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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contribution of atmospheric processes affecting the dynamics of air pollution in south-western Europe during a typical summertime photochemical episode" *by* M. Gonçalves et al.

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Interactive comment on "Contribution of atmospheric processes affecting the dynamics of air pollution in south-western Europe during a typical summertime photochemical episode" by M. Gonçalves et al.

The authors gratefully acknowledge the comments of the Anonymous Referee #1, which may help to improve the quality of this manuscript and to clarify some issues that were not previously addressed. Please find below the item by item response to the Referee comments.

Q: Page 18,467, line 24. Please update the US-EPA 2005 reference. The final guidance was published in 2007 and is called "Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, PM2.5, and Regional Haze", EPA -454/B-07-002, 262 pages. Please also note that this final guidance does not suggest any MNBE, UPA, and MNGE threshold criteria to determine acceptable model performance in regulatory applications (see section 18.6 of the above document)

A: The authors have updated the aforementioned reference and changed the text as follows: "The US Environmental Protection Agency guidelines (US-EPA, 1991; 2007) recommend the use of different statistical parameters and combination of methods to assess a model performance, among them the mean normalized bias error (MNBE), the unpaired peak prediction accuracy (UPA) and the mean normalized gross error (MNGE)" Although the US-EPA (2007) guideline does not provide any threshold criteria to determine acceptable model performance, it suggests several statistical parameters to be used for the model evaluation. Among them, those used by the authors to provide a measure of the quality of model predictions have been: Mean Bias, Mean Error, Mean Normalized Bias, Unpaired Peak Accuracy and Mean Normalized Gross Error. The authors think that these parameters together with the evolution graphics shown for several air quality stations (Figures 2 and 3) provide an overview of the evaluation tasks carried out and ensure the model performance for the case of study. The evaluation results are discussed in section 3.1 of the manuscript for the regions of study (Northeastern and Central Iberian Peninsula) and for specific rural and urban background stations in both areas.

The following questions are related to several hypotheses made by the authors to explain the general trends of the model performance in predicting the concentration of gaseous pollutants. Their main purpose is to identify the major causes of uncertainty, taking into account the trends in the statistical parameters used (Bias, Error, MNBE, MNGE and UPA). The modelling results (but also previous studies for similar situations)

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are taken as a basis for the discussion. The authors agree with the referee in considering that not only the meteorological simulation inaccuracies may be in the origin of the deviation of pollutants concentrations. All the questions suggested for the referee were addressed as follows:

Q: Page 18,468, lines 11-13: Was the WRF-ARW model evaluated for this particular case study to confirm that daytime wind flows were underestimated? The findings by Jimenez et al. (2006) were based on MM5 simulations and may not be applicable here. In the absence of a more detailed analysis of the meteorological fields, this possible explanation for the ozone overpredictions remains a hypothesis that cannot be substantiated.

A: Meteorological data from the CEAM, METAR and AEMET networks were used to assess the WRF-ARW performance in predicting wind speed at 10 m, wind direction and temperature at 2 m. Data from 18 stations were available in the NEIP domain and 24 in the CIP domain. The WRF-ARW model evaluation for the case of study, 17-18 June, 2004, shows a differentiated behaviour for the wind speed estimates as a function of the geographical location. The daily wind flows (from 600 to 1800 UTC) in the North-eastern Iberian Peninsula are overestimated for some stations (MB below 1.0 ms-1) and underestimated for those stations located in coastal areas (MB below -1.5 ms-1). The average MB for the whole NEIP the 18 June, 2004 is -0.15 ms-1. Nevertheless the wind speed estimates in the CIP are overestimated during daytime (MB around 0.8 ms-1 for the 18 June, 2004) and underpredicted during the 1800 to 600 UTC period (17-18 June, 2004)- MB=-0.7ms-1 -. These differences were stated in the manuscript, pointing out that the underestimations in daily wind speeds occur mainly in the NEIP or coastal domain: "Previous studies (Jiménez et al., 2006; Jiménez-Guerrero et al., 2008) point that under low pressure gradient situations mesoscale models tend to underestimate daytime wind flows in coastal areas, which would favour the O3 accumulation" Those results are also supported by previous works performed in the Earth Science Department of the Barcelona Supercomputing Center (see Jiménez

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et al., 2006; Jiménez-Guerrero et al., 2008, as referenced in the manuscript) and the current meteorological forecast evaluation for the whole national territory available on line at http://www.bsc.es/projects/earthscience/metar/sub_fc_val_google.php

Q: Page 18,468, lines 18 - 20: Which analyses were performed to establish that the weakness of the meteorological model indeed is responsible for the NO2 overpredictions in the CIP? How did the authors rule out that errors in emissions and/or chemistry contributed to the overprediction?

