Commentary on **"How realistic are air quality hindcasts driven by forcings from climate model simulations? "**

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The authors would like to thank the referees for their comments and suggestions on this paper.

Response to comments by refeeres

Reply to referee ♯1 remarks

Referee: Sect. 2: What do you mean exactly by "two-way nested domains"? Does the chemistry in the smaller domain feed back to the larger domain? Or does the chemistry influence the meteorology (ok, this seems to be unlikely for a CTM)?

Indeed by "two-way nested domains", we mean that the smaller domain can influence the larger domain while the larger domain provides the boundary conditions to the smaller domain. The model is run as a CTM and meteorology is not affected by atmospheric composition.

Referee: Sect. 2: page 2089, line 4: What does "SSTs evolve along the simulation" mean? Are the observed SSTs prescribed for all simulations, or does ARPEGE include an ocean model, or something else?

To provide more details, we modified the text: *"(...) Anthropogenic forcings of ARPEGE-Climate (GHG, aerosols) refer to the climatology of the present time. For the present simulation, ARPEGE-Climate is driven by prescribed observed SSTs (Sea Surface Temperatures) and for the future simulations, the SSTs are thus from RCP8.5 scenario ocean atmosphere coupled simulations. (...)"*

Referee: Sect. 2.3: Which simulation are you evaluating for the use of the classes? Only ANALY seems to make sense, but this should be stated somewhere.

We have used ANALY to illustrate the impacts of the selection of the site classes.
Page 2092, from line 5, we modified the text: *"In order to highlight the effect of site representativeness, we have compared the summertime (JJAS) average diurnal cycles for classes 1–2 (1 and 2), 1–5, 1–10 and 6–10 over France 5 for O3 and NOx (Fig. 1) for the simulation ANALY ".*

Referee: Sect. 3.1.1 Fig. 2: It is really confusing that precipitation is plotted in the second line of Fig. 2 but nothing is said about it in this place. Even if a short reference to it is given in the next subsection. I recommend to add the comparison for the precipitation fields.

Page 2094, from line 3, we expanded the text: *"(...) representative of the current decade. Higher precipitations are simulated by INT over Europe and North Africa, but the spatial patterns are similar between the two simulations. (...)"*

Referee: page 2096, line 12: Fig. 2 displays the difference of ANALY-INT, but in Sect. 3.1.2 INT-CLIM is investigated. I would expect that the temperature differences from ANALY-INT and INT-CLIM differ.

The differencies between ANALY, INT and CLIM are used to separate the effects due to the meteorological forcings and surface exchange fluxes. The meteorological parameters have an influence on the atmospheric distribution of species: on surface processes as deposition velocity and on emissions (sea salt, biogenic volatile organic compounds, desert dust). In the atmosphere, meteorological forcings do not differ between INT and CLIM. The only differences are the species that depend upon meteorology.

Referee: page 2098, lines 22-24: From my point of view the annual amplitude of the ANALY simulation is overestimated as well and not "accurate".

Page 2098, from line 22, we modified the text: *"(...) The model-observation comparisons of NO_x presented in Fig.9b highlight a slightly overestimated annual amplitude of the concentrations for ANALY (with too high winter*

values and too low summer values) while the winter NO_x values simulated by CLIM are much more overestimated ($MBNO_xDM=8\mu g.m^{-3}$). (...) "

Referee: page 2100, line 27: "vertical resolution" of what? The models should use the same resolution, if I understood correctly.

Page 2100, line 27, we modified the text: *"(...) as seen in Fig. 9 due to the differences in deposition velocities and in the concentrations in the lower atmosphere. (...) "*

Referee: Fig. 4: I understood from your model description that ANALY, INT and CLIM use the same horizontal and vertical resolution. Why do the pictures for ANALY have a much better resolution as for INT (e.g., the resolution of the mountain ridge)?

ANALY, INT and CLIM are computed with the same CTM setting, the same horizontal and vertical resolutions. But the meteorology forcings used have different resolutions: ARPEGE-Climate provides coarser resolution than ARPEGE for the model. For this reason, the pictures for ANALY exhibit better resolution than INT, as displayed by the mountain ridge (coming from the better description of surface pressure).

