Atmos. Chem. Phys. Discuss., 11, C8391–C8398, 2011
www.atmos-chem-phys-discuss.net/11/C8391/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Singular vector decomposition for sensitivity analyses of tropospheric chemical scenarios" *by* N. Goris and H. Elbern

Anonymous Referee #2

Received and published: 31 August 2011

General comments:

This study uses singular vector decomposition to conduct a wide range of sensitivity analyses across various tropospheric chemistry scenarios. This study is set in the context of adaptive observations for air quality forecasting where sensitivity analyses identify model parameters leading to maximum error growth.

Overall, the manuscript is very well written, is well structured and does a very good job of explaining the various technical details. I have a series of comments regarding revisions that I believe should be made prior to publication in ACP.

I think further interpretation of the results is needed. Both for photochemical and mech-

anistic influences on the various results, and for what these results imply for adaptive observations or observing networks. In the case of the latter, the abstract second paragraph leads with "As a preparation for targeted observation calculations, the concept of adaptive observations is studied with a chemistry box model." This is an intriguing, relevant, and timely research topic. However, there is no discussion of how the results relate specifically to adaptive observations or to targeted observations, and instead the authors seem to lose track of this objective in the discussion of the results and in the conclusion. The authors should distil the various qualitative statements about each scenario and analysis into a series of statements and recommendations that relate directly to this topic, and make it relevant to real observing systems and forecasting problems if possible. As it stands, the authors have undertaken a very thorough and detailed series of sensitivity analyses using a new and little-used technique without substantively placing it in context. The authors do not conduct a significant investigation into the photochemical and mechanistic causes of their results. If a reader does not possess detailed knowledge of the chosen scenarios it is difficult to interpret the results. One area that could be expanded would be a discussion of the temporal evolution of the VOC and NOx sensitivities in the various scenarios. Why do certain scenarios tends towards either the NOx or VOC limited regimes with passing time? Various model outputs could be used to explain this aspect of the results.

The authors should relate the chosen scenarios to air quality forecasting over populated areas, i.e. PLUME, URBAN, BIO, and LAND. What do the results imply for observing systems in these environments?

The authors should justify the merits of performing this analysis in a box modelling context. For instance, I imagine it will be harder to isolate specific photochemical environments in the follow-up study using a chemical transport model. The authors should discuss the other advantages. Additionally, the authors need to evaluate the other possibilities for performing sensitivity analysis in both a CTM and a box model, i.e. brute force and adjoint. Although brute-force and adjoint sensitivity methods are feasible for

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a box model, are they feasible for a CTM given the specific aims of the study, and is the SVD method feasible in a CTM, and for the future in an operational system?

The relative and absolute error growth statistics need to be related to one another in a clearer way. Since the grouped absolute error growth statistics for NOx and VOC are presented in relative terms with respect to each other it is very difficult to relate the absolute and relative error statistics. I think if the authors addressed the point raised earlier in the general comments regarding the influence of photochemistry on the results it might go some way to resolving this issue. Indeed, the authors seem to be aware of the problem to some extent as they note that "Remarkably, there is no similarity between the grouped error growth (Sect. 4.1) and the grouped relative error growth." Another consideration is that the only model concentrations listed are initial concentrations (presumably at t0), but are the weightings for the relative statistics created from the tI concentrations? If so, it is rather hard to understand the difference between the relative and absolute statistics without the concentrations over the full course of the forward model runs.

Specific comments:

Page 16762, refs to table 2 and 3. No mention in either table is made of the water vapour concentration used in each scenario. This should be stated, as water vapour abundance plays a key role in differentiating the clean scenarios e.g. MARINE versus FREE. The FREE scenario should have a demonstrably longer ozone lifetime due to the lower water vapour concentrations in that environment.

Much of the discussion on page 16764 describes the various aspects of the TSVD plots and the analyses that comprise them. I found this section to be somewhat confusing. The TSVD figures and the figure 2 schematic loosely imply that the model start time is varied from 2nd July noon (for tI=t0 cases) through to 6th July noon (for tI=tn cases), but this is not stated clearly in the text, and in fact in a previous instance it was stated that all scenarios were started on July 1st (+24 hours of spinup to get to noon July

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2nd). These matters should be clarified in the text by the authors. I assume too that the model finish times used in the TVSD analysis vary from noon July 6th to July 10th for t0 → tn and tn → t2n, respectively. Additionally, the sentences regarding how tn and tf relate to one another in the sentence beginning "For the sake of clarity....." could be moved further up the in the discussion to aid the reader. The clarity may perhaps also be improved with a demonstration of what m means in figure 2 and how it can be used to calculate the simulation length.

Page 16766, lines 5-6. "The specific initial value at different day or night times does not seem to affect the results much." This is an imprecise statement. There is some variability according to tI for specific different day and night start times. For instance, all of the NOx grouped singular vectors appear to show some variability in the daytime due to changes in tI (in some cases variability of up to 0.2 occurs).

Page 16766, line 17. "Secondly, simulations with initial time tI during hours with decreasing or increasing insolation are disregarded for categorisation. More precisely, hours with increasing insolation are defined to be between sunrise and 3 h after sunrise and hours with decreasing insolation are defined to be between 4 h before sunset and sunset." Can the authors specify why they introduce this criteria? Also, the initial description "with decreasing or increasing insolation" should perhaps be changed to "with rapidly decreasing or increasing insolation" since insolation exhibits a sinusoidal variability it will still be increasing and decreasing for all but a small period of time in the daytime.