A: The authors agree with the referee, the overpredictions of NO2 may have several causes, among them the meteorological fields (RMSE in the CIP during the 18 June, 2004 is on average 2.2 ms-1), but also emissions account and gas-phase chemistry uncertainties in the model, to clarify this aspect the sentence was reworded as follows: "NO2 concentrations are overpredicted in the CIP domain (positive bias of 5.89%), which could be attributed to the weaknesses of the model to represent wind flows under this low pressure gradient situation. The uncertainties related to the emissions account and the atmospheric chemistry representation in the CMAQ model could also play an important role"

Q: Page 18,468, lines 20 - 23: Which analyses were performed to establish that inaccuracies of the model chemistry are responsible for the NO2 overpredictions in the NEIP domain? How did the authors rule out that errors in emissions and/or meteorology contributed to the overprediction? Page 18,468, lines 23 - 29: Couldn't this point to a problem with the emission inventories?

A: There's not a unique cause for NO2 underprediction in the NEIP. The authors base their assumptions in previous works and discuss the effect on NO2 predictions when increasing the vertical resolution of the model. The fact that increasing resolution does not improve clearly the NO2 predictions could lead to the assumption that there are other factors producing this underprediction, among them the chemistry represented in the model, but also the emissions account. This is now reflected in the manuscript

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as follows: "This fact suggests that discrepancies between modelled and measured levels do not only depend on transport patterns, but also on the chemical behaviour represented by the model and on the emissions estimates."

Q: Page 18,470, lines 3-6: Was the WRF-ARW model evaluated for this particular case study to confirm that surface winds were overestimated during this time period? Was this overestimation observed at all observation sites?

A: The wind speed at 10 m for the 1600 to 2000 UTC period is overestimated at some stations in the NEIP, maximum mean bias of 1.6 ms-1 the 17 June, 2004 and 6.45 ms-1 the 18 June, 2004 for the Cabo Creus station (42.31N,3.31E). Nevertheless the behaviour of different locations significatively differs, as pointed out by the referee's question. The MB for the Els Torm station (41.39N, 0.72E) in the same period of 18 June, 2004 is -2.27 ms-1. From those stations located in the coastal area, only the Barcelona airport station (41.28N, 2.07E) and the Tortosa station ((40.82N, 0.49E) which corresponds to the Ebro river mouth) showed slight underestimations of the wind speed during the dust period. Moreover, previous works confirms the weakness of the WRF-ARW model in predicting the diurnal to nocturnal wind speeds change in coastal areas (Jiménez-Guerrero et al., 2008)

Q: Page 18,471, lines 17 - 18: Please specify if these concentration gradients are primarily horizontal, vertical, or both.

A: The authors are referring mainly to the effect of the vertical gradients of O3 concentration, which enhance the vertical diffusion processes. Although this effect also exists in the horizontal component, the contribution of horizontal diffusion is much lower. This is now clarified on the text as follows: "The vertical gradients of O3 concentration generated in these areas, larger during daytime, increase the contribution of diffusion processes to ground-level O3 (up to 200 μg m-3 h-1 fluxes, mainly from upper vertical layers)"

Q: Page 18,472, lines 14 - 24: Please clarify if the description of circulation patterns in

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the coastal domain was based on previous work or was established specifically for this episode. If it was based on previous work, please provide references and information to support that these findings are applicable to the episode studies here. If it was based on an analysis of the WRF-ARW fields for this study, please provide the supporting figures and analyses.

A: This discussion is based on the current simulation data, but also agrees with previous studies (both based on experimental campaigns and on modelling results, as cited in the text: Baldasano et al., 1994; Millán et al., 1997; Soriano et al., 2001; Pérez et al., 2004; Jiménez et al., 2006, among others confirm the modelling results of our case of study). The meteorological simulation results which were not included in the text in order to avoid redundancies, but they can be provided as additional material for the final version of the manuscript. Moreover the authors provide Figures 5 to 6 in the manuscript, where the transport patterns are depicted for each domain in order to illustrate this discussion.

Q: Page 18,473, lines 15 - 18: Figure 5 shows ozone production only for the 10:00 UTC panels which does not really correspond to the "central hours" of the day

A: The O3 chemical production occurs during morning and central hours of the day (from 8:00 to 16:00 UTC). The maximum production could be fixed around 11.00-12.00 UTC. The 10.00 h and 18.00 h panels were selected as representative of a daytime and a night-time period behaviour. The authors agree with the referee in considering that the previous wording could lead to misunderstandings compared to the figure and have modified the paragraph as follows: "The process analysis indicates that maximum chemical production of O3 does not occur in the first vertical layer of the model, but in layers aloft during the morning and central hours of the day" Moreover, as stated in the manuscript a deeper discussion on the formation periods is provided in the next section of the article for each specific domain analysed.

Q:Page 18,477, lines 13 - 27: Please clarify how you define "chemical sensitivity" and

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"NOx sensitive" in this context. Is it based on indicator ratios, NOx/VOC ratios, etc? Please provide an explanation and references. Maybe it would be better to think of this analysis in terms of chemical regimes?

