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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Radiative forcing estimates in coupled climate-chemistry models with emphasis on the role of the temporal variability" *by* C. Déandreis et al.

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We thank the two referees for their constructive remarks that enable improvement of the quality and the clarity of the paper. A new version of the manuscript is proposed in supplement with corrections highlighted in red.

Response to the referee 1 comments

Response to the general comments: 1- The treatments of aerosols and aerosol-cloud interactions in this model are not as sophisticate as it should be, which may affect some conclusions of the current study. For example, only sulfate aerosols are included in the INCA model, while in most next generation of global aerosol-climate models, sulfate

aerosols are treated as internally mixed with other primary aerosol species. It is also not clear whether sulfate aerosol size distributions are predicted in the INCA model, but aerosol size distribution are predicted in most next generation aerosol-climate models. The activation parameterization used in this model is an empirical formula, and activated droplet number concentrations depend only on sulfate aerosol mass, but not on sulfate size distributions and on subgrid vertical motion from turbulence. The simple treatment in this study may affect some of their conclusions. For example, if sulfate aerosol mass and number concentrations in both the Aitken and accumulation modes are predicted in the model and used in the activation parameterization, the difference in the shortwave fluxes at the top of the atmosphere between the "MONTH" and "VAR" simulations (section 3.3.3) can be smaller, as the non-linear dependence of activated droplet number concentrations on the accumulation mode sulfate number concentrations should be smaller than that on total sulfate mass.

Response: The central objective of this paper is to analyse the radiative impact when the aerosols temporal variability is introduced in climate simulation by coupling the radiative code of a GCM with a chemistry-aerosol module. To simplify the analysis and to avoid the noise due to other effects, we decided to use a simple aerosol treatment. We limited the number of aerosol species to sulfate only and the activation parameterization is based upon an empirical formula. Though empirical relationships present several weaknesses (not treating explicitly the microphysical process and of the variability of the CNDC/Aerosol numnber relationship in different regions and for different aerosols types), prognostic activation schemes present also disadvantages like the difficulty to calculate a realistic cloud parcel updraft velocity in GCM (Pringle et al., 2009). Finally, we think that today the choice of the level of complexity to treat aerosol indirect effect in climate simulation is still an open question and empirical schemes are still widely used in climate modelling studies (IPCC AR4). To bring the attention of the reader to these limitations in our study, we highlight the simple aerosol treatment that is made in the introduction and in the conclusion of the paper

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2- As the aerosol-cloud interaction in this model is treated in a simple way (no cloud life time effects, and droplet number concentrations are directly diagnosed from sulphate aerosol mass), the approach explored in this study is only useful to their particular model and is not applicable to most next generation aerosol-cloud models that solve a prognostic equation for cloud droplet number concentrations, and accounts for other aerosol effects on climate, such as cloud lifetime effects and aerosol effects on ice clouds. Even if you can get the instantaneous PI aerosol field in a PD simulation, it is still not clear how you can calculate the needed PI droplet number concentrations online in a PD simulation for those next generation aerosol-cloud models. As aerosols and clouds are closely coupled in those next generation aerosol-climate models, it is not practical to estimate the first indirect effects online in my opinion.

Response: As stated in response to the general comment 1, we decide to use a simple aerosol representation. Our methodology doesn't apply to models using prognostic activation schemes or to effects that impact directly meteorological fields like cloud lifetime effect or semi-direct effect. The treatment of such effects is beyond the scope of this study. Here, we focus on the direct and first indirect effect.

Response to the specific comments:

1- Title: I would suggest to add "of sulfate aerosol' after "Radiative forcing estimates', as the paper only focus on the estimate of radiative forcing of sulfate aerosols.

Response: Following the 2 reviewers recommendation, we added the term "sulfate" in the title. The title is now: "Radiative forcing estimates of sulfate aerosol in coupled climate-chemistry models with emphasis on the role of the temporal variability"

2- p. 24314, line 10-11: these modifications are not large, as it is 0.20 W m-2 out of 240 Wm-2. It is not fair to compare this value to the radiative forcing of sulfate aerosols, as the radiative forcing is the difference between PD and PI simulations. The estimates of the radiative forcing are quite similar for both approaches (see Section 3.1 and 3.2).

