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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Mapping the drivers of uncertainty in atmospheric selenium deposition with global sensitivity analysis" *by* Aryeh Feinberg et al.

Anonymous Referee #3

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The authors carry out a sensitivity analysis of atmospheric selenium deposition using polynomial chaos expansion as a surrogate model. In general, this is a new and interesting application of surrogate modelling and sensitivity analysis and so has the potential to represent a forward step in the field. Additional information is required on the methods carried out to convince me that the surrogate model is suitable for the sensitivity analysis, particularly for Se Lifetime.

In particular: 1. There are some choices made in the application of the polynomial chaos expansion that are not explained and it's difficult to know whether they are rule of thumb decisions or made specifically for this application. How was q=0.75 decided

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upon and what other choices were there to least-angle regression?

2. I don't follow the logic of explaining LOO validation and then using a model that doesn't actually leave any of the training runs out. Can you explain why this is appropriate and how the validation might change if you had used LOO? Is it appropriate to continue to call it LOO validation with a full model? It may be explained in one of the given references but I'd like to see some explanation here.

3. You choose the degree based on whether the validation doesn't decrease in the next step. Why and how does the validation increase? I would expect the validation to always decrease to some extent with extra terms. If a threshold is applied, what is it?

4. It seems counter-intuitive that calculating the surrogate separately for burden and deposition yields better results for lifetime than directly modelling lifetime? I would expect there to be double the errors. How was 'better' calculated and can you explain a bit more why it's a fair result?

5. Can you explain a little more about how the sensitivity analysis is derived from Equation 4? It's not enough here to refer to previous work.

6. Given the results in Figure 5 I'm not convinced that double counting interactions because they are less than 0.05 is a good idea. 0.05 is quite a large fraction of the 0.15 that is the largest main effect. Can you find a way to investigate the effect this is having?

7. In Figure 5, the main effects are quite low and nowhere near adding up to 1 – was the amount of interaction in this model expected?

8. Still with Figure 5, in previous experience seeing interactions that are large and consistent between multiple variables is a sign that the model fit is actually poor. It's not clear because of the way you have carried out LOO and added the interaction terms whether this is indicating poor model fit or whether these are real interactions.

9. The main effect figures show that there is not much range on the y-axis covered by

the central line – it's highlighted by the uncertainty in the remaining parameters. Could you add some information on how much uncertainty there is from using PCE as your surrogate? I would like to see this to show me that most of what you are seeing is not simply a result of the use of a surrogate model.

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