Review #2:

The authors highly appreciate the comments and suggestions of Referee #2, which helped considerably to strengthen the presentation of our results.

General comments:

Comment 1:

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 mechanistic 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.

Response 1:

We agree that "track is lost" for the objective of observation targeting, and the mere identification of sensitivities appears to be somewhat short to accomplish this task in the sense of Buizza and Palmer (1993). In our modifications, we have now addressed this issue in a more focused way. Optimal sensitivities are the critical quantities to be identified, prior to the final identification of optimal observation configurations. Accordingly the following paragraphs were added to the introduction:

"Singular vector analysis accomplishes the identification of measurement priorities by detecting the most sensitive species (here equated to the fastest growing uncertainties). Therefore the first objective of the present work is the singular vector based sensitivity analyses of specific photochemical scenarios, while the second objective is the generation of sensitivity based measurement strategies. [...]

The present singular vector based sensitivity analysis seeks to give insight into the impact on chemical evolution due to uncertainties in emission strengths and initial species concentrations. Special features of interest are the identification of typical temporal patterns of sensitivities in dependence of the time of day and chemical evolution lengths. Finally, the detected sensitivities are applied to answer the question, which chemical species have to be measured with priority. A follow up study will address the same problem with a full 3-dimensional model."

In order to better establish the link from sensitivities to observation targeting, we added a section called "Singular vector based sensitivity analyses" at the end of the theory part explaining the analogy of sensitivity ranking and measurement priority (for details see our Response 1 to Reviewer #1).

Furthermore, we reformulated our sensitivity-findings in terms of targeted observations by adding a section called "Summary of sensitivity results and associated measurement strategy" at the end of each error growth investigation (parts of these modifications are included in Response 3). Finally, we added a summary of the different measurement strategies at the end of the revised paper (for details see our Response 2 to Reviewer #3).

Comment 2:

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.

Response 2:

We thank you for your comment. In our manuscript we calculated leading singular vectors of different scenarios to detect linear species combination that leads to maximum error growth. We placed out study in the quite well established frame of the NO_x and VOCs limitation of the ozone-formation, since the sensitivity of ozone to NO_x and VOC is known to be regime dependent. The chosen context therefore gives us the opportunity to test how these features change for different metrics and to suggest associated measurement strategies. However, we do not believe that SVD is able to add substantial new insights in the quite well established understanding of the respective roles of NO_x and VOCs in formation of ozone (for an approach on this by OH recycling, along with a still existing gap in understanding, see for example Hofzumahaus et al., 2010). On the contrary we want to use well-known facts about the VOC-NO_x-ozone chemistry to confirm our results. Explaining the reasons for the temporal evolution of the VOC and NO_x sensitivities in detail would result in a far too extensive manuscript. We therefore propose to adhere to the key objectives of the paper, but also emphasize its limits in the introduction.

In order to follow the Referees' advice and to help the reader interpreting the results, we extended Section 4.1 (page 16769, line 2) to:

"The finding for simulations with initial time t_I at day is in accordance with the statement of Seinfeld and Pandis (1998) that the availability of NO_x governs the ozone production in rural environments. It can be explained by the fact that the VOC-to-NO_x-ratio is highest for scenario URBAN, followed by scenario PLUME, scenario BIO, scenario LAND, scenario MARINE and finally scenario FREE. At high VOC-to-NO_x-ratios, the hydroxyl radical (OH) reacts preferably with VOC, while at low VOC-to-NO_x-ratios it reacts mainly with NO_2. This is of special importance for the ozone sensitivity to VOC and NO_x since OH is the key reactive species in the chemistry of ozone formation (for more details see Seinfeld and Pandis, 1998). The differing ozone sensitivities to VOC and NO_x for initial time at t_I night are caused by negligible OH-concentrations at nighttime. Here, reactions of VOC and NO_x with NO_3 play an important role. Only with beginning insolation VOC and NO_x start reacting with OH again, initiating the oxidation sequence. Due to the previous nighttime chemistry, the VOC-to-NO_x ratios and the VOC and NO_x mixture itself have already changed and lead therefore to different evolutions of the ozone perturbation."

Comment 3:

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?

