Review #1:

The authors are grateful for the fundamental criticism of Referee #1, which is welcomed as it helped substantially in the process of clarifying the description of both the objectives and the methodological approach of the paper.

General comments

Comment 1:

The singular vector decomposition is applied for the determination of the sensitivity of _one_ single variable (ozone concentration at the end of the box-model run) with respect to a few input parameters (initial concentrations or emission factors of chemical species). For this task the application of an adjoint model or, in the case of only a few parameters of interest, even "brute-force" runs of the original model (modifying one parameter per run) would be sufficient - and even superior to the singular vector decomposition (SVD), which has the following disadvantages:

1) The programming effort before running the SVD is large.

2) The numerical effort (CPU time) of the SVD is very large.

Response 1:

We acknowledge that we did not properly explain the need to calculate singular vectors as **optimal** perturbations, aiming to find maximal sensitivity in the following sense (which is deviating from results based on proposed alternative approaches):

We are seeking the **most quickly** growing perturbation (in the linear approximation) to each central chemical scenario run over a given time period — i.e. the singular vectors corresponding to the largest singular value of the linearized propagator for the chemical evolution over that time interval.

As an extension to this, we want to investigate, how singular vectors change continuously with varying time intervals (that is, due to ageing of air over days) and initial times (that is, the dependence on the start time in the diurnal cycle). In our manuscript, we describe the first application of our SVD scheme where we limit the investigation to a few input parameters. For future applications, we want to keep the dimension of the problem flexible, i.e. optionally as high as the number of species. We also anticipate the application of our model in a full three-dimensional chemistry transport model. As we point out in our manuscript, this use of SVD methods is a well proven approach to examine the stability of complex dynamical systems.

In contrast to singular vector analysis, the application of an adjoint model as well as the utilisation of the “brute-force” approach do result in first-order local concentration sensitivity coefficients and **do not result in an optimal initial perturbation** in terms of a preselected metric (and subsequent lower ranking orthogonal perturbations).

Further, our objective and claim is not to suggest a method of low programming effort. Rather, the optimality property of the fastest growing singular vector is required. As the adjoint and tangent-linear model versions with respect to initial values are provided by the software package KPP (Kinetic PreProcessor), there was no substantial programming effort. The elevated numerical effort (CPU time) of singular vector calculations is the prize to be paid for identifying the initial perturbation with maximal growths. Approximating singular vectors with iterative applications of brute force methods would result in similarly high CPU time.

As a consequence of this and other reviewers' remarks, we added a new paragraph to the introduction explaining the need for singular vector analysis and distinguishing the applied method from other proposed sensitivity approaches (for further details see also our Response 1 to Reviewer #3).

Furthermore we propose to add a section called "Singular vector based sensitivity analysis" at the end of the theory part explaining the link from sensitivities to observation targeting:

"On the basis of different error norms the singular vectors introduced in this section all identify the most quickly growing perturbation (in the linear approximation) of a chemical scenario run over a given initial forward time period. These optimal perturbations can be regarded as the most critical errors. The magnitude of the perturbation entries of one compound reveals how sensitive the considered forecast result is to small errors in the initial concentrations or emission factors of the associated species. The order of magnitude of the singular vector entries equals therefore the order of sensitivity. Following Palmer (1995), dynamically sensitive model states (i.e. high singular vector entries) identify moreover valuable measurements sites. In a chemical box model context, where only one location is considered, dynamically sensitive model states identify chemical species that have to be measured with priority. Hence singular vector analysis solves the question of measurement priority in terms of sensitivity ranking. Optimal sensitivities are the critical quantities to be identified, prior to the final calculation of optimal observation configurations."

Comment 2:

3) Singular vectors, as presented in the manuscript, are normed (norm = 1). That is why they can represent only _relative_ sensitivity measures (importance of one species _relative_ to another one).

Response 2:

For our linear analysis, a relative sensitivity measure is what we want, with the generalisation of introducing operators for specific metrics. Particularly for targeted observations, sensitivities of species relative to each other as well as their relative time and regime dependence are of great importance considering the objective to prioritise measurement of one species over measurement of another.

Comment 3:

4) As both the vectors v and -v are singular vectors, the information on the sign of sensitivity is lost. However, this is an important information: for instance, as correctly pointed out by the authors, an increase of the initial NOx concentration may lead to an increase or decrease of the final ozone concentration, depending on the conditions (initial values) considered.