Reply to referee ♯1 technical corrections

Referee: Abstract: From my point of view SOMO35 is not a well known abbreviation, so it would be better to not used it without explanation in the abstract.
We removed the abreviation SOMO35 from the abstract.
Page 2084-2085, we modified the text: *"(...). We conclude that the indicators such as mean bias, mean normalized bias, RMSE and deviation standards can be used to interpret the results with some confidence as well as the health-related indicators such as the number of days of exceedance of regulatory thresholds.(...)"*

Referee: Sect. 2: As the chemical mechanism used here is the mixture of different existing reaction mechanisms and few additions, I suggest to give a list of the full reaction mechanism in the electronical supplement.
Done. We now provide a list of the reaction mechanism in the electronical supplement.

Referee: page 2095, line 17/18: "Concentrations ..." this sentence seems to be incomplete. Do you mean "High concentrations ..." or generally "SO2 leads to sulphate formation"?
We mean that SO_2 generally leads to sulphate formation.
Page 2095, line 17/18, we modified the sentence: *"(...). Once emitted in the atmosphere, SO_2 leads to the formation of sulphate aerosols. (...) "*

Referee: page 2096, line 17: Fig. 7b is referenced before Fig. 6. Rotate figures or include Fig. 7b into Fig. 5.
The figures Fig.6 and Fig.7 are rotated in the new version.

Referee: Sect. 3.1.2 and Fig. 7: Fig. 7 a,b,c,d,e are cited but no letters are given in the figure.
The letters are now given in the figure.

Referee: Sect. 3.1.2 and Fig. 7: the unit of the deposition flux in the text and the caption are given as μg.m^{-3}.s^{-1}. In the plot itself it is given as μg.m^{-2}.s^{-1}, which is more likely the unit of a flux.
The deposition flux is in μg.m^{-2}.s^{-1}.
Page 2096, line 25/26, we modified the text: *"(...). The mean deposition fluxes ($\mu g.m^{-2}.s^{-1}$) of O3, NOx and SO2 have been computed for the summertime period. (...) "*
Fig.7, we modified the caption: *"(...). Deposition flux are in $\mu g.m^{-2}.s^{-1}$ (...)."*

Referee: page 2097, line 12: "In summary, the comparisons between ANALY and CLIM (Fig. 8) have revealed ... " This sentence is really confusing as Fig. 8 was not mentioned before. Please rewrite the sentence, e.g., "In summary, Fig. 8 providing a comparisons between ANALY and CLIM reveals ... "
Page 2097, line 12/13, we modified the sentence: *"(...). In summary, the comparisons between ANALY and CLIM represented in Fig.8 have revealed the contribution of both meteorological and flux changes on simulated air pollutants. (...)"*

Referee: page 2098, line 16/17: "In Table 6 are summarized the statistics ..." ! "Table 6 summarizes the statistics ..."
Done

Referee: page 2098, line 20: the minus sign before 4.6 is missing.
Done

Referee: Sect. 3.2.1: It would be good to state somewhere that the Tables providing the error statistics are discussed in detail in the next subsection.
A paragraph has been added before section 3.2.1 to describe the subsections. *"First, in Sect. 3.2.1, we discuss the interannual variability simulated by the model. The statistical records are then described in detail in Sect. 3.2.2. To finish, Sect. 3.2.3 evaluates the impacts of chronology of pollution events on the skill scores. "*

Referee: page 2099, line 18: add "(Table 10)" behind bias.
Done

Referee: page 2099, line 23: Fig. 9 and Table 9
Done

Referee: page 2101, line 24: Figure 11c ! Figure 11b
Done

Referee: Table 1: 6 yr (? , the text says 5 yrs); 2003-2008 (? , the text says 2004-2008)
The simulations were performed over 6 years including 2003. However as explained in the paper (section 2.1), we did not consider year 2003 in the statistical comparisons for clarity and the results for this year are not presented in the paper. For clarity, we indicated that simulations are performed for 5 years.
We modified the abstract, line 4: *"(...) using three 5 yr simulations (...)*
Page 2087, line 7: *"(...) the present time (2004-2008) over (...)"*
Page 2089, line 19: *"(...) Three five year (...)"*
Table1: *"2004-2008", "5 yr of 2000-2010 climate", "5 yr of 2000-2010 climate "*

Referee: Table 3, caption: et ! and
Done

Referee: Table 5: for PM10 classes 1-2 (text says 1-5)
Done

Referee: Table 9: DM is missing in all entries in the left column.
Done. DM has been added.

