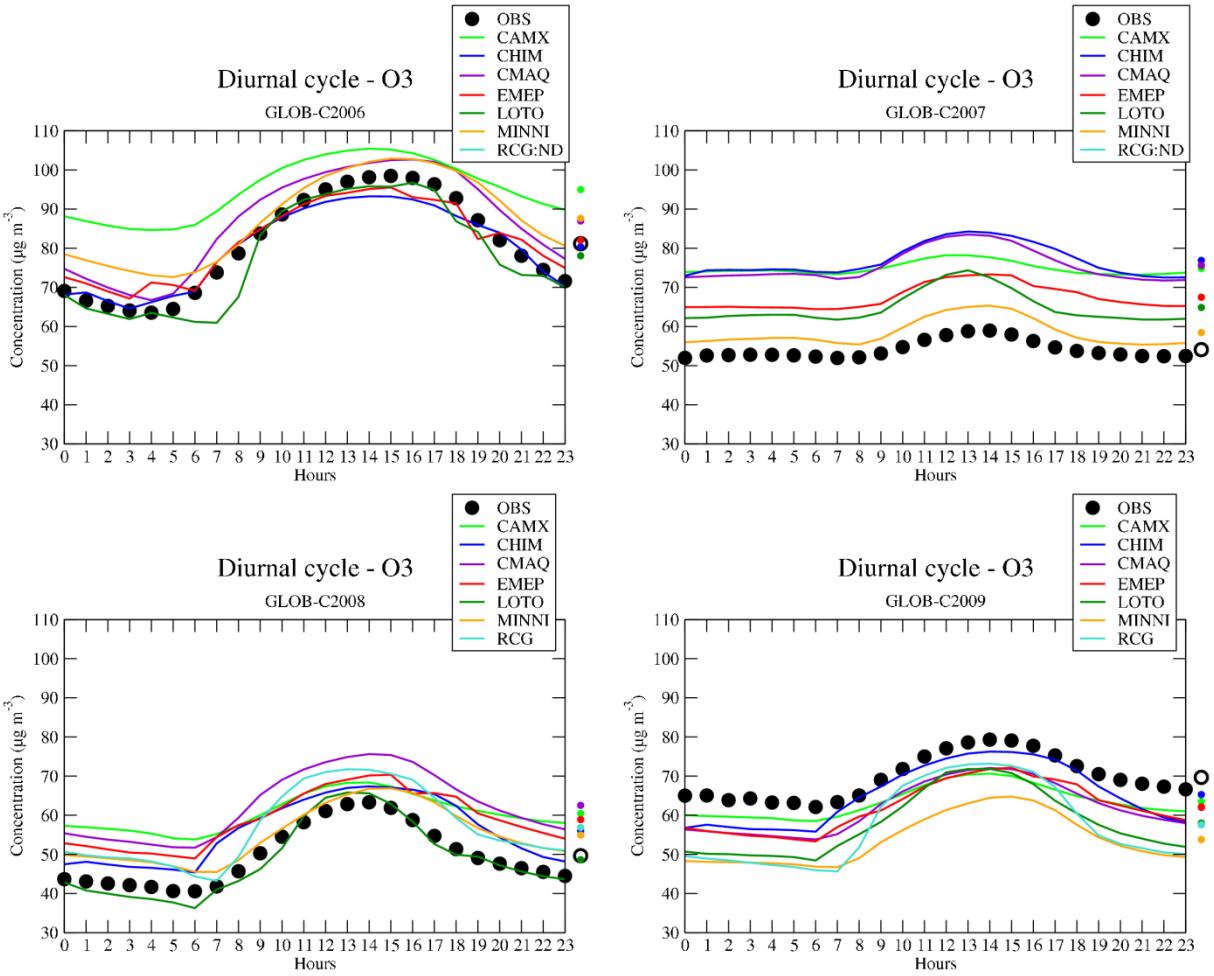



**Fig. 67: Mean ozone diurnal cycles for all campaigns simulated by the models compared with observations. Averaged concentrations are provided on the right side of the charts.**