Response 3:

We agree. We reformulated our findings in view of this advice at all applicable positions in the paper. As already mentioned above, we added a section called "Summary of sensitivity results and associated measurement strategy" at the end of each error growth investigation and added a summary of the different measurement strategies at the end of the revised paper. Here, we also respond to regime restricted measurement strategies (i.e. measurement strategies that are dependent on the scenario). For example, the measurement section for the grouped error growth reads:

"The sensitivity findings above indicate measurement priorities given the objective of reducing the uncertainty of the ozone-forecast. For measurements at daytime the calculated sensitivities suggest to focus on VOC-measurements in polluted air (scenario URBAN). For moderately polluted areas (scenario PLUME) or urban plumes with biogenic impact (scenario BIO) both VOC and NO_x measurements are of importance. Carefulness is required for measurements in cleaner air (scenarios LAND, MARINE, and FREE). Here, NO_x-measurements are indeed more advisable than VOC-measurements. Nevertheless the ozone sensitivity to the VOC family has a considerable impact of over 20% and VOC-measurements are not negligible. Only for the objective of improving short-term forecasts in the free troposphere VOC-measurements can be omitted.

Referring to measurements by night the above findings do not dictate a clear measurement strategy. Due to constant NO-emissions, scenario URBAN is still NO_x saturated and therefore VOC sensitive. Here, measurements of VOC are most valuable and NO_x-measurements can be omitted. For the other scenarios the ozone sensitivity to VOC and NO_x is dependent on simulation length and photochemical scenario. Due to an ozone sensitivity of over 20% to both VOC and NO_x none of these scenarios allow for omission of VOC- or NO_x-measurements.

In general, the measurement strategy associated to the ozone sensitivity to VOC and NO_x is dependent on photochemical scenario, measurement time and objective. Desirable are measurement strategies where at least one family or species can be omitted from measurement for all considered measurement times and forecast intervals. Since these desirable measurement strategies are advantageous for instrumentation they are denoted as profitable measurement strategies in the following. For the grouped error growth, a profitable measurement strategy is only admissible for urban plumes."

Comment 4:

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.

Response 4:

We are grateful for this advice. Accordingly, we added the following paragraph to the introduction:

"As a preparation for targeted observations of a fully coupled 3-dimensional chemical transport model (CTM) the present work considers the problem of measurement optimisation in a zero dimensional model set-up. This box-model context has the advantage to permit an isolation of specific photochemical environments. Furthermore, the reduced numerical effort (CPU time) of the box-model allows for a comprehensive investigation of special features of interest, like the singular vector dependence on varying time interval lengths (that is, due to ageing of air over days) and initial times (that is, the dependence on the start time in the diurnal cycle). "

Comment 5:

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 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?

Response 5:

We agree with Referee #2. As already pointed out in our Response 1 to Reviewer #1, we see that we did not properly explained the need to calculate singular vectors as **optimal** perturbations, aiming to find maximal sensitivity (which is deviating from results based on proposed alternative approaches). In order to clarify the relation between singular vector analyses and other sensitivity methods, we added a corresponding paragraph to the introduction (see our response to Reviewer #3, Response 1).

Comment 6:

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.

Response 6:

The authors do agree that the relation between relative and absolute error growth statistics need to be presented in a clearer way. In order to make the weights (i.e. the t_I concentrations) more visible for the reader, we added one more figure illustrating the course of the initial concentrations for changing initial times for VOC and NO_x family. Furthermore the link between relative and absolute error statistics is explained by adding the following paragraph to the "Summary and Conclusion"-section (page 16780, line 18):

"These different sensitivities of ozone to particular VOC and NO_x compounds for projected error growths and projected relative error growths can be explained by associated initial concentrations. Indeed, multiplication of the leading projected singular vectors with the associated initial species' concentrations gives an approximation to the ranking of measurement priorities as induced by the leading relative singular vector. In the same manner the behavioural pattern of the leading projected relative singular vector can be reconstructed. Due to a slightly different mix of VOCs and NO_x for projected error growth and projected relative error growth, this reconstruction is only approximately accurate. […]

For all calculated types of projected error growth it is found that summation of the most important projected singular vector entries (associated to the most sensitive species) roughly approximates the associated grouped singular vector entry."

Specific comments:

Comment 7:

Page 16762, refs to table 2 and 3. No mention in either table is made of the water vapor 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.

Response 7:

Thank you for this comment. We added the water vapour concentration to Table 2 (meteorological parameters).

Comment 8:

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 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.