Referee: Fig. 2: everywhere "PBL" is used, change the annotations in the figure accordingly from BHL to PBL.

Done

Referee: Figs. 2,3,6,9,10,12: Figures are much too small. I had to enlarge them on the screen by more than a factor of 3. This should be readable without zoom in the final version.
The size of figures have been enlarged to fit as much as possible. In our opinion, it is beneficial to keep related figures on the same page in order to allow the reader to see a quick comparison of the results. PDF publications allow also to zoom in if the readers want to see the details.

Referee: Fig. 6: a,b,c,d, are far too small.
The letters a,b,c,d are enlarged in the new version.

Referee: Fig. 6: "For INT, daytime and nighttime mean deposition velocities reach 0.57 cm/s and 0.24 cm/s, respectively over land (0.06 cm/s and 0.05 cm/s over sea). For CLIM, day-time and nighttime mean deposition velocities reach 0.54 cm/s and 0.24 cm/s over land (0.05 cm s1 and 0.04 cm s1 over sea)." This information does not belong into a caption.
Fig.6, we removed this informations from the caption.
Page 2096, from line 21, we modified the paragraph: *"(...). For INT, daytime and nightime mean deposition velocities reach 0.57 cm.s^{-1} and 0.24 cm.s^{-1}, respectively over land (0.06 cm.s^{-1} and 0.05 cm.s^{-1} over sea). For CLIM, daytime and nightime mean deposition velocities reach 0.54 cm.s^{-1} and 0.24 cm.s^{-1} over land (0.05 cm^{-1} and 0.04 cm.s^{-1} over sea). Over land, similar deposition velocities are thus calculated in INT and CLIM. (...)"*

Referee: Fig. 6+7: showing the sulphate difference in Fig. 7, but no sulphate in Fig. 6 is inconsistent.
As suggested by the referee, we added the mean deposition fluxes of sulphate. In order to be consistent with Fig.7, we also added the deposition fluxes of isoprene in Fig.5 (in the new version).
In the new version, we modified the figures as follow:
• Fig.5 (represented by Figure.1 here), we modified the caption: *"a)Emissions of isoprene for the summertime period, averaged for 2004–2008 in the INT (left) and CLIM (middle) simulations. Differences between INT and CLIM are shown on the right figure. b) Deposition flux ($\mu g.m^{-2}.s^{-1}$) of isoprene, averaged for the summertime of INT and CLIM simulations."*
• Fig.6 now displays O_3 deposition velocity. We modified the caption: *"O_3 deposition velocity averaged for the summertime period simulated by INT (left) and CLIM (middle). Differences between INT and CLIM are shown on the right."*