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Response: We modified the abstract as follow: “Sulfate temporal variability causes a modification of the short wave net fluxes at the top of the atmosphere of +0.24 and +0.22 W.m-2 for respectively the present and preindustrial periods. This change is small compared to the value of the net flux at the top of the atmosphere (about 240W.m-2). However, it corresponds to 10% of the total anthropogenic radiative forcing for the 20th century. The effect is more important in regions with low-level clouds and intermediate sulfate aerosol concentrations (from 0.1 to 0.8 μg(SO4)m-3 in our model).”

3- p. 24314, lines 16-19: I think the reason is not because of the meteorological trajectories used, but is because of the monthly mean PI aerosol fields are used in the second radiation call (section 4.1). The difference between the ‘extended off-line’ method and the offline method is 0.22 W/m2, which is the close to the difference in the shortwave fluxes at the top of atmosphere between the “MONHT” and “VAR” examined in section 3.3.2. This suggests that the use of monthly vs. instantaneous aerosol fields is the reason.

Response: An error in the original text p24330, line 8 brought this intrepretation. We corrected this sentence, changing “section 3.2” by “section 3.1 (on-line method)”. For the reference method, the on-line approach (interactive calculation of sulfate fields) is applied to both present and preindustrial emissions fields. Both simulations are performed with the same meteorological conditions based on present-day fully variable sulfate aerosols. This error has been corrected several times in the text, improving the explanation of the “off-line extended method” results.

In addition, a sensitivity test has been done (not shown in the paper) to define the reasons for the results of the extended off-line method. This test consisted in performing the off-line extended method reading preindustrial concentration fields every 30 minutes instead of using the monthly data. Results were very similar to those obtained with the original extended off-line methods. This test confirms that the meteorological trajectories explain the results obtained in the off-line extended method. We preferred not to include this test in the paper as we thought it might add complexity.

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4- p. 24318, line 28: the formula of re. As the relationship between effective radius and volume-mean radius are different over ocean and over land, it will be better to use different formulas over land and over ocean (Martin et al., 1994).

Response: These different relationships from Martin et al. (1994 have not yet been taken into account in the LMDZ model.We might include them in the near future to improve the representation of the 1st indirect effect. Nonetheless, this addition is of secondary importance for this paper as its focus is not on the land/ocean contrasts.

5- p. 24320, section 2.2.2: how is the size distribution of sulfate calculated? Does this model include any new particle formation from aerosol nucleation? How about other aerosol species, such as dust, sea salt? These primary aerosols can affect size distributions, and can affect droplet number concentration and further affect the estimate of the indirect effect of sulfate aerosol.

The chemical transformation of the gaseous sulphur species requires oxidants either in the gas-phase or in the liquid-phase. The sulfur chemistry implemented in INCA is similar to the one described in Boucher et al. [2002]. The oxidant fields are estimated in INCA as part of the dynamic chemistry scheme. DMS and its product DMSO are oxidised using the actual concentrations of OH and NO3. SO2 is transformed to sulfate by H2O2 and O3 in cloud liquid water. The formation of sulfate is limited by the acidity formed in the oxidation process within cloud droplets. SO2 is also oxidised in the gas-phase. Gaseous H2S and aerosol methane sulphonic acid (MSA) are also included as minor species of the sulphur cycle. The mass of sulfate produced through these reactions is directly injected into the soluble accumulation mode. The nucleation mode is not treated explicitly, hence, there is no new particle formation in this size range and the very fine particles are not represented. The mass median diameter (MMD) of sulfate depends upon the mixing of the ratio of sulfate formation from clouds and via gas phase oxidation. Furthermore, the MMD varies as deposition takes place since large particles will be preferentially scavenged by sedimentation and below cloud scavenging. . We have added this description to the paper in line 22 p24320.

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6- p. 24324, section 3.3.1: It is not clear to me how the authors keep the meteorological fields exactly the same in the four runs (two MONTH simulations, and two VAR simulations). Does this mean the model run in an offline model, without aerosol effects on meteorological fields?

Response: To clafity this point, we modified the text p 24324. Following the proposition of Referee 2, lines 20-21 were replaced with: "The MONTH simulation is run off-line with respect to the meteorological trajectory of the VAR simulation (temperature, pressure, wind, humidity, cloud fraction, LW path,...)." We replaced lines 26-27 with "Preindustrial simulations have also been performed off-line with the meteorology based on the present-day fully variable sulfate aerosol retroaction." We also corrected the reference to a section on line 20 "Sect. 3.2" has been replaced by "Sect. 3.1".

7- p. 24325, lines 24-28, and p. 24326, lines 1-2: As I mentioned above, this results can be model-dependent. If aerosol number concentrations in both the Aitken and accumulation modes are simulated, the non-linear dependence between droplet number concentrations and aerosol fields can be smaller.