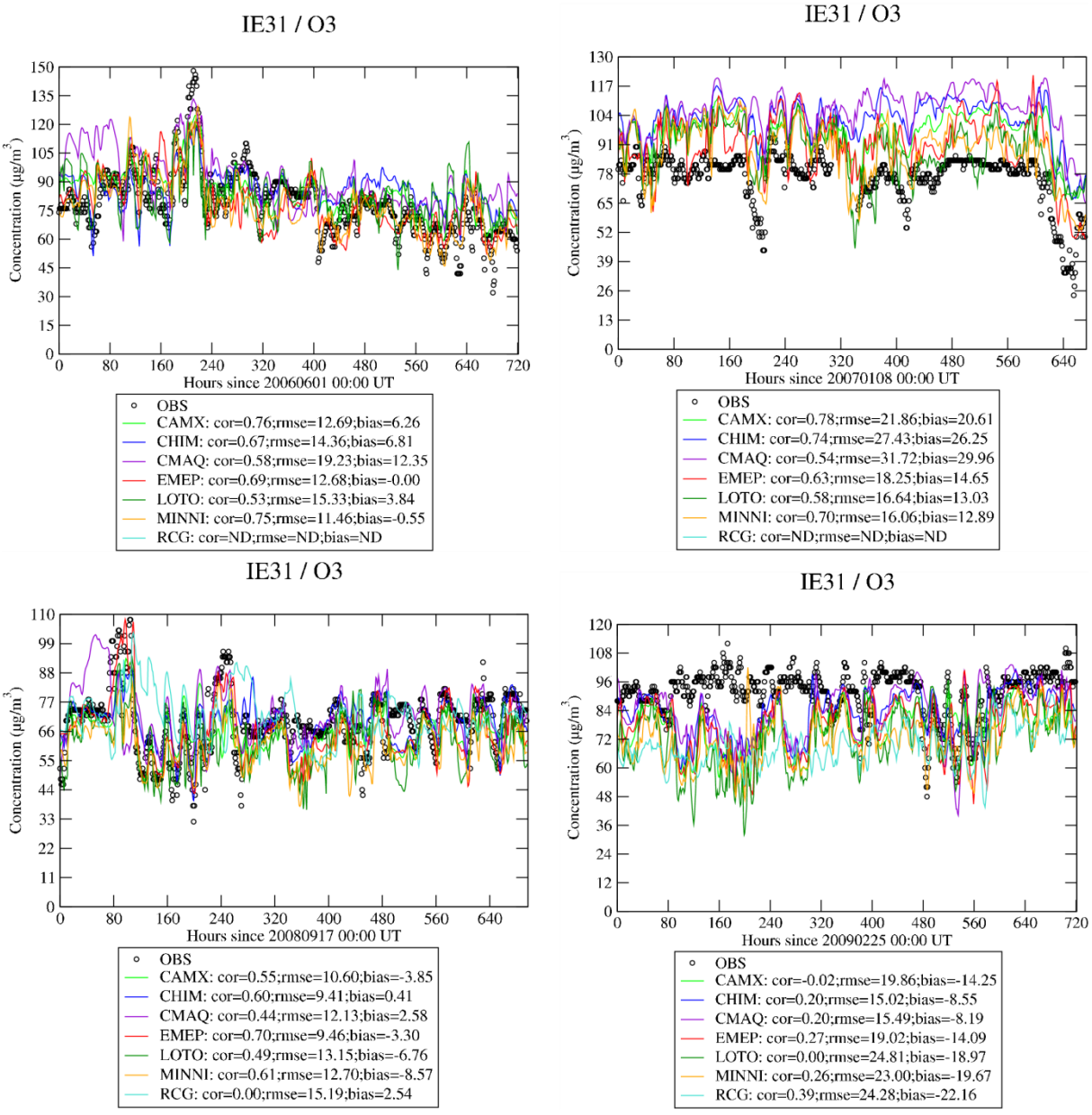



**Fig. 78: Timeseries of hourly concentrations at Mace Head for all models and campaigns**

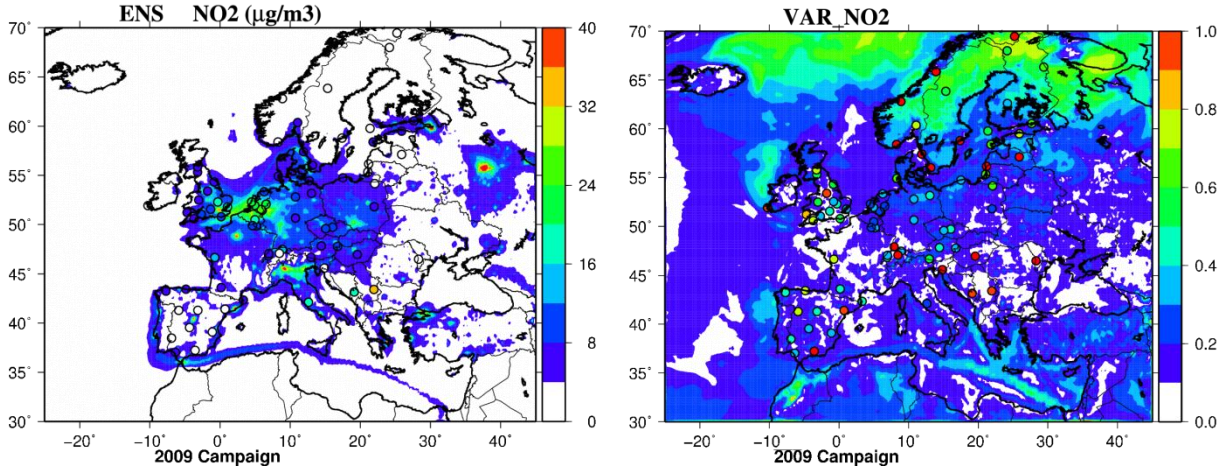



**Fig. 89: *Left column*: Mean nitrogen dioxide concentrations (µg $m^{-3}$) of the "ensemble" (ENS) for the 2009 campaign with corresponding observations (coloured dots). *Right column*: coefficient of variation of models (no unit) constituting the ensemble with corresponding normalized root mean square errors of the "ensemble" (coloured dots). Red color is assigned for values exceeding the color scale.**

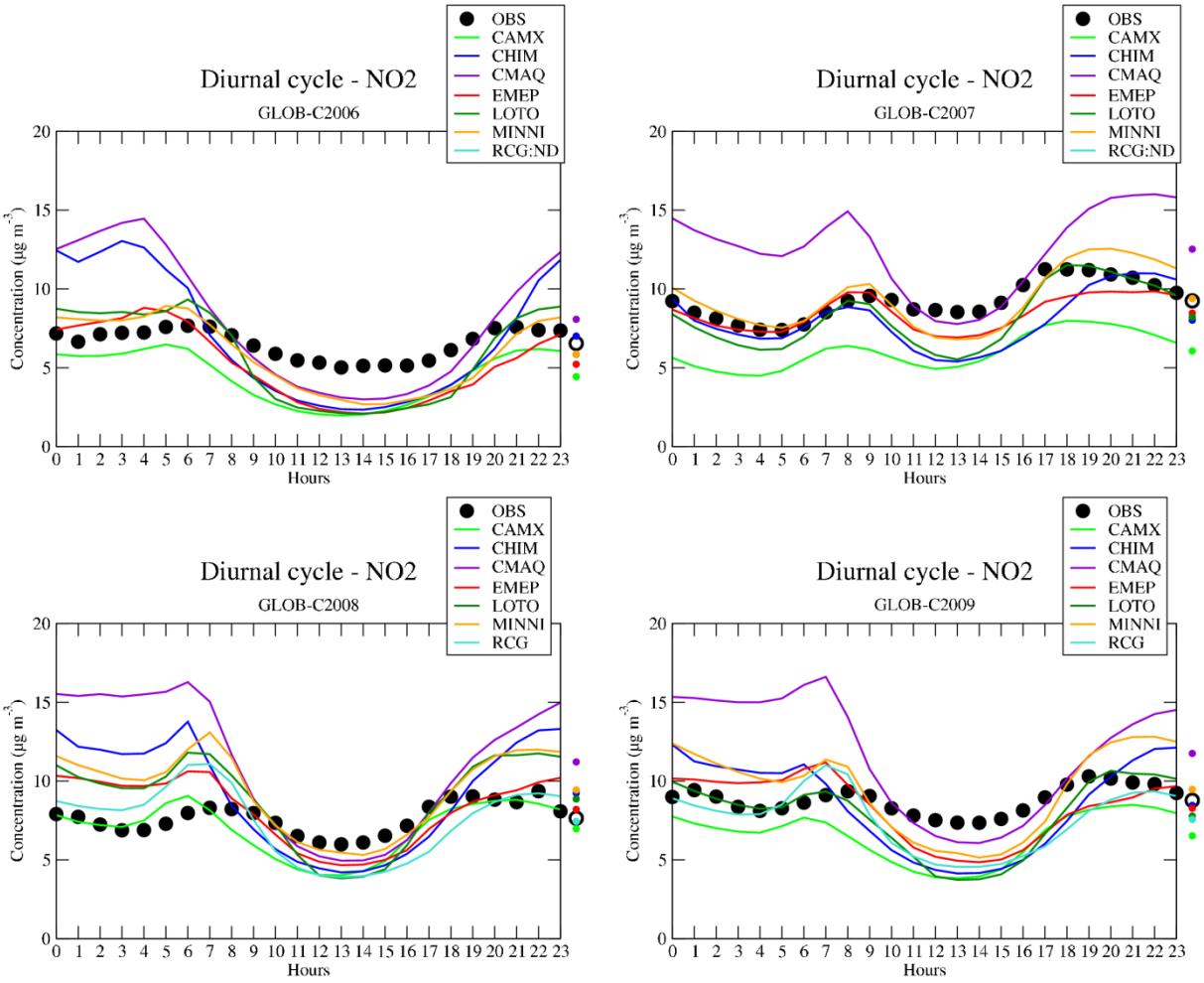



**Fig. 9~~10~~: Mean diurnal cycles of nitrogen dioxide for all campaigns simulated by the models compared with observations. Averaged concentrations are provided on the right side of the charts.**

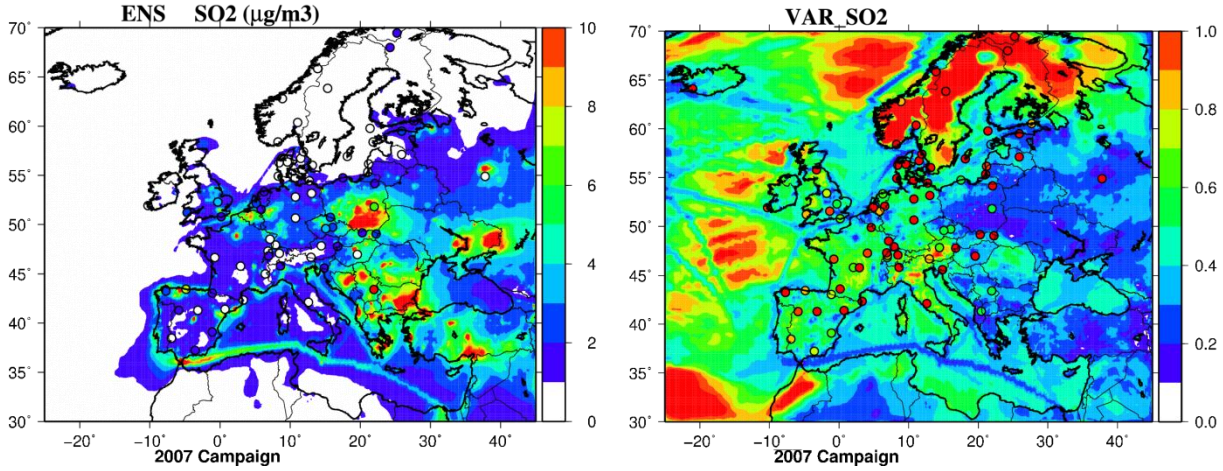



**Fig. 1011: *Left column*: Mean $SO_2$ concentrations (µg $m^{-3}$) of the "ensemble" (ENS) for the 2007 campaign with corresponding observations (coloured dots). *Right column*: coefficient of variation of models (no unit) constituting the ensemble with corresponding normalized root mean square errors of the "ensemble" (coloured dots). Red color is assigned for values exceeding the color scale.**