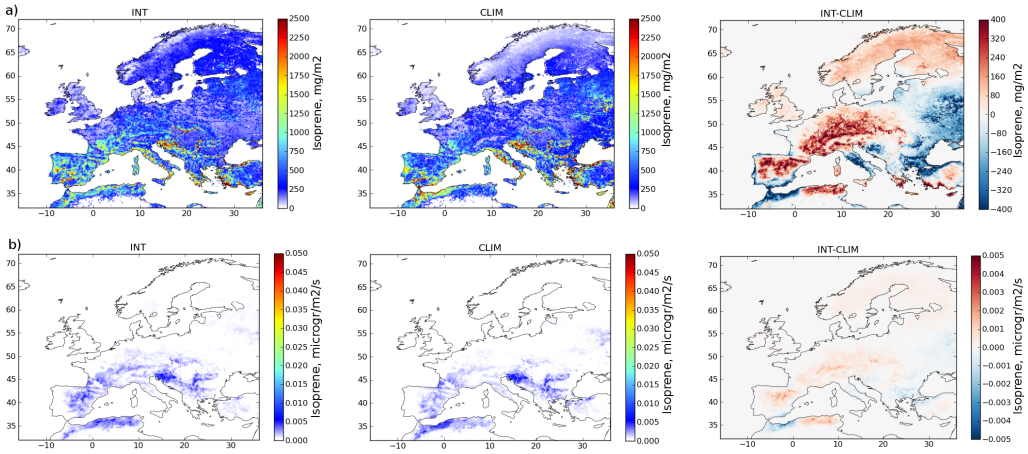

Figure 1: **(a)** Emissions of isoprene for the summertime period, averaged for 2004–2008 in the INT (left) and CLIM (middle) simulations. Differences between INT and CLIM are shown on the right figure. **(b)** Deposition flux (μg.m^{-2}.s^{-1}) of isoprene, averaged for the summertime of INT and CLIM simulations.

• A new figure (Fig.7 in the new version and represented by Figure.2 here) shows the deposition fluxes of O_3, NO_x, SO_2 and sulphate. The caption is: *"From top to bottom: (a) deposition flux of O_3, (b) deposition flux of NO_x, (c) deposition flux of SO_2 and (d) deposition flux of sulphate. Deposition flux are in $\mu g.m^{-2}.s^{-1}$ and averaged for the summertime period of INT and CLIM simulations."*

Referee: Fig. 9, caption last lines: Please state in order, what is displayed in 1) and what in 2).
Fig.9, we modified the caption: *"1) Simulated (ANALY: black lines; CLIM: gray lines) and measured at the AirBase stations (red lines) time series of monthly mean concentrations of O_3 **(a)**, NO_x **(b)**, SO_2 **(c)** and PM_{10} **(d)**. The time series are plotted from 1 January 2004 to 31 December 2008 and averaged over the European domain. Concentrations are in $\mu g.m^{-3}$. 2) Anomalies calculated when substracting the average annual series from the time series in 1)."*

Referee: Fig. 12: The cross for ANALY is too small.
The cross for ANALY has been enlarged.