Response: As stated previously, our results are limited to model that calculate cloud droplet number from an empirical relation. In the INCA model, the mass of sulfate formed participates to the aerosol accumulation mode, and we do not treat the Aitken mode. Since the aerosol mass in the Aitken mode is considerably samller that in the accumlation mode, the contribution of the Aitken mode to the relations based on aerosol mass is much less important.

8- p. 24326-24327, section 3.3.3: Again, many discussions in this section regarding the role of sulfate concentrations are likely to be model-dependent too.

Response: see response to specific comment n°7.

9- p. 24329, lines 14-22: I think the reason is not because of the meterological trajectories used, but is because of the monthly mean PI aerosol fields are used in the second

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radiation call .The difference between the 'extended off-line' method and the offline method is 0.22 W/m2, which is the close to the difference in the shortwave fluxes at the top of atmosphere between the "MONHT" and "VAR" examined in section 3.3.2. This suggests that the use of monthly vs. instantaneous aerosol fields is the main reason.

Response: We explain in specific comment n°3 that the confusion was brought by an error in the text that we have corrected.

10- p. 24330, Section 4.2: Is this reference method the same as the off-line approach in Section 3.2, but it reads aerosol fields every half hour instead of using the monthly data?

Response: This has been now corrected as stated in specific comment n°3.

11- p. 24332, lines 1-2: here the authors provide the correct explanation why the 'extended off-line method' gives a quite different estimate compared to the offline method. The explanation in the abstract (p. 24313, lines 16-17) and in section 4.1 (p. 24329, line 14-22) are not accurate.

Response: This confusion has been taken out as we did the corrections indicated in specific comment n°3.

12- p. 24333, section 4.4: in this approach, you will have to finish PD and PI simulations first to calculate "preindustrial aerosol fraction", and then rerun the PD simulations.

Response: Section 4 aims to explore different methods, their advantages and disadvantages. A disadvantage in method detailed in section 4.4 consists in the need to run a chemistry-transport model to get PI and PD concentration fields. The same is true for the off-line method.

13- Fig. 1: In the new approach (solid), cloud droplet number concentrations are quite low even at very high sulfate concentration. Is simulated droplet number concentrations compared with observations?

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Response: CDNC variations play an important role in the computation of the first indirect effect radiative forcing. Comparison of several GCMs, including LMDz ,with observations from satellites are the cornerstone of the Quaas et al. (2009) paper. Figures 2a and 3 from this paper show that CDNC variation relative to aerosol optical depth perturbation is quite well captured by LMDZ-INCA with respect to satellite observations.

14- Fig. 4: Why is the 1st indirect forcing positive over some regions?

Response: It is due to the decrease of SO2 emission over these regions due to a reduction of biomass burning (see figure displayed in response to the detailed comment of Referee 2). This information has been added in the text p 24323 line 25.

Response to the referee 2 comments

Response to the general comments:

1- General comments The paper give a good overview of the possible variations caused by off-line and on-line calculations and the effect of temporal variability. It goes through a number of methods on calculating radiative forcings, and a thorough documentation and validation of the method. As expected there are some effects of temporal variation of aerosols, although surprisingly small, despite the focus on this. -0.2Wout of 200 is a number well below the uncertainty threshold. It should of course be taken into account however and the metoods may be taken into account when calculating the effect of aerosols on climate in general, not only the effect on changes in aerosols. Actually, although to some extent shown in other papers, finding that PD-PI aerosol forcing is not very dependent temporal variability is just as interesting and important than the temporal variability in total aerosol forcing. If this is the case also for a more complex aerosol, this will simplify most forcing calculations.

What I do not like however is that the use of word "radiative forcing" is sometimes ill-defined. Sometimes it is taken to be present day - pre-industrial. With regard to temporal variability it is taken to be the difference between variability and monthly mean

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values. I think it is especially problematic in the description of the initial extension method (4.1) that give a larger deviation from PD-PI forcing than any of the other. Online -0.73 W/2 ; Off-line -0.70, Off-line monthly means -0.64 and then the extended method (before the improvement is -0.49).