Response 8:

We adopted the advices given. We tried to clarify this section with the following reformulation at the relevant place (page 16764, line 10): "For a comprehensive investigation of those effects, a temporal singular vector diagram (TSVD) is implemented. Each TSVD consists of a complete set of singular vectors comprising several starting times within a chosen time interval as well as different associated simulation lengths. For spin up reasons, the left boundary of this chosen time interval is 2 July, 12h while the right boundary is 6 July, 12h. The hourly time points within these interval boundaries are starting points for a series of singular vector calculations. For sake of clarity, the interval boundary of 2 July, 12h is called starting time t_0 henceforth (corresponding to the first chosen starting point of the singular vector calculations), while the starting point of each individual singular vector calculation is called initial time t_I. Further, the interval boundary of 6 July, 12h is denoted as end time t_n (corresponding to the last chosen starting point of the singular vector calculations), whereas the end point of each individual singular vector calculation is denoted as final time t_F. For each hourly (Δt=1h) initial time t_I, I=0,...,n-1 within the interval $[t_0,t_n]$ a time row TR(I) of singular vector calculations is carried out, leading to $n=(t_n-t_0)/\Delta t=96$ time rows per TSVD. Each time row TR(I) consists of n individual singular vector simulations, all starting at time

$t_I=t_0+I\cdot\Delta t$, but differing in terms of simulation length, which equals $(m+1)\cdot\Delta t$ for calculation m, m=0,..,n-1. Figure 2 illustrates that time row TR(0) consists of 96 simulations with fixed initial time $t_I=t_0$ and varying final times t_F (in Fig. 2 each of these simulation is denoted as $[t_0,t_F]$, F=1,...,96). For the first simulation of TR(0) the simulation length is $(m+1)\cdot\Delta t$ =1h (m=0). Therefore the first simulation begins at July 2, 12h and ends one hour later at July 2, 13h (in Fig. 2 the final time of July 2, 13h is denoted as t_1 since the final time is one hour after starting time t_0). Accordingly, the last simulation of TR(0) has a simulation length of $(m+1)\cdot\Delta t$=96h (m=95) and begins at July 2, 12h and ends at July 6, 12h (denoted as t_{96}). With 96 different initial times and 96 different simulation lengths each TSVD comprises n^2=9216 singular vector analyses."

Comment 9:

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).

Response 9:

The authors are happy to formulate their statement more precisely: "The importance of NO_x and VOC changes with initial time t_I at sunset or sunrise and the behavioural pattern for simulations starting at day or night recurs. Thereby the behavioural patterns of different starting times differ less for starting times within one regime (day time or night time) than for starting times across different regimes."

Comment 10:

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.

Response 10:

Thank you for pointing this out. We changed the sentences in question to "Secondly, simulations with initial time t_I during hours with rapidly decreasing or increasing insolation are disregarded for categorization. More precisely, hours with rapidly increasing insolation are defined to be between sunrise and 3h after sunrise and hours with rapidly decreasing insolation are defined to be between 4h before sunset and sunset. During these hours the behaviour of the calculated sensitivities can differ from the otherwise clearly defined behaviour of the sensitivities with initial time t_I at day. This is due to the fact that the transition from day- to night-time characteristics does not occur abruptly for each scenario. Rather, it tends to proceed more steadily (most prominent for scenario FREE, see Fig. 3c). Omitting these transition-hours from categorization enables to gain plain results for sensitivities with initial time t_I at day."

Comment 11:

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.

Response 11:

We propose to reformulate the statement to "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). Accordingly, only simulations with initial time t_I within the biogenic part of the scenario are taken into account. The biogenic part of the URBAN/BIO scenario is denoted as scenario BIO."

Comment 12:

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

Response 12:

Thank you for pointing this out. The references to t_I and t_F do actually mean the specific instances of the simulation intervals, since the definition of the number of sunrise is dependent on the initial time of each particular simulation. In order to clarify this issue and furthermore define the categories in a more transparent way, we extended our explanations to: "… simulation length tends to be another influential feature. Therefore categories C_a and C_b are further subdivided into 4 subcategories depending on simulation length leading to 8 categories in total. These categories are denoted as C_{ak} and C_{bk}, k=1,2,3,4. Here, C_{a1} represents simulations with initial time t_I at day with a rather shorter simulation length, while

C_{b4} represents simulations with initial day t_I at night with a rather long simulation length. For the subdivision into simulation length, sunrises are chosen as separation criteria. In detail, each category $C_{ak/bk}$ contains results of calculations ending between sunrise k-1 and sunrise k. Thereby, sunrise k, k=1,2,3 specifies the k^{th} sunrise after the initial time t_I of each particular simulation. For simulations starting at July 2, 12.00h, the first sunrise is at July 3, 04.45h, while the first sunrise for simulations starting at July 3, 16.00h is at July 4, 04.45h. Differing from this definition, sunrise 0 is defined to equal the initial time t_I of each simulation and sunrise 4 is defined to equal the final time t_F of each simulation, respectively. Accordingly, category C_{a1} includes results of all simulations with initial time t_I at day that end before their associated first sunrise."