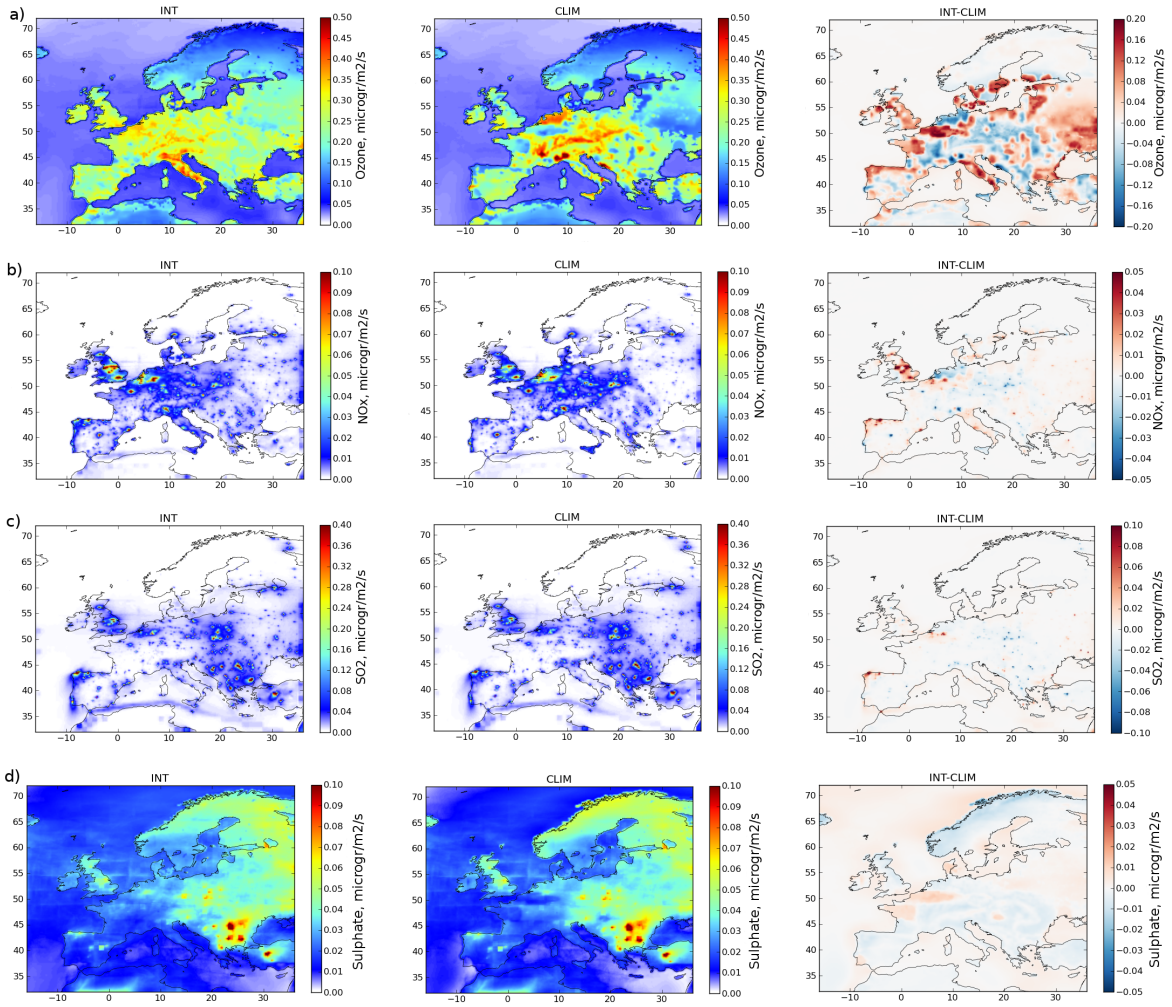

Figure 2: From top to bottom: **(a)** deposition flux of O_3, **(b)** deposition flux of NO_x, **(c)** deposition flux of SO_2 and **(d)** deposition flux of sulphate. Deposition flux are in μg.m^{-2}.s^{-1} and averaged for the summertime of INT and CLIM simulations.

Reply to referee ♯2 comments

Referee: Introduction: The authors refer to previous relevant articles but they do not clarify if the results of these studies are related purely to climate change or emissions change. So the introduction needs practically to be put under a more thorough structure and perspective. Also, there are a number of recent relevant publications which are not referred at all. See for example (Andersson Engardt, 2010, JGR, doi: 1029/2008JD011690; Huszar et al., Climate Research, 2012, doi: 10.3354/cr01036; Katragou et al., JGR, 2011, doi:10.1029/2011JD015899; Juda Rezzler et al., Climate Research, 2012). There is also a recent evaluation study by Zanis et al., Atmospheric Environment, 2011, doi:10.1016/j.atmosenv.2011.09 with a similar concept to that of this paper comparing long-term air quality simulations forced with reanalysis or GCM fields. An update of the literature with relevant work for Europe is needed.

As the referee suggested, the paragraph has been organised, with an update of the literature. The question of interactions between a changing climate, changing emissions is indeed a very topical and active field and it is a challenge to highlight "all "relevant results outside of a review paper. Several references have been added .
We modified the introduction : *"(...) The interactions between climate change and air quality have been already extensively studied. At the global scale, studies [Prather et al., 2003; Dentener et al., 2006] have for instance evaluated the effects of changing emissions and climate on surface O_3 concentrations under an A2 scenario (IPCC AR4). Dentener et al. [2006] showed that global mean surface O_3 may increase by about 4.3 ± 2.2 ppbv by the year 2030 and the area of global natural ecosystems exposed to critical nitrogen deposition may increase up to 25 % by this time. Regional models centered over the continental United States have been used to examine US air quality in the future due to climate change alone independently of evolution in emissions in North America and elsewhere [Hogrefe et al., 2004; Knowlton et al., 2004; Dawson et al., 2009]. Hogrefe et al. [2004] concluded that the average daily summertime maximum 8-h O_3 concentrations will increase by 2.7 ppbv and 4.2 ppbv for summers in the 2020 s and 2050 s, respectively. In the literature, a set of regional models have similarly focused on the European region to isolate the impacts of climate change [Langner et al., 2005; Meleux et al., 2007; Giorgi and Meleux, 2007; Carvalho et al., 2010; Andersson and Engardt, 2010; Katragkou et al., 2011; Huszar et al., 2012; Juda-Rezler et al., 2012].*