Response: We modify section 4.1 as follow: p24328 lines 22-23: "The modification of the direct effect due to the change of aerosol concentration between present-day and preindustrial conditions results in a difference of net fluxes at the top of the atmosphere that amounts to –0.32 Wm-2. It is very close to the value of the direct radiative forcing obtained with the off-line method (-0.31 Wm-2, Sect 3.2). Computing sulfates interactively increases the difference of net fluxes at the top of the atmosphere by 3% relative to the off-line method when these concentrations are read in. The geographical distribution of these two fields is also very similar (Fig. 10a and 10c). Industrial regions where are located SO2 emissions sources are more sensitive to the temporal variability of aerosol. . . ."

p24329 lines 4-5: "The modification of the first indirect effect due to the change of aerosol concentration between present-day and preinsustrial conditions results in a difference of net fluxes at the top of the atmosphere estimated at -0.27 Wm-2. This value is much lower (a 60% decrease in absolute value) than the radiative forcing obtained with the off-line method (-0.39 Wm-2, Sect 3.2). The geographical difference of these two fields displays . . ."

2- Also I think that the authors should consider making a test or at least discuss whether they expect their conclusion to be different in case of a more complex aerosol representation, in particular in the presence of an absorbing aerosol.

Response: For model using an empirical relationship to calculate CDNCs, the presence of an absorbing aerosol would not have a strong effect. In this case, we would have used the adjusted parameters proposed by Quaas and Boucher (2005) for the relation number of CDNCs=f(aerosol mass) when the sum of the masses of all hy-

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drophilic aerosols (sulfate, the hydrophilic fractions of black and organic carbon and submicrometer sea-salt) is taken into account. This comment has been added in the conclusion.

Response to the details: 1- Title: The title is too general, taken into considerations that all variations in forcing is connected to sulphate. Suggest that "of sulphate" should be added after radiative forcing

Response: We added the term “sulfate” in the title. The new title now is: “Radiative forcing estimates of sulfate aerosol in coupled climate-chemistry models with emphasis on the role of the temporal variability”

2- p 24314 line 17. "than that" not needed ?

Response: This correction was included in the text

3- p 24314 It is unclear to me what the 60 % refer to.

Response: The abstract has been modified and the sentence has been rewritten: ‘If this condition is not satisfied, it introduces a 60% error on the estimation of the first indirect radiative forcing’.

4- p 24315 line 9. " It causes a surface cooling" This is generally not true on a local / regional scale. The sentence should be replaced by negative radiative forcing on top of the atmosphere. line 22 estimation –> estimate ? line 26 "few days" delete few ? A few days is a relatively long life-times for many aerosols.

Response: The corresponding editions have been included into the text

5- p24316 , line 2-3 I do not understand the first part of this sentence their large size âĂŤ> large differences in size ?

Response: This sentence has been re-written. It is now: ‘High temporal variability of aerosol is induced by their large difference in size and the many processes that affect their properties while they are in the atmosphere (nucleation, coagulation, sedimenta-

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tion, wet deposition, humidity growth. . .).'

6- p 24316 line 8-10, Possibly true for sulphate, but unsubstantiated for other type of aerosols, absorbing aerosols, coarse aerosols?

Response: What we meant here is that in the case of absorbing aerosols, the semi-direct effect will cause strong non-linearities.

7- p 24316 line 26-27. I do not understand this sentence. Many models has calculated off-line aerosol effects, e.g. IPCC 2007, page 162, or is the point that few or none? has done a off-line model for a long model simulation, e.g. 20th century run.

Response: This sentence has been replaced with: "But, to our knowledge, none of these studies include on-line radiative forcing calculation taking into account aerosol temporal variability."

8- p 24317, line 26 resolved –> resolve ?

Response: We corrected this sentence.

9- p 24319 line 8-10. The high end of the Polder numbers are quite small compared to a number of other sources, e.g. Hegg et al. ACPD 11, 28663–28687, 2011, which give a CDNC of 300-400 in clean maritime regions.

We agree with the reviewer that Polder numbers are on the low end. We would like to point out that the Figure 3 from Quaas et al., (2009) indicates that CDNC number sensitivity to aerosol optical depth perturbation is well captured by LMDZ-INCA with regards to the observations and compared to other GCMs. This reference has been added.

10- p 24320 line 19 Is size distribution taken into account for calculation of direct effect?

Response: Yes, the aerosol optical parameters are calculated taking into account the size distribution of the aerosols. We introduce this point in the paper p24318 - line 19.

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11- p 24321 line 18. Full stop before "They .."

Response: It has been corrected in the text

12- p 24323 line 2-11 The use of the word "higher" confuses me "Fm_pd is higher than Fvpd" From figure 2 Fm_pd = 240.57 Fvpd = 240.72 At the same time "DF_M is 12% higher than DFv" DF_M = -0.64, DFV = -0.73. DF_M is less negative yes, but the aerosol effect is higher in the on-line simulation.