Comment 13:

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'.

Response 13:

Thank you for pointing this out. We changed the according sentence to "Notable findings of the categorization are summarized in the following paragraphs."

Comment 14:

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.

Response 14:

We revised this statement, which now reads "For all scenarios, the subdivision in terms of simulation length leads to a reduction of the standard deviation. Still there are some cases where the standard deviation is relatively large, for example the standard deviation for category C_{a4} for scenario MARINE (see Fig. 5). For the sake of clarity a further subcategorisation, which would allow for a larger reduction of the standard deviation, is not applied."

Comment 15:

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.*

Response 15:

We thank you for this comment. We reformulated the respective sentences to "…that scenarios with rather clean air are in general more NO_x sensitive than scenarios with polluted air. While scenario FREE (which represents the cleanest air) is characterised by high ozone sensitivity to NO_x, scenario BIO is nearly in VOC-NO_x balance. Eye-catching is the extreme ozone insensitivity to NO_x for case URBAN (representing the most polluted air)." Furthermore, we followed the recommendation to adopt the usage of "sensitivity of ozone to ......" in place of impact.

Comment 16:

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 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.

Response 16:

On page 16765, line 22 we extended the explanations to "Since all singular vectors are set to unit length, a vector component of 1 indicates that the ozone concentration at final time is solely influenced by this particular compound or family. Furthermore, a NO_x vector component larger than 0.5 indicates that the ozone evolution is dominated by NO_x, whereas a NO_x vector component smaller than 0.5 indicates VOC dominance. The opposite applies for VOC vector components." Furthermore, on page 16768, line 14 we stated "...Table 5 shows two common features for scenarios LAND, MARINE, PLUME and BIO. Firstly, their mean ozone sensitivity to NO_x is smaller than 0.5 for the shortest time interval, which implies VOC dominance. Secondly, these scenarios show decreasing ozone sensitivity to VOC with growing simulation length." The new formulation states the implications of the NO_x-values and furthermore clarifies that scenario FREE is excluded from these statements.

Comment 17:

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.*

Response 17:

In fact there is no heterogeneous chemistry HNO_3 sink included in our tangent-linear and adjoint RACM set-up, that is, no dissolution in cloud droplets or aerosol surface reactions are introduced. Further investigation showed that scenario FREE indeed reaches a steady state between NO_x/HNO_3. However, as already pointed out in Response 2, we do not apply singular vector analysis to gain new insights in the roles of NO_x and VOC in the formation of ozone, but to investigate the problem of targeted observation of chemical constituents in a well-known context. Since furthermore a detailed explanation of the sensitivity results would result in a far too extensive manuscript, we propose to adhere to the key objectives of the paper. Nevertheless, we outlined the reasons for varying sensitivities according to the initial time t_I of the model (see Response 2).

Comment 18:

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

Response 18:

In order to answer this question, we added the following sentences: "However, scenario FREE does not share all these features. Here, the ozone sensitivity to VOC is mainly determined by species MGLY, DCB, CSL and ALD, while the contribution of species HC3, KET and ETH is negligible. The particular contribution of each VOC-species to the ozone sensitivity to VOC does not increase or decrease continuously with growing simulation length, but changes from decreasing to increasing or vice versa."

Comment 19:

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.

Response 19:

The authors agree that this is not ideal. Therefore we added a Table with definitions of the RADM2 species abbreviations for the VOC-compounds and referred to this Table when first mentioning these species.

Comment 20:

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."

Response 20:

Thank you for this recommendation. We reformulated the sentence as proposed.

Comment 21:

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 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, HC5, and HC8......"

Response 21:

We reformulated the sentence as recommended.

Technical comments 22:

- *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 sufficient 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 mechanisms 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 (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.*

Response 22 (to all technical comments above):

Thank you for pointing out and correcting these mistakes. We revised our manuscript accordingly.

Comment 23:

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

Response 23:

According to the ACPD Conventions (Abbreviations and Acronyms, at http://www.atmospheric-chemistry-and-physics.net/submission/manuscript_preparation.html) the abbreviations "Sect." and "Fig." should be used when they appear in running text followed by a number unless they come at the beginning of a sentence. Since the 'Figure'-references of our manuscript all appear at the beginning of a sentence they are consistent with these guidelines.