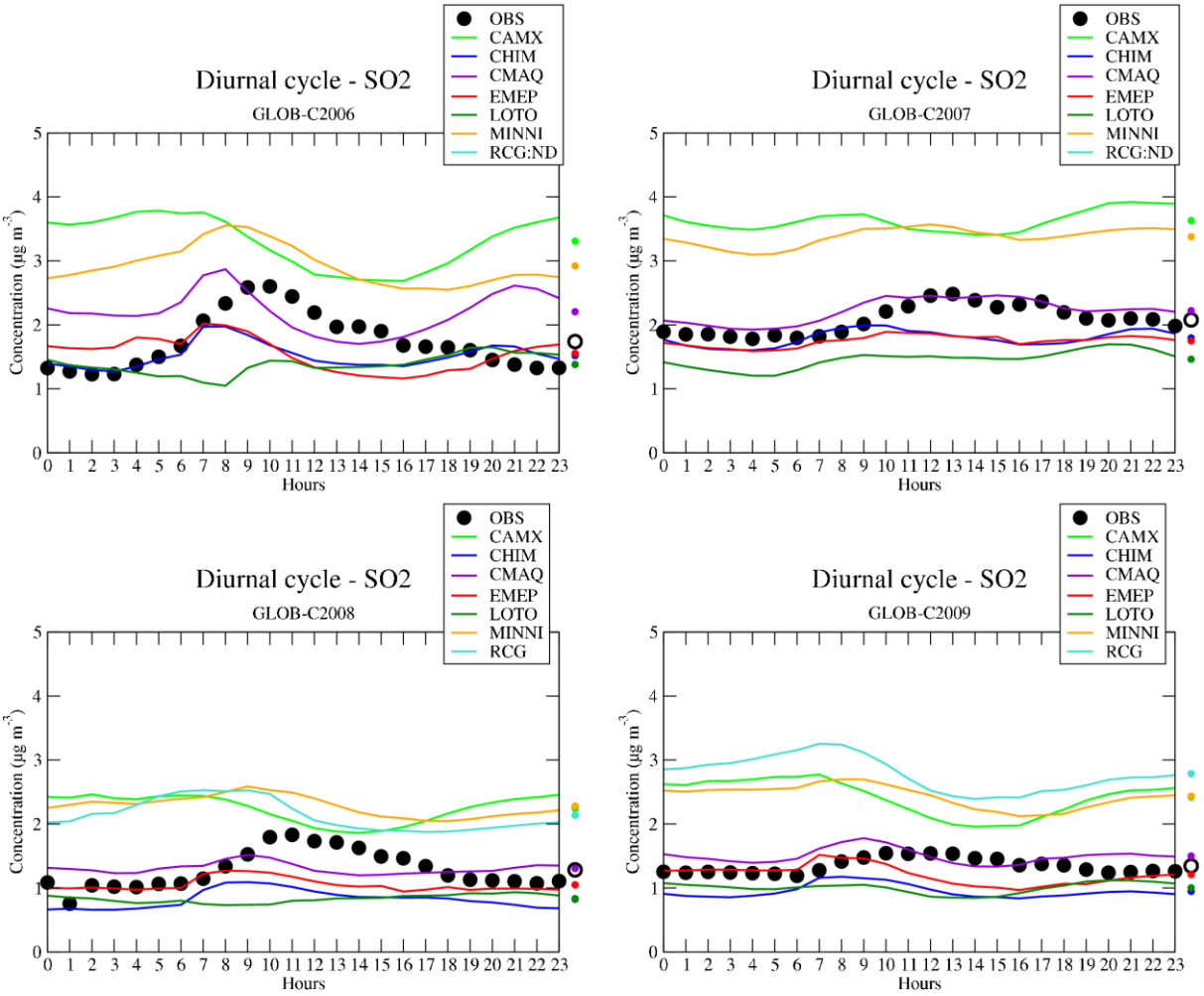



**Fig. 11~~12~~: Mean SO2 diurnal cycles for all campaigns simulated by the models compared with observations. Averaged concentrations are provided on the right side of the charts.**

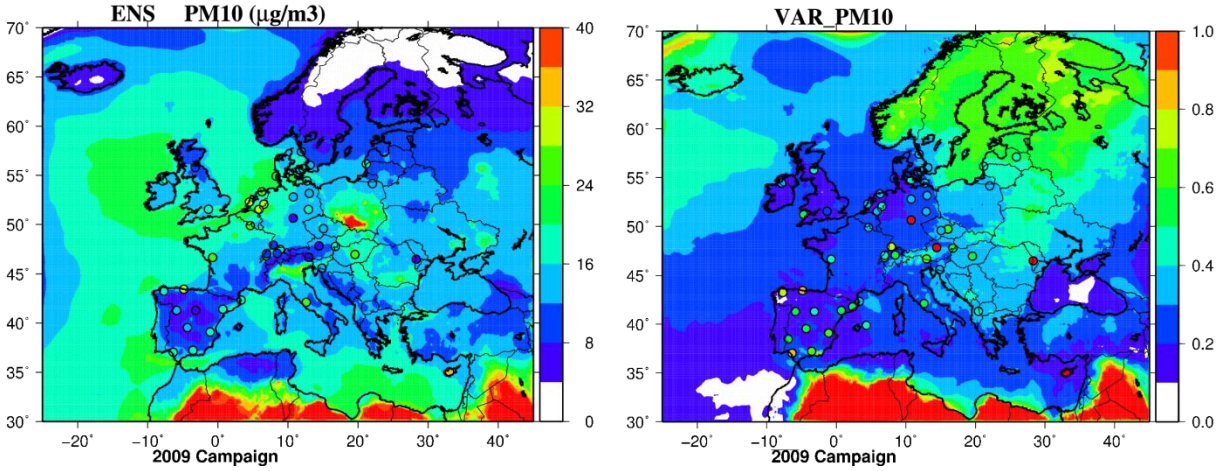



**Fig. 12~~13~~: *Left column*: Mean ~~$PM_{10}$~~PM10 concentrations (µg $m^{-3}$) of the "ensemble" (ENS) for the 2009 campaign with corresponding observations (coloured dots). *Right column*: coefficient of variation of models (no unit) constituting the ensemble with corresponding normalized root mean square errors of the "ensemble" (coloured dots). Red color is assigned for values exceeding the color scale.**

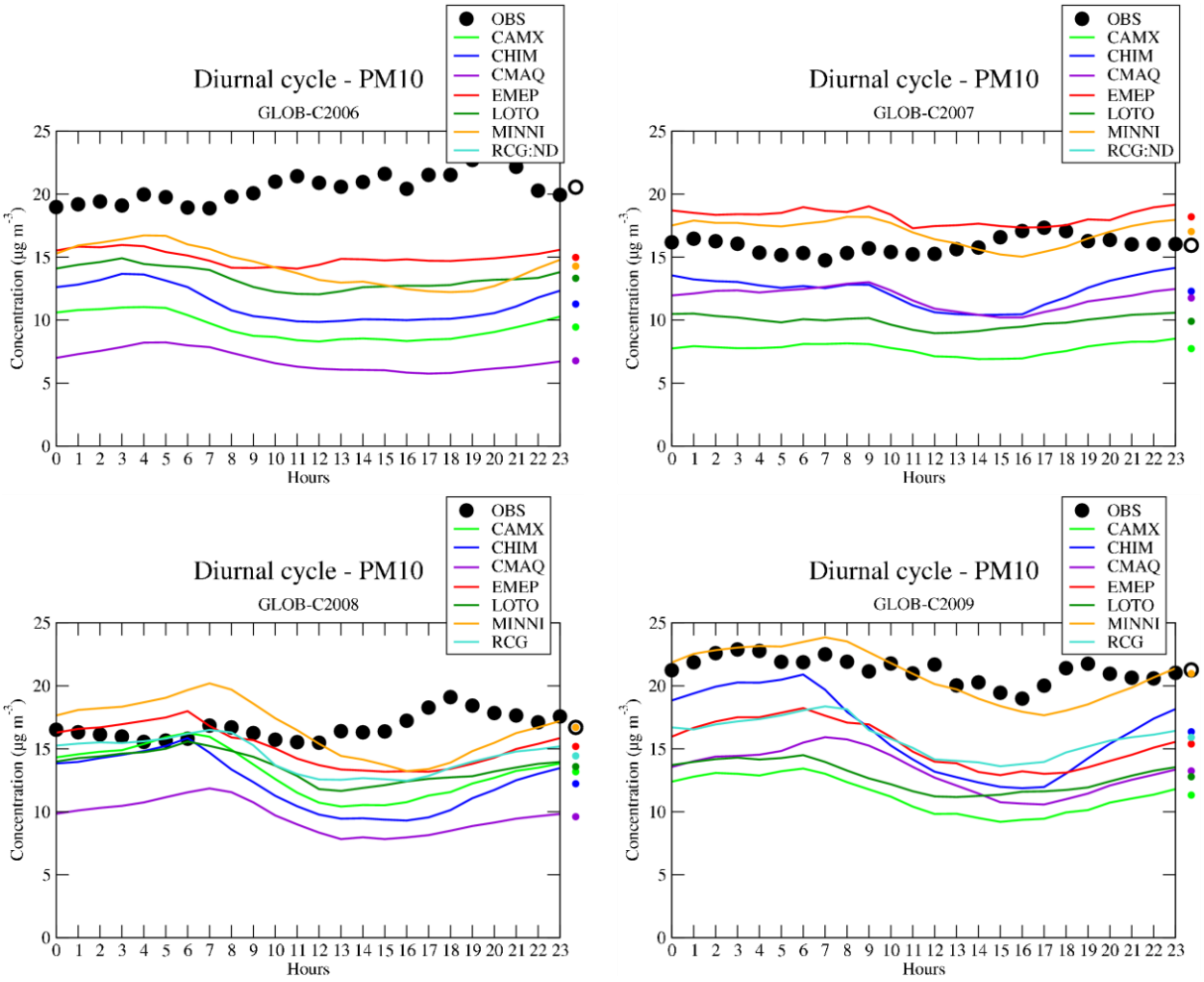



**Fig. 1314: Mean diurnal cycles of ~~$PM_{10}$~~PM10 for all campaigns simulated by the models compared with observations. Averaged concentrations are provided on the right side of the charts.**

