Review of "**Efficient ensemble generation for uncertain correlated parameters in atmospheric chemical models**"

The authors presents a systematic and efficient approach to generate emission perturbations in chemical transport models. They applied their method to biogenic emissions of isoprene (in particular). As they pointed out, perturbing emissions can easily require an extremely large number of perturbations. First they derive an important result that under linearity assumption (or TLM) the sensitivity of perturbed model parameters when considering all combinations scales as R^{J} where J is the number of model parameters and R the number of cases considered (generally 2 – one for control the other for the perturbed. It is shown that under the assumption of linearity, that this total sensitivity can be obtained by perturbing each model components individually, this significantly reducing the total number of perturbation. Secondly by constructing the error covariance of those perturbations (in model space), it is possible to obtain the leading eigenvalues/eigenmodes of the covariance matrix by using a large scale eigenvalue software known as APRACK that does not require storing a (very large) covariance matrix. Finally, an approximation of the perturbations can be obtained from these leading eigenvalues/eigenvectors by using a Karhunen-Loeve expansion. Although, as the authors discusses, the equivalence between the combined sensitivities and the independent sensitivities are not meet with certain nonlinear processes, such as meteorology, this is nevertheless an important contribution that should be published. However, the document somewhat hard to follow, especially in the mathematical description of the method, and some clarifications and a rearrangement of the theory would be most beneficial. Although this may represent some rewriting, it is believed that it can be easily accomplished.

Major issue:

Section 2.1 is hard to follow and uses concepts that are not well defined. Table A1 gives examples that greatly help understand what the concepts may actually mean. It is unfortunate that this table appears in the appendix section. The authors should provide examples (as in Table 1) of the concepts introduced – especially for the second paragraph Lines 105 to 111. It maybe worth considering splitting section 2 into a section on "Efficient sensitivity calculation" using sections 2.1, 2.1.1, and then 2.1.2, followed by a new section (section 3) discussing the algorithm which would include lines 82-91, figure 1, section 2.2 and 2.3. It could also be welcomed to have a figure in the section 2 (around equation 9) that shows the required number of forecasts J as a function of I for the combined sensitivity and independent sensitivity calculation (for a few values of R).

Minor points:

1 – line 67-69. Should draw the parallel and differences between the Principal component analysis and the Karhunen-Loeve expansion for discrete functions. Also the Principal component analysis is in fact widely used in geophysical sciences for example in climatology.

2 – line 98. It is not clear what is multi-variational covariance ? Do you mean multivariate covariance ? If not this has to be defined.

3 – line 203. Please define s/s’

4 – line 262. Is the joint perturbation is define by using a multivariate covariance matrix, C. Or is it an observation the results for each species leads to similar eigenvectors ?

5 – Lines 333-337. The results not shown, should be shown as it is part of the main finding of this approach and study.

6 – Figure 5. Not sure what the numbers above each panel refers to. Please explain or drop. Same for figure 7.

7- Lines 355-356. Don’t quite follow the argument of this sentence “The approach is based … “ . Please expand and explain.

8 – Section 4. Discussion and conclusions. Could you comment on how this method may provide an error uncertainty associated with each eigenvector of the expansion, and thus how the method could be used in inverse modelling.