Precisely, Zlatev [2007] and Langner et al. [2005] presented the impacts of climate change on air quality over Europe with a constant emission rate and showed an increase in photochemical production in future climate scenarios. In Meleux et al. [2007], the authors isolated the impacts of summer European climate change on the increase in O_3 levels by using the same emissions and global chemical boundary conditions for the present day and future periods. Katragkou et al. [2011] investigated the sensitivity of surface ozone to the future climates of the 2040s and 2090s by studying changes in meteorological parameters under an A1B scenario. Andersson and Engardt [2010] suggested changes in surface ozone between -4 to 13 ppbv on average from 1961-1990 to 2071-2100, based on the A2 scenario and highlighted the role of surface deposition processes. Carvalho et al. [2010] concluded that PM_{10} levels will be impacted by climate change depending on the month and region, with a maximum increase reaching 30 $\mu g.m^{-3}$ in September over Portugal. Szopa et al. [2006] investigated impacts of local anthropogenic emission changes and background O_3 changes. They estimated that the O_3 concentration in July may increase up to 5 ppbv across Europe by 2030. (...)"

Referee: The authors claim that they want to study the impact of different climate forcing on air quality. In the 1st experiment (ANALY), the chemistry transport model MOCAGE is driven by reanalysis and then by ARPEGE climatology (INT). The comparison of these 2 runs would have provided an idea of how the hindcast (driven by reanalysis) can be different from the control (driven by the climate model ARPEGE) in case the model set up had been the same. However, if I understand correctly, different resolutions are used for the two models configuration. Thus, the differences between ANALY and INT simulations do not show only the impact of reanalysis vs control, but also the differences of a coarse vs fine resolution meteorological forcing on air quality.

Indeed, the meteorological forcings we have used differ in resolution: ARPEGE-Climate outputs are in a coarser resolution than ARPEGE. However, the configuration is the same in both simulations. It is true that comparisons between ANALY and CLIM show impacts of climate forcings (no assimilation of observations) and also coarser meteorology.

Referee: 2003 is mentioned to be omitted from the analysis as it is considered to be a climatic anomaly. This needs to be better justified. Climate extremes are part of the present-, and even more importantly for the future-climate, thus potential climate-air qual-

ity models should be assessed for their potential to capture such cases, as well.

We agree with the referee that climate air quality models should be able to capture extreme events. However, in future climate, 2003 is certainly a climatic anomaly. The purpose of this paper is to assess the use of a climate model to represent air quality by comparing simulations statistically averaged over a period of time, too short to be not too much affected by such events. The results for year 2003 (in ANALY) are clearly unexpected in CLIM. By taking into account 2003, we would have added biases in the comparisons and statistical results would have been much more difficult to interpret.

Referee: Page 2088, line 21: Organic and Nitrate aerosols are not taken into account, which is an important drawback for the assessment of PM.
We agree with the referee when he states that it is a drawback that organic and nitrate aerosols are not taken into account. Futher developments in MOCAGE concerning these aerosols will be assessed to improve the negative biases in total PM.

Referee: Page 2094, lines 15-26: The authors claim that temperature change is the most important factor that determines the ozone change. In what physical sense? Due to temperature dependence in reaction rates? I think this is an oversimplification. The authors should consider that anticyclonic anomalies lead to positive temperature anomalies but at the same time also lead to positive anomalies in incoming solar radiation and more stagnant conditions with longer residence times of air masses over certain areas which favour ozone production. In general, circulation changes is an important factor that is not referred at all.

The change in temperature is one of the most important factors to explain ozone change as described in Dawson et al. [2007]. Increases in temperature imply higher biogenic emissions and faster chemical reactions, which cause higher pollutant concentrations, as ozone. Among the meteorological parameters, changes in solar radiation, humidity, cloudiness, atmospheric circulation are also considered important.
Page 2094, from line 15, we have slightly modified the sentence to reflect this: *"(...). Nevertheless, as explained in Katragkou et al. (2010), other variables such as differences in solar radiation, zonal and meridional winds and changes in atmospheric stability also impact ozone concentrations. (...)"*

Referee: Page 2096, lines 3-4: The authors state that for all pollutants the differences are primarily due to PBL height differences. This process is more important for primary pollutants while for secondary pollutants the situation is more complex. Furthermore, especially for ozone which has also a source from above the situation is even more complex.