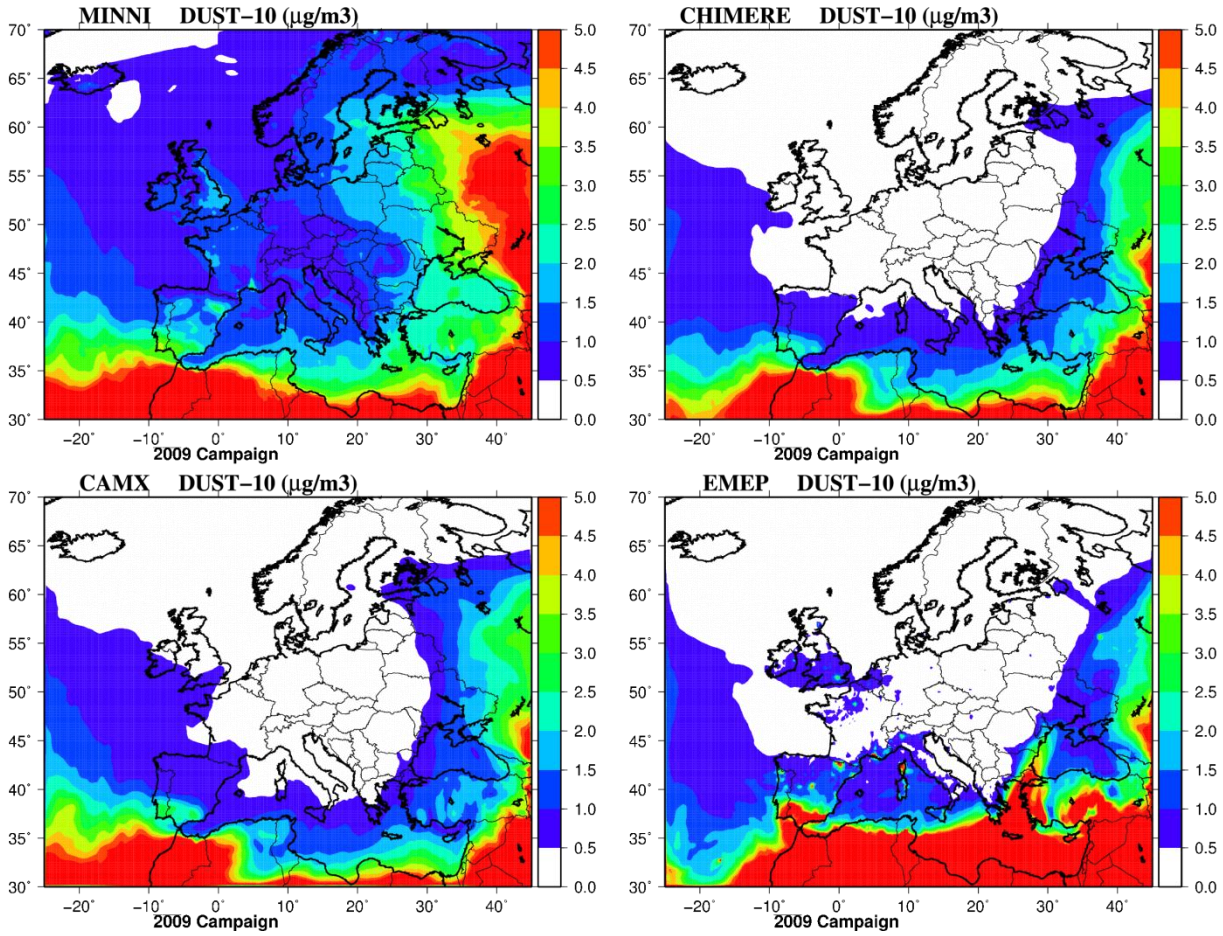



**Fig. ~~14~~15: Mean dust concentrations (µg $m^{-3}$) in the ~~$PM_{10}$~~PM10 fraction for the 2009 campaign computed by the MINNI, CHIMERE, ~~CAMx~~ CAMx and EMEP models.**

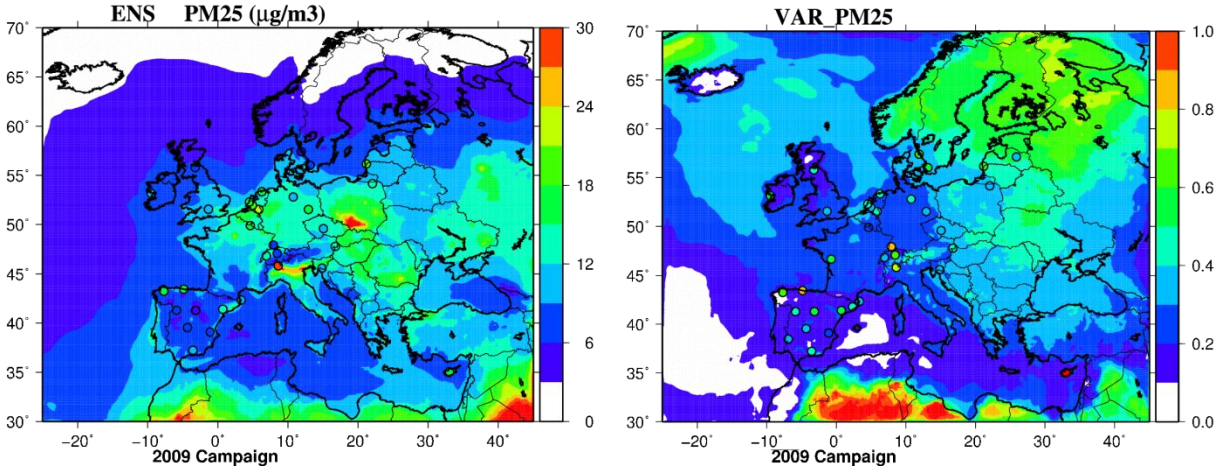



**Fig. 1516: *Left column*: Mean ~~$PM_{2.5}$~~PM2.5 concentrations (µg $m^{-3}$) of the "ensemble" (ENS) for the 2009 campaign with corresponding observations (coloured dots). *Right column*: coefficient of variation of models (no unit) constituting the ensemble with corresponding normalized root mean square errors of the "ensemble" (coloured dots). Red color is assigned for values exceeding the color scale.**