Page 16766, line 21. "Thirdly, for scenario URBAN/BIO only the biogenic part of the scenario is considered, since the first 36 h equal those of scenario URBAN (remember the spin up run of 24 h). The biogenic part of the URBAN/BIO scenario is denoted as scenario BIO."The authors should state whether the exclusion of the URBAN scenario relates to tI, tF, or for both tI and tF.

Page 16767, line 18. "Hence categories Cak/bk, k=1,2,3,4 represent results of calcula-

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tions ending between sunrise k-1 and sunrise k. Thereby, sunrise k, k=1,2,3 specifies the kth sunrise after initial time tI . Sunrise 0 equals initial time tI and sunrise 4 equals final time tF , respectively." This section is somewhat confusing. In the first sentence, do the authors mean "Hence categories Cak/bk, k=1,2,3,4 represent results of calculations ending between sunrise 1 and sunrise 4."? The authors do not state categorically when all of the simulations are initiated. Finally, when the authors refer to tI and tF do they actually mean t0 and tn? Earlier it is stated that t0 and tn define the bounds of the model run period and tI and tF define specific instances of simulation intervals.

Page 16768, line 5. "Notable findings of the categorisation are summarised in the following." The following what? The authors probably need to add the word sections or paragraphs after 'following'.

Page 16768, line 1. "Not in all cases the reduction is large enough to declare the mean impacts to be representative." This sentence needs to be revised as it doesn't make sense.

Page 16768, line 8. "The high NOx values for case FREE (representing the cleanest air) and the low NOx values for case URBAN (representing the most polluted air) are most remarkable." Consider revising the use of the word values to indicate that it is in fact values of ozone sensitivity to NOx. Note too that "that scenarios with rather clean air are in general more NOx sensitive than scenarios with polluted air." doesn't prepare the reader for the extreme cases of ozone insensitivity to NOx shown in the urban cases. Table 4 uses a slightly different nomenclature to this section of text opting for the use of impact, which is consistent with the definition of mi. I am not happy with the use of impact, however, since impact could falsely imply that the 'NOx impact' is in fact ozone production due to NOx, which is altogether different from sensitivity. I think that the authors should adopt the usage of "sensitivity of ozone to ....." in place of impact.

Page 16768, line 17. "For the shortest time interval, there is VOC dominance," For clarity, the authors should note that this is directly implied by values of the sensitivity to

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NOx that fall below 0.5. Perhaps this point should be made earlier in the text to aid the reader. Note that the FREE scenario appears to be an exception to this statement.

Page 16768, line 11. "Further, simulation length tends to change the amount of the NOx sensitivity, but no clear chains of cause and effect are identifiable." Various model outputs could be used to determine the cause of this behaviour. I would urge the authors to spend time examining NOx and ozone lifetimes, model sensitivity to initial NOx concentrations, and the temporal evolution of NOx sink trace gases (e.g. HNO3 etc.). One possible cause of the changes shown in table 5 for the MARINE and LAND cases is that as NOx is destroyed over time the photochemical regime reverts to a more NOx limited conditions. Longer model runs would allow the regime to shift back to the lower NOx concentration/more NOx sensitive cases. Quite why the FREE case does not show similar behaviour is unknown, but perhaps this is linked to the treatment of HNO3 loss terms, i.e. is there deposition of the HNO3 onto ice? If not then the FREE model will reach a steady state between NOx/HNO3. Likewise, the authors should explain why the sensitivities vary according to when the model was initialised, i.e. day or night.

Page 16770, line 11. "Scenario FREE, however, does not share all these features." What features does FREE exhibit?

When the authors discuss chemical species within the mechanism they use abbreviations. It is not always obvious to which chemical species these abbreviations refer, e.g. CSL.

Page 16772, line 16. "For longer simulation lengths however (i.e. simulation lengths longer than (tn−t0)/2), the relative influence of VOC is decreasing with increasing simulation length." Repetition within this sentence should be resolved. Perhaps end sentence with "the relative influence of VOC is decreasing."

Page 16778, line 19. "Since the structural pattern of TOL and XYL is less pronounced than the structural pattern of HC3, HC5, and HC8, the order of maximum impact does

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not match the order of mean influence." Consider revising "structural pattern of TOL and XYL is less pronounced" to "sensitivity of TOL and XYL shows less variability with tI and tF compared to HC3, HC%, and HC8....."

Technical comments:

Page 16746, line 11. Change "More precisely uncertainties....." to "More precisely, uncertainties....."

Page 16751, line 17. "The term singular vector analysis refers to the fact, that.....". Remove comma.

Page 16753, line 15. Change "In case of....." to "In the case of....."

Page 16756, line 16. "For the latter it is of importance, that....." Change to "For the latter, it is of importance that....."

Page 16756, line 20. "This formula is caused by the fact, that....." Change to "This formula is caused by the fact that....."

Page 16748, line 12: remove the two commas: "By investigation of the linearised model, Khattatov inferred, that a linear combination of 9 initial species'concentrations is suïňČcient to adequately forecast the concentrations of the complete set of 19 simulated species 4 days later"

Page 16748. Recommend changing: ".....motivated to further examine the....." to "motivated further examination of the....."

Page 16750, line 19. Second subscript 1 is bold. I think it should be normal font.

Page 16764, line 3. Recommend changing "A detailed description of these mechanism can be found in Seinfeld and Pandis (1998)" to "A detailed description of these mechanisms and regimes can be found in Seinfeld and Pandis (1998)."

Page 16767, line 16. Mispelling of the word length. "According to the second criterion

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(Simulation lengt),"

Page 16768, line 7. "For simulations with initial time tI at day, Table 4 indicates, that scenarios". Second comma needs to be removed.

Various instances of figure references in text exist as 'figure' instead of Fig.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 16745, 2011.