This sentence is in fact a conclusion of our results. As expected, different PBL mixing heights have an impact on primary pollutants but also on secondary pollutants. We agree that in general, species like ozone, are affected by many more processes.
Page 2096, line 4, we have slightly modified the sentence: *"(...) For all pollutants, primary as well as secondary, (...) "*

Referee: Page 2097, lines 20-21: According to the authors, the changes in isoprene are mainly attributed to dynamical processes of the Boundary layer. Following the Guenther approach to calculate biogenic emissions it is sensible that changes in temperature and solar radiation are at least as important.

Biogenic emissions are indeed related to changes in meteorology, such as surface temperatures and solar radiation [Guenther et al., 2006], as stated in the paper. The differences between ANALY and INT are explained only by differing meteorology, no differencies in the flux (this is a sensitivity study, as shown in Table.1, where the same primary emissions are used). The comparisons between INT and CLIM focus on the impacts of varying surface exchanges and the biogenic emission fluxes are different. With the two comparisons, we can understand the changes due to meteorology and surface emissions.
Page 2097, from line 20, we modified the sentence: *"(...) The major changes in isoprene concentrations (Spain, North Africa, Greece) are attributed to both changes in atmospheric circulation and stability (ANALY vs INT), as well as to differences in surface emissions and deposition (INT vs CLIM).*

Referee: The effect of the lateral and top boundary conditions is not discussed at all. Especially for ozone, these play a very important role (e.g. Stratosphere-Troposphere Transport, intercontinental transport etc). In general a paragraph should be added with the limitations of the modeling approach.

Due to the configuration of the model, two-way nested as explained in Section.2, these conditions (longe range transport, Stratosphere-Troposphere ex-

changes) are provided by the global domain of MOCAGE. This is a typical issue of regional air quality modeling.

Referee: Another important point is that the statistical results are presented but they are not discussed or explained in a physical sense.

We do not understand what is mean exactly by this remark. We believe that we have commented as much as possible the main outcome of the study.

Referee: The Section 3.2.3 is not clear to understand. Please provide more clarifications for the methodology used.

The section 3.2.3 is introduced by the last paragraph of section 3.2.2. In CLIM, the meteorological forcings are representative of the current decade: there is no particular match in the sequence of years and the representativeness of the skill scores can be assessed by permutations of all years. By doing the same for ANALY, the comparisons will allow us to determine which statistical tools are useful to consider for future studies. The way to do this is explained in the beginning of Sect. 3.2.3. We have modified the comparisons between ANALY and CLIM by making arbitrary permutations of the years in order to:
• for CLIM: no particular sequence of years is more possible than another one. This is only relevant for the statistical comparisons.
• for ANALY: by using permutations, we evaluated the performances in a statistical way more comparable with CLIM.

Reply to referee ♯2 technical comments

Referee: The abstract needs to be revised as it is too generic. It should focus more on the basic findings of the work performed.

We believe that the abstract focus on the main findings provided by this work.

Referee: In the abstract are mentioned 6 year of simulations while in the manuscript the time slice 2004-2008 appears to be selected. The inconsistency needs to be corrected.

We have not shown the results of year 2003 for the reasons explained in the paper. For clarity, we have modified the abstract and the table to reflect what is actually used. (The modifications have been previously explained.)

Referee: 12 figures and 12 Tables for a single paper may be a little bit more than a reader can follow. I tend to think that some filtering will be necessary. The description of the presented material could be more concise and more focus could be given on the analysis of physics and chemistry, their linkages and the implications.

We believe it is a criterion of GMD to accept large results for publication. We also believe that these results are useful for other modellers.

Referee: The writing (language) does not always meet the required standards. Several paragraphs need to be more carefully rewritten.

One of the co-author is a native english speaker who has carefully removed the english grammar errors.

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