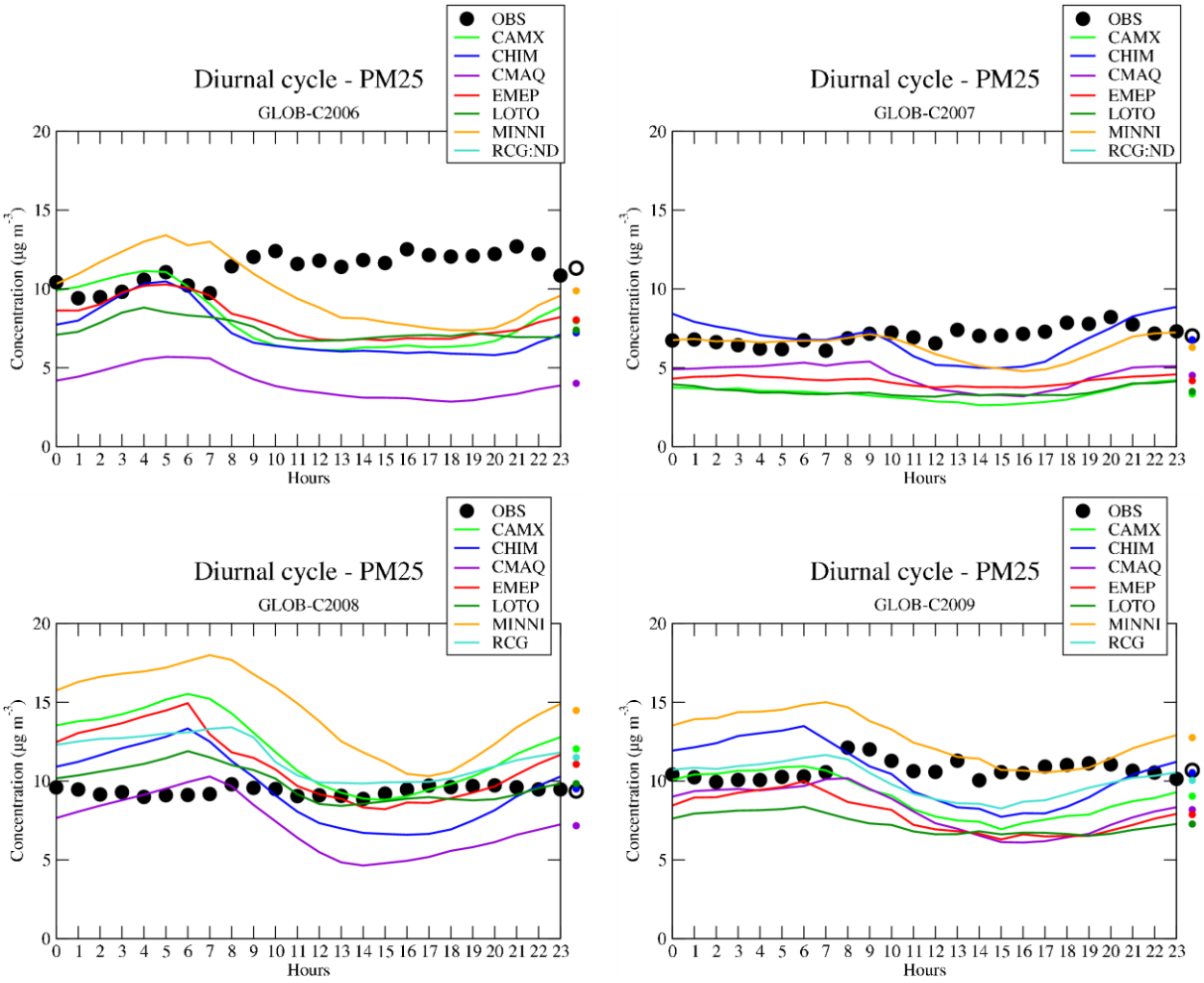



**Fig. 16~~17~~: Mean diurnal cycles of ~~PM$_{2.5}$~~PM2.5 for all campaigns simulated by the models compared with observations. Averaged concentrations are provided on the right side of the charts.**

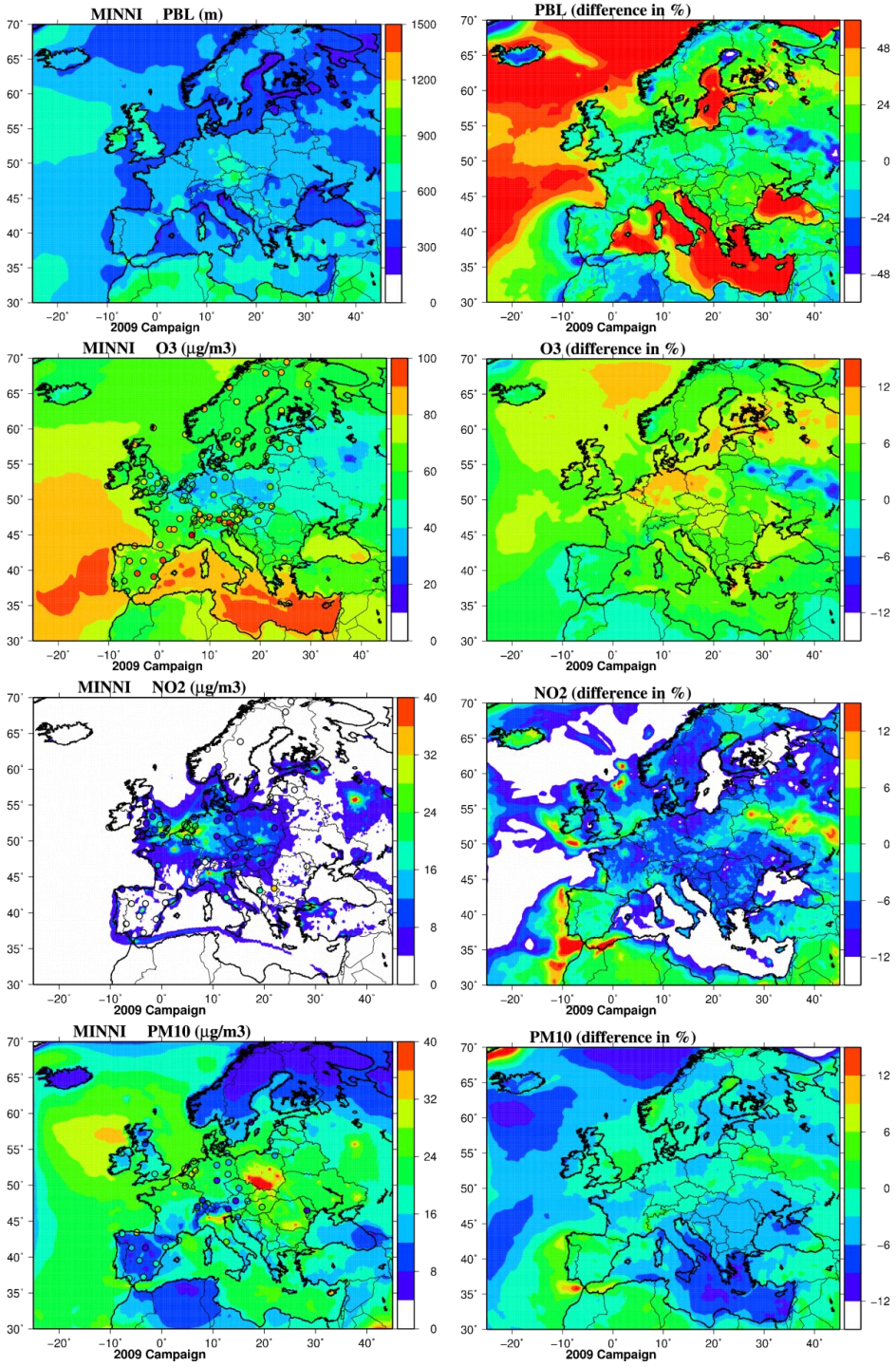



**Fig. 17~~18~~: Left graphs show the average PBL heights and the average concentrations for $O_3$, $NO_2$ and ~~$PM_{10}$~~PM10 using original MINNI's parameterizations. Right graphs show the percentage difference between the average concentrations calculated with PBL heights given by IFS ($PBL_{IFS}$) and by MINNI's parameterizations ($PBL_{MINNI}$). Red color is assigned for values exceeding the color scale.**