Response: These sentences have been modified accordingly.

13- p 24323 / Figure 2: As far as I can understand the on-line radiative perturbation is closer to the calculated off-line aerosol effect than the off-line perturbation. Does not this mean that you get almost identical on-line and off-line forcing perturbation. (0.73 – 0.70) ?

Response: Yes, the on-line radiative perturbation (-0.73Wm-2) is closer to the off-line total radiative forcing (-0.70Wm-2) than the off-line perturbation (-0.64Wm-2). The difference between the off-line and on-line radiative perturbation is up to 10%.

14- p 24323 line 25 -> / figure 4 Is it possible that the areas of positive in-direct effect is caused by a decrease in so2 emissions numbers from PI to PD in these regions. Check e.g. the aerocom sets for so2 from biomass burning. If that is the case you can mention this?

Your hypothesis is confirmed by the figure 1 (in attachment). It displays the difference of SO2 emitted by biomass burning (in 106kg/year) between present and pre-industrial periods. In the two regions of positive radiative forcing, we observe a decrease of this source of SO2 emissions. We thank the reviewer for her/his suggestion and have added a comment in section 3.2.

15- p 24324 / Figure 5. It is a bit unclear to me how the meteorological trajectory is included. Based on figure 5 I assume that all 4 simulations is based on present-day fully variable sulphate aerosols, and then all other results is calculated off-line wrt to this

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meteorology? I suggest that you specify this also in the text and remove the sentence about using the same physical forcings etc, since this looks like you have stored and read in some forcing data into the model?

Response: We modify the text p 24324 to clarify this point. Following the reviewer's proposition, we replaced line 20-21 by: "The MONTH simulation is run off-line with respect to the meteorological trajectory of the VAR simulation (temperature, pressure, wind, humidity, cloud fraction, LW path,...). We modified lines 26-27 by "Preindustrial simulations have also been performed off-line with the meteorology based on the present-day fully variable sulfate aerosol retroaction." We also corrected an error line 20 "Sect. 3.2" has been replaced by "Sect. 3.1".

16- p 24327 line 10 et –> and

Response: It has been corrected.

17- p 24344 Table 2 Partly written in French

Response: This part is now written in English

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C15314/2012/acpd-11-C15314-2012-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 24313, 2011.

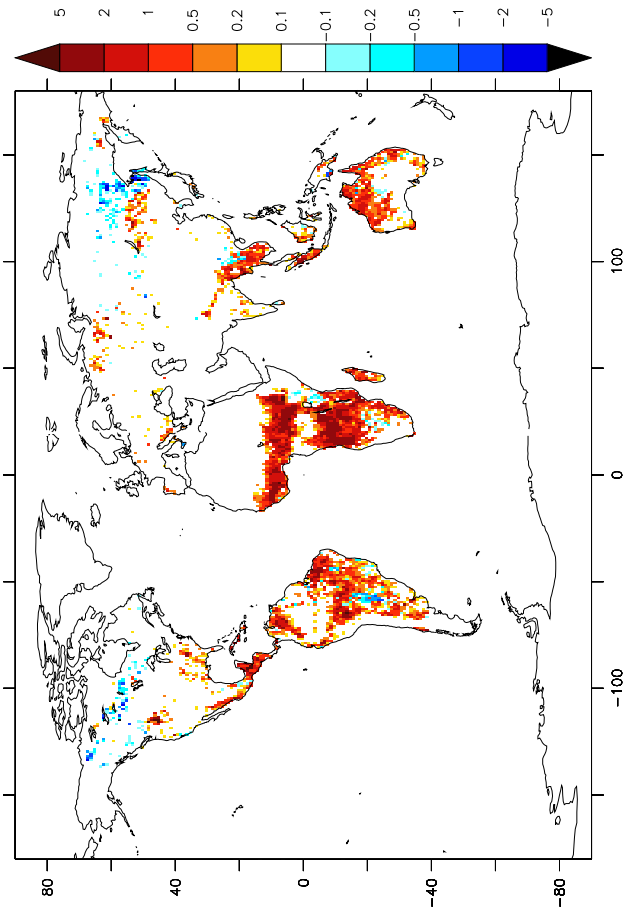

Fig. 1. difference of SO2 emitted by biomass burning (in 1E+06kg/year) between present and pre-industrial periods

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Interactive Comment