Response 3:

We fully agree that information about the sign of sensitivity is useful. However, the information on the sign is not lost within the singular vector analysis, since it can be provided by multiplication of specific entries of initial and final singular vector. A positive sign for e.g. $(\delta NO_2(t_I))*(\delta O_3(t_F))$ indicates that the final ozone concentration changes in the same direction (increase or decrease) as the initial NO_2 concentration, while a negative sign signals the opposite. As singular vectors are stored at initial and final time, this information of sign is available without any extra costs.

Comment 4:

The authors seem to be aware of this general problem, because in the abstract, introduction, and summary they stress the usefulness of the singular vector decomposition for solving another problem: optimization of measurement configurations of chemical compounds. However, contrary to the statement at the beginning of the summary, this topic is not even mentioned in the main body of the manuscript. Now the main logic behind the manuscript appears to be: "In order to demonstrate that method M1 (singular vector decomposition) is suitable for problem P1 (optimization of measurements), we show that it works for a related problem P2 (sensitivity analysis), which can be solved better by a simpler method M2 ('brute-force' sensitivity analysis or adjoint model)".

Response 4:

We agree that the presentation of the logic in the manuscript may appear in the criticised way. But as already pointed out in previous comments, we utilized the method of singular vector analysis (method M1) to calculate optimal sensitivities, while the simpler method M2 is not providing the results, we are looking for. In terms of targeted observations, a ranking of the singular vector entries by magnitude equals the measurement priorities. Therefore, singular vector based sensitivity analysis does solve problems P1 and P2 simultaneously. By formulating the objective of the paper more clearly (for details see our Response 1 to Referee #2) and furthermore analysing the results in terms of targeted observations (for details see our Response 2 to Referee #3) we try to emphasize the analogy of sensitivity ranking and measurement priority in a more focussed way. Additionally, we propose to introduce the section "Singular vector based sensitivity analysis" which establishes the link from sensitivities to observation targeting (for details see our Response 1 above).

Specific comments:

Comment 5:

The results are very detailed, probably too detailed for the majority of the readers. The individual analysis of the impact of several (groups of) chemical species (NOx, VOC, CSL, XYL, TOL, MGLY etc.) for 8 categories of model runs (different initial time and length of the simulation) for 6 different scenarios (LAND, MARINE etc.) makes it difficult to present a "take-home message" for the reader.

Response 5:

In our revised version of the manuscript we follow the Referee's advice and draw a unified picture of the results. As a first step, the categorisations in terms of scenario, starting time (day/night), and integration lengths are already serving this purpose. With help of those categorisations we identified differences in the sensitivity patterns for different scenarios and their modification with increasing integration time or ageing of polluted air.

In order to present a "take-home message" for the reader, we focus more on feasible measurement strategies (for details see our Response 2 to Reviewer #3). Apart from establishing the link between sensitivity ranking and targeted observations in the theory section, we added a section called "Summary of sensitivity results and associated measurement strategy" at the end of each error growth investigation. Finally, we revised the conclusions to contain a summary of the different measurement strategies at the end of the revised paper (for details see our Response 2 to Reviewer #3) leading to the final conclusion that "While the sensitivity of ozone to the VOC or NO_x family varies with time and suggests therefore mostly time-dependent measurement strategies (i.e. for a time-independent measurement strategy all species have to be measured), the relevance ranking of species within one family remains scenario-wise relatively stable. Here time-invariant targeted observations can effectively be applied omitting measurements of several less sensitive compounds. Given the multitude of reactions and the time and scenario dependence of the chemical evolution, the potential of ranking species into less, middle or very important is a very positive result showing that singular vectors can be utilised effectively for optimal campaign and instrumentation planning."

Technical comments:

Comment 6:

Most parts of the manuscript are technically well written, containing some minor errors, e.g.:

- page 16748, line 12: "linear combination of 9 initial species'" should be "9 linear combinations of initial species'" (cf. Khattatov et al., 1999, p.18724)

Response 6:

We are happy to correct this.

Comment 7:

- reference "Sandu et al. (2006)" should be "Liao et al. (2006)"

Response 7:

This has been corrected.

Comment 8:

- Equ. (38) is a system of ordinary, not partial, differential equations.

Response 8:

We modified the revised version of our manuscript accordingly.

Comment 9:

- Fig. 4 is probably wrong (now it is a copy of Fig. 5)

Response 9:

Thank you for pointing out this fault, which escaped our attention, after the manuscript was re-edited by the journals' office. In the revised version of our manuscript this has been corrected. Furthermore, the correct Fig. 4 can now be found in a supplement to the article, together with an explanation of the Copernicus Office.