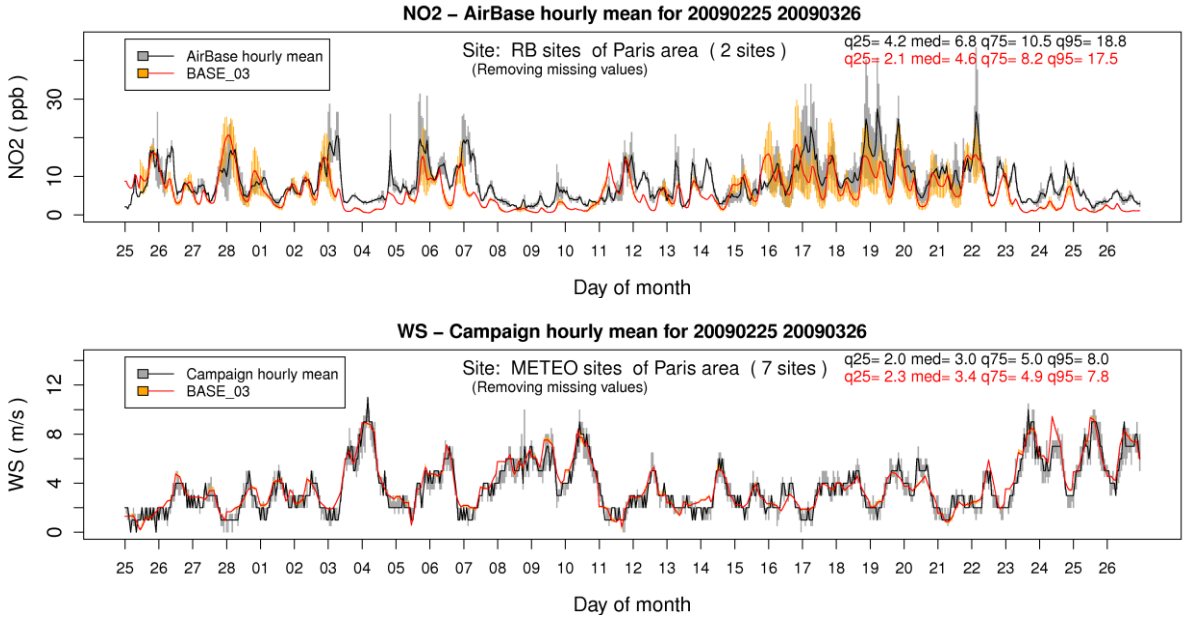



**Fig. 18~~19~~: Time series of hourly Box plots showing the distribution of the observed and computed $NO_2$ concentration (top) and wind speed (bottom) for ~~CAMx~~ CAMx (meteorology from IFS). Observations are in black/grey; modelled values in red/orange. Bars show the 25$^{th}$ -75$^{th}$ quantile interval, while the median is displayed by the continuous line. The 25$^{th}$, 50$^{th}$, 75$^{th}$, and 95$^{th}$ quantile of the whole campaign are reported too. Comparison of computed and observed boxplot time series evaluated at Airbase and meteorological sites, available over the Paris area.**

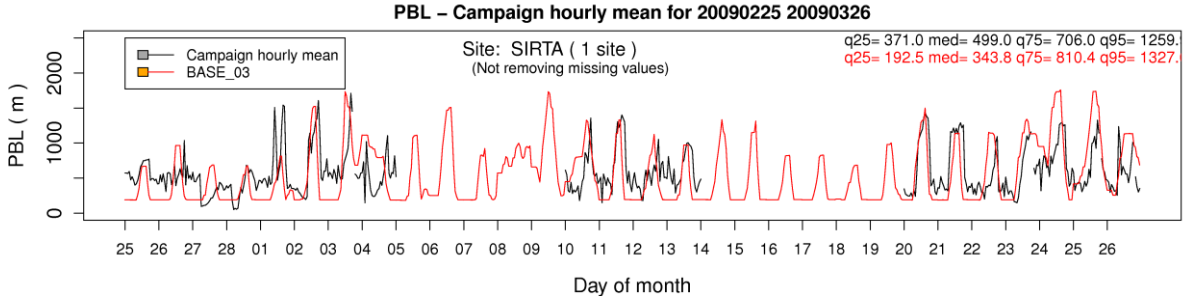



**Fig. 19~~20~~: Time series of hourly Box plots showing the distribution of the observed and computed PBL height. Observations are in black/grey; modelled values in red/orange. Bars show the 25 $^{th}$ -75 $^{th}$ quantile interval, while the median is displayed by the continuous line. The 25 $^{th}$, 50 $^{th}$, 75 $^{th}$, and 95 $^{th}$ quantile of the whole campaign are reported too. Comparison of computed and observed boxplot time series evaluated at SIRTA site.**

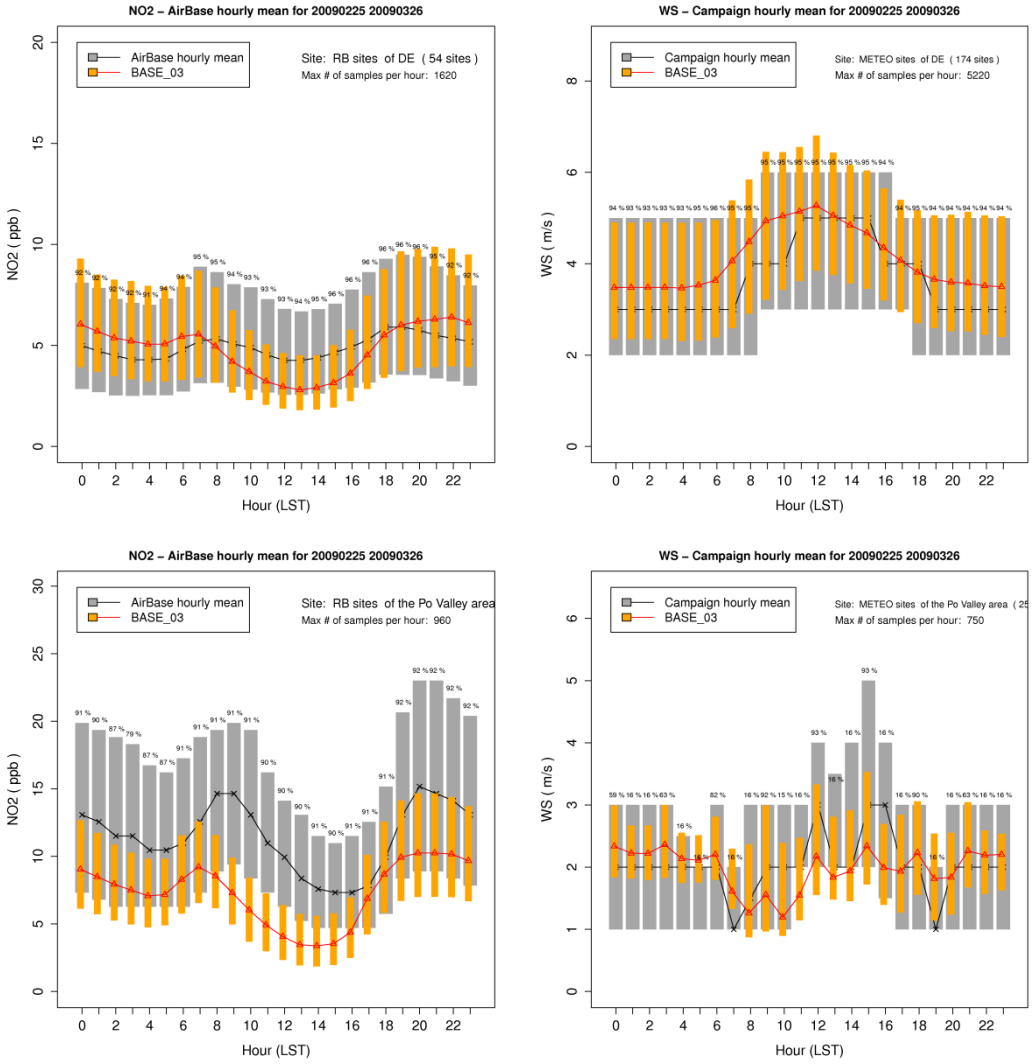



**Fig. 2021: Time series of hourly Box plots showing the distribution of the diurnal cycle observed and computed $NO_2$ concentration (left) and wind speed (right) over Germany (top panels) and Po valley (bottom panels). Observations are in black/grey; modelled values in red/orange. Bars show the 25$^{th}$ -75$^{th}$ quantile interval, while the median is displayed by the continuous line. The 25$^{th}$, 50$^{th}$, 75$^{th}$, and 95$^{th}$ quantile of the whole campaign are reported too. Comparison of computed and observed boxplot time series evaluated at AirBase and meteorological sites, available over Germany and Po valley. Hour is in UTC time.**