A:The chemical sensitivity regimes in those areas were studied in previous works (Palacios et al., 2002; Jiménez et al., 2004; Gonçalves et al., 2008) and were observed to be different. Nevertheless the authors here wanted to explain the different gas-phase chemistry behaviour observed in the study areas and are not referring specifically to O3 response to emissions abatement or sensitivity studies based on chemical indicators. Intending to clarify this point, the paragraph was modified as follows: "Differences in chemical behaviour are observed for the studied domains (Figure 11). The coastal domains present similar chemical behaviours; the NMVOCs are the only locally emitted precursors, which react with transported NOx generating O3 by gas-phase chemistry. The CIP-MAX1 domain includes emitters of NOx and NMVOCs, which react forming O3, nevertheless the O3 chemistry does not involve destruction of NMVOCs in the same magnitude as the coastal domains. In absence of solar radiation, when the O3 stopped forming, the NO oxidation continues producing net NO2 (1800-1900 UTC period). In the CIP-MAX2 domain the emissions are an important source of NMVOCs. These are chemically destroyed in the 1000 to 1900 UTC period on 17 June, being the generation of O3 by chemistry relatively low. The low contribution of net transport of NOx and the absence of emissions of these compounds in the area may be the main causes for the low O3 production. On the second day of the episode the horizontal advection directly affect the MAX2 involving a much higher net chemical production of O3, being the chemical oxidation of NMVOCs lower"

Q:Page 18,489, line 10: please modify the wording given that the latest US-EPA modelling guidance does not recommend strict pass/fail criteria for model acceptance.

A:The phrase was changed to: "The model performance agrees with European Directives recommendations, nevertheless specifically in background air quality stations tends to overestimate the O3 morning concentrations and underestimate the O3 levels

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during the afternoon"

Q: Page 18,489, lines 11 - 22: Please make sure to distinguish between hypotheses and speculation on the one hand and conclusions on the other hand in this section.

A:This paragraph in conclusions was reworded as follows. "The model performance agrees with European Directives recommendations, nevertheless specifically in background air quality stations tends to overestimate the O3 morning concentrations and underestimate the O3 levels during the afternoon. The findings of this work together with previous studies results allows us to depict the main causes for these deviations, being aware that there is not an unique reason for a model failure in predictions. The chemical destruction may be underpredicted during night-time, which favours the high estimated morning concentrations. Moreover the overpredicted flows during the afternoon and dusk and night-time could cause an enhanced venting of pollutants. The mesoscale meteorological models have shown inaccuracies in predicting wind flows during the dominant stagnant conditions; which together with the underestimations in the emissions account play a fundamental role in these deviations. NO2 is clearly linked to O3, with both inaccuracies in transport and chemical behaviour being on the origin of the errors in the simulated concentrations. On the other hand, O3 peaks and NO2 underprediction in the coastal domain may be related to difficulties for the model to define the accumulation layers formed and characteristic recirculation of pollutants due to the very complex terrain. In the case of SO2 and PM10 a slight underestimation of emissions could be the main cause for the underestimation of these pollutants."

Q: Page 18,489, lines 24 - 25: How was mass consistency tested in this study?

A: The mass consistency for the gaseous pollutants concentrations was tested by a mass balance. It is possible to estimate each hour concentration by adding the processes contribution to the concentration in the previous hour for each cell in the model. In our case this fact was ensured by using the Yamartino scheme for advection in the CMAQ simulation, which includes adjustments for mass consistency. Nevertheless,

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this analysis does not provide any additional result that deserved being discussed in the manuscript, and therefore this sentence was removed from the Conclusions section.

Q: Figure 4: Suggest replacing the full date/time labels with labels showing only the simulation hour (1 - 48) for better readability. Figures 8 - 11: It is very difficult to distinguish between the colors and shading patterns in these figures. This is most pronounced for Figures 8 and 10. Please rework the color scale using a more distinguishable palette, possibly along the lines of Figures 2-3 which are much easier to read.

A: The suggested changes in figures were done intending to make easier their interpretation.

Editorial comments:

All the editorial comments below were added to the manuscript. Please find details on those needing clarification:

Page 18,459, line 10: please specify what these air quality targets are or provide a reference The European Directive considered is the DIR 2008/50/EC which is now indicated in the text and included in the reference list.

Page 18,459, line 11: suggest inserting "processes contributing to" between "fundamental tool to assess the" and "air quality levels

Page 18,460, line 4: please replace "pretends to assess" with "aims to assess"

Page 18,465, line 3: please replace "being the sum of abs(%PCi) exactly" with "but the sum of abs(%PCi) is exactly"

Page 18,466, line 6: please remove "by" from the phrase "contributing also by 24%"

Page 18,468, line 9 / Table 2: Please clarify if these statistics are based on all hours, daytime hours, or peak values.

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The statistics are based in the 48 simulated hours, which was clarified in the manuscript.

Page 18,478, line 3: please change "information about atmospheric processes" to "information about simulated atmospheric processes"

Page 18,479, lines 10 - 11: Please reword the following phrase: "but having the chemical formation some importance at low levels"

This phrase was reworded as follows:

"The central-continental domain (CIP) behaviour slightly differs: the horizontal advection is also the main contributor to O3 surface concentrations, but the chemical formation takes place in the whole vertical column below the PBL"

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 18457, 2008.