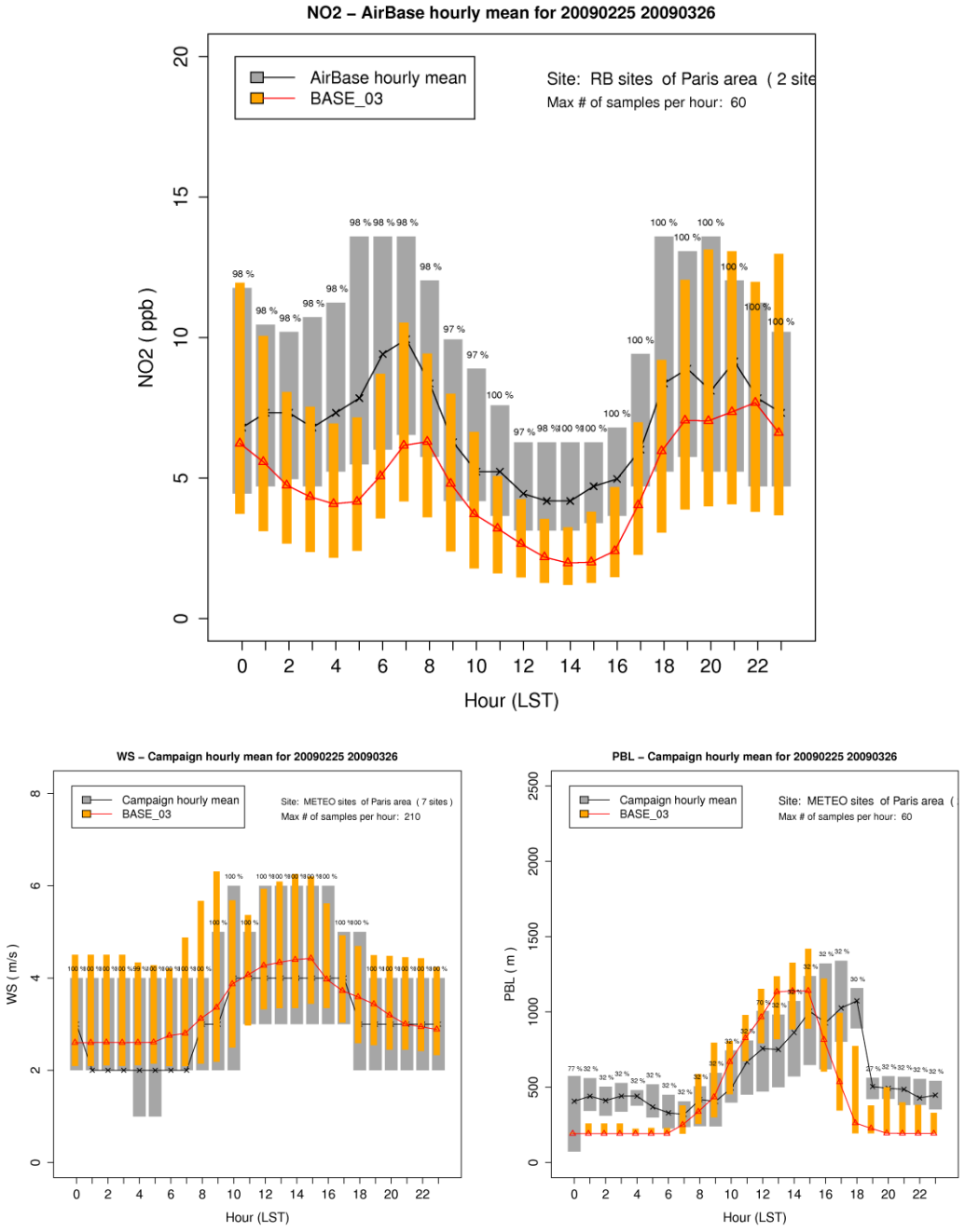



**Fig. 2122: Time series of hourly Box plots showing the distribution of the diurnal cycle observed and computed $NO_2$ concentration (top), wind speed (bottom left) and PBL height (bottom right). Observations are in black/grey; modelled values in red/orange. Bars show the 25$^{th}$ -75$^{th}$ quantile interval, while the median is displayed by the continuous line. The 25$^{th}$, 50$^{th}$, 75$^{th}$, and 95$^{th}$ quantile of the whole campaign are reported too. Comparison of computed and observed boxplot time series evaluated at AirBase and meteorological sites, available over the Paris area. Hour is in UTC time.**

**List of tables**

**Table 1: Models involved in the study**

| Teams | Models with references | Model acronym in this study | Simulated periods |
|---|---|---|---|
| PSI/RSE | **CAMx** (ENVIRON, 2011~~03~~) | CAMX | 2006, 2007, 2008, 2009 |
| INERIS | **CHIMERE** (Menut *et al*., 2013) | CHIM | 2006, 2007, 2008, 2009 |
| HZG | **CMAQ** (Byun *et al*., 2006; Matthias et al., 2008) | CMAQ | 2006, 2007, 2008, 2009 |
| MSC-W - Met.NO | **EMEP** (Simpson *et al*., 2012) | EMEP | 2006, 2007, 2008, 2009 |
| TNO | **LOTOS-EUROS** (Sauter *et al*., 2014) | LOTO | 2006, 2007, 2008, 2009 |
| ENEA/ARIANET | **MINNI** (ARIANET, 2004) | MINNI | 2006, 2007, 2008, 2009 |
| FUB | **RCG** (Stern *et al*., 2006) | RCG | 2008, 2009 |

**Table 2: Synthetic description of models (part 1)**

| | EMEP | CHIMERE | LOTOS-EUROS | RCG | CMAQ | MINNI | CAMx |
|---|---|---|---|---|---|---|---|
| **version** | rv4.1.3 | Chimere2013 | v1.8 | v2.1 | V4.7.1 | FARM V3.1.12 | V5.40 |
| | | | **VERTICAL MODEL STRUCTURE** | | | | |
| **Vertical layers** | 20 sigma | 9 sigma | 4 (3 dynamic layers and a surface layer) | 6 fixed terrain following layers | 30 sigma | 16 fixed terrain-following layers | 33 sigma |
| **Vertical extent (hpa or m)** | 100 hPa | 500 hPa | 3500 m | 3000 m | 100 hPa | 10000 m | 8000 m |
| **First layer depth** | 90 m | 20 m | 25 m | 25 m | 42 m | 40 m | 20 m |
| **Correction of first level concentration** | Yes | No | Yes | No | No | No | No |
| | | | **NATURAL EMISSIONS** | | | | |
| **Biogenic VOC** | Based upon maps of 115 species from Koeble and Seufert (2001), and hourly temperature and light. See Simpson *et al.* (2012) | MEGAN model v2.04 | Based upon maps of 115 species from Koeble and Seufert (2001), and hourly temperature and light (Beltman *et al.*, 2013) | Based upon maps of 115 species from Koeble and Seufert (2001), and hourly temperature and light.using emissions factors of Simpson *et al.* (1999) | BEIS 3.14 emission inventory (Vukovich and Pierce, 2002) | MEGAN model v2.04 | MEGAN model v2.1 |
| **Soil NO** | After Simpson *et al.* (2012) | MEGAN model v2.04 | Not used here | From Simpson *et al.* (1999) | BEIS 3.1.4 | MEGAN v2.04 | MEGAN model v2.1 |
| **Lightning emissions** | Climatological fields, Köhler *et al.* (1995) | No | No | No | No | No | No |
| **Sea salt** | Monahan (1986) and Martensson (2003), see Tsyro *et al.* (2011). | Monahan *et al.* (1986) | Martensson *et al.* (2003) and Monahan *et al.* (1986) | Gong *et al.* (1997) and Monahan *et al.* (1986) | Zhang *et al.* (2005) and Clarke *et al.* (2006) | Zhang *et al.* (2005) | Not used |
| **Windblown Dust** | After Simpson *et al.* (2012) | No | Denier van der Gon *et al.* (2009). | Loosemore and Hunt (2000), Claiborn *et al.* (1998) | No | Vautard *et al.* (2005) | No |
| **Road traffic suspension** | Denier van der Gon *et al.* (2009). | No | No | No | No | No | No |
| | | | **LANDUSE** | | | | |
| **Landuse database** | CCE/SEI for Europe, elsewhere GLC2000 | GLOBCOVER (24 classes) | Corine Land Cover 2000 (13 classes) | Corine Land Cover 2000 (13 classes) | Corine Land Cover 2006 (44 classes) | Corine Land Cover 2006 (22 classes) | USGS data |
| **Resolution** | Flexible, CCE/SEI ~ 5 km | About 300 m | 1/60 x 1/60 degrees | 1/60 x 1/60 degrees | About 250 m | About 250 m | 10 minutes |

**Table 3: Synthetic description of models (part 2)**

| | EMEP | CHIM | LOTO | RCG | CMAQ | MINNI | CAMX |
|---|---|---|---|---|---|---|---|
| | | | | **METEOROLOGY** | | | |
| **Driver** | ECMWF IFS | ECMWF IFS + urban mixing | ECMWF IFS | ECMWF IFS + Observations | COSMO CLM | ECMWF IFS | ECMWF IFS |
| **Resolution** | 0.25°x0.25° | 0.25°x0.25° | 0.25°x0.25° | 0.25°x0.25° | 24 km x 24 km (Lambert Conformal Conic Projection) | 0.25°x0.25° | 0.25°x0.25° |
| | | | | **PROCESSES** | | | |
| **Advection scheme** | Bott (1989a,b) | Van Leer (1984) | Walcek (2000) | Walcek (2000) modified by Yamartino (2003). | Blackman cubic polynomials (Yamartino, 1993 | Blackman cubic polynomials (Yamartino, 1993) | Bott (1989a,b) |
| **Vertical diffusion** | Kz approach following O'Brien (1970) and on Jeričevič *et al.* (2010) for stable and neutral conditions | Kz approach following (Troen and Mart, 1986) IFS PBL | Kz approach IFS PBL | Kz-approach and IFS PBL | ACM2 PBL scheme (Pleim, 2007a) | Kz following Lange (1989). PBL from Maul (1980) version of Carson (1973) algorithm for day times. | Kz approach following O'Brien (1970) IFS PBL |
| **Dry deposition scheme** | resistance approach for gases, Venkatram and Pleim (1999) for aerosols, Simpson *et al.* (2012) | resistance approach Emberson (2000a,b) | Resistance approach,DEPAC3.11for gases, Van Zanten *et al.* (2010) and Zhang et al (2001) for aerosols | resistance approach, DEPAC-module | Resistance approach, Venkatram and Pleim (1999) | Resistance model (Walcek and Taylor, 1986; Wesely, 1989) | Resistance model for gases (Zhang *et al.*,2003) and aerosols (Zhang *et al*., 2001) |
| **Compensation points** | No, but zero $NH_3$ deposition over growing crops | No | Only for $NH_3$ (for stomatal, external leaf surface and soil = 0) | No | No | No | No |
| **Stomatal resistance** | DO3SE-EMEP: Emberson *et al.* (2000a,b), Tuovinen *et al.* (2004), Simpson *et al.* (2012) | Emberson (2000a,b) | Emberson (2000a,b) | Wesely (1989) | Wesely (1989) | Wesely (1989) | Wesely (1989) |
| **Wet deposition of gases** | In-cloud and sub-cloud scavenging coefficients | In-cloud and sub-cloud scavenging coefficients | sub-cloud scavenging coefficient | pH dependent scavenging coefficients | In-cloud and sub-cloud scavenging which depends on Henry's law constants, dissociation constants and cloud water pH. Chang *et al.* (1987) | In-cloud and sub-cloud scavenging coefficients (EMEP, 2003) | In-cloud and sub-cloud scavenging model for gases and aerosols (Seinfeld and Pandis, 1998) |
| **Wet deposition of particles** | In-cloud and sub-cloud scavenging | In-cloud and sub-cloud scavenging | Sub-cloud scavenging coefficient | Sub-cloud scavenging coefficients | In-cloud and sub-cloud scavenging | In-cloud and sub-cloud scavenging coefficients | In-cloud and sub-cloud scavenging model for gases and aerosols (Seinfeld and Pandis, 1998) |
| **Gas phase chemistry** | EmChem09 (Simpson *et al*. | MELCHIOR | TNO CBM-IV | CBM-IV | CB-05 with chlorine chemistry | SAPRC99 (Carter, 2000a,b) | CB-05 (Yarwood *et al.*, 2005) |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | 2012) | | | | extensions (Yarwood *et al.*, 2005) | | |
| **Cloud chemistry** | Aqueous $SO_2$ chemistry | Aqueous $SO_2$ chemistry and ph computation | No | Simplified aqueous $SO_2$ chemistry | Aqueous $SO_2$ chemistry (Walcek and Taylor, 1986) | Aqueous $SO_2$ chemistry (Seinfeld and Pandis, 1998) | Aqueous $SO_2$ chemistry RADM-AQ (Chang *et al.*, 1987) |
| **Coarse nitrate** | Yes | No reactions with Ca or Na but coarse might exist with transfer from smaller particles | Yes | Yes | No | No | No |
| **Secondary Inorganic equilibrium** | MARS (Binkowski and Shankar,1995) | ISORROPIA (Nenes *et al.*, 1999) | ISORROPIA v.2 | ISORROPIA | ISORROPIAv1.7 | ISORROPIA v1.7 (Nenes *et al.*, 1998) | ISORROPIA (Nenes *et al.*, 1998) |
| **SOA formation** | VBS-NPAS – Simpson *et al.* (2012) | After Bessagnet *et al.* (2009) | Based on Bergström et al (2012) | SORGAM module (Schell *et al.*, 2001) | SORGAM module (Schell *et al.*, 2001) | SORGAM module (Schell *et al.*, 2001) | CAMx-VBS (beta version) (Koo *et al.*, 2014) |
| **VBS** | Yes, Bergström *et al* (2012), Simpson *et al.* (2012) | No | Yes, based on Bergström et al (2012) | No | No | No | Yes based on Koo *et al.* (2014) |
| **Aerosol model** | Bulk- approach (2 modes) | 8 bins (40 nm to 10 µm) | Bulk- approach (2 modes) | Bulk approach (2 modes) | AERO5 (Carlton *et al.*, 2010), Log-normal approach (3 modes) | AERO3 (Binkowski, 1999); 3 modes: Aitken, accumulation, coarse | Bulk- approach (2 modes) |
| **Aerosol physics** | No dynamics | Coagulation/condensation/nucleation | No dynamics | No dynamics | Coagulation/condensation/nucleation | Coagulation/condensation/nucleation | No dynamics |

**Table 4: Error statistics used to evaluate model performance (M and O refer respectively with Model and Observations data, and N is the number of observations)**

| | |
|---|---|
| **Mean Bias** | $(\bar{M} - \bar{O})$ with $\bar{M} = \frac{1}{N}\sum_{i=1}^{N} M_i$ and $\bar{O} = \frac{1}{N}\sum_{i=1}^{N} O_i$ |
| **Normalised Mean Bias** | $NMB = (\bar{M} - \bar{O})/\bar{O}$ |
| **Mean Bias** | $MB = (\bar{M} - \bar{O})$ |
| **Mean Gross Error** | $MGE = \frac{1}{N}\sum_{i=1}^{N} \lvert M_i - O_i \rvert$ |
| **Standard Deviation** | $SD_X = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(X_i - \bar{X})^2}$ with X=O or M |
| **Root Mean Square Error** | $RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(M_i - O_i)^2}$ |
| **Normalized Root Mean Square Error** | $NMSE = \frac{1}{\bar{M}}\sqrt{\frac{1}{N}\sum_{i=1}^{N}(M_i - O_i)^2}$ |
| **Correlation Coefficient** | $R = \left(\sum_{i=1}^{N}(M_i - \bar{M})(O_i - \bar{O})\right) \Big/ \left(\sqrt{\sum_{i=1}^{N}(M_i - \bar{M})^2 \times \sum_{i=1}^{N}(O_i - \bar{O})^2}\right)$ |

**Table 5: ~~$PM_{10}$~~PM10 and ~~$PM_{2.5}$~~PM2.5 spatial correlations for all campaigns**

| | 2006 | | 2007 | | 2008 | | 2009 | |
|---|---|---|---|---|---|---|---|---|
| | ~~$PM_{10}$~~PM10 | ~~$PM_{2.5}$~~*PM2.5* | ~~$PM_{10}$~~PM10 | ~~$PM_{2.5}$~~*PM2.5* | ~~$PM_{10}$~~PM10 | ~~$PM_{2.5}$~~*PM2.5* | ~~$PM_{10}$~~PM10 | ~~$PM_{2.5}$~~*PM2.5* |
| **CAMx** | 0.58 | *0.32* | 0.24 | *0.60* | 0.32 | *0.47* | 0.07 | *0.46* |
| **CHIMERE** | 0.65 | *0.32* | 0.58 | *0.78* | 0.39 | *0.42* | 0.55 | *0.66* |
| **CMAQ** | 0.50 | *0.19* | 0.50 | *0.80* | 0.11 | *0.42* | 0.11 | *0.37* |
| **EMEP** | 0.75 | *0.24* | 0.56 | *0.62* | 0.34 | *0.48* | 0.68 | *0.61* |
| **LOTOS-EUROS** | 0.34 | *0.05* | 0.50 | *0.61* | 0.27 | *0.37* | 0.50 | *0.37* |
| **MINNI** | 0.61 | *0.43* | 0.55 | *0.58* | 0.20 | *0.45* | 0.32 | *0.51* |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **RCG** | ND | *ND* | ND | *ND* | 0.62 | *0.32* | 0.44 | *0.36* |