Response to Referee #2 for the manuscript: “Estimating aerosol emission from SPEXone on the NASA PACE mission using an ensemble Kalman Smoother: Observing System Simulation Experiments (OSSEs)”

Dear Referee #2,

Thank you for reviewing our manuscript. Your comments definitely help to improve the readability and define some aspect of our work better. Below you can find our point-by-point responses to all of your comments.

Best regards,
Athanasios Tsikerdekis

**Format**
Questions
Responses
“Quotes from the manuscript and revised or added text.”

**Main Comments**
This paper describes an ensemble of OSSEs to estimate the top-down constraints on aerosol emissions based on observations provided by SPEXone. While the experimental setup and the scientific results are interesting, the paper needs improvement before publication. In particular, the english should be checked throughout. Additionally, the text needs to be more concise and some sections are unclear.

Thank you for your comments and appreciate your criticism. We have reread and improved the manuscript where we deemed necessary. Specifically we have given special attention to the subsections you mentioned on you minor comments (Abstract, Subsection 3.1 and Subsection 3.2).

**Minor Comments**
Abstract: please shorten the abstract, which currently reads more like a conclusion. Indeed the abstract was very detailed and we shorten it considerably from 483 words to 381 words (reduced by 20%). We hope the new version gives a more concise and laconic summary of our work.

L168-170: The use of the term analysis for 1 cycle and posterior for several ones here makes little sense. Please find another terminology.
In our work emissions are estimated iteratively in time. Which means that the estimated emissions are going to be affected from observations on the same day, but also by observations in subsequent days. Therefore the term analysis is reserved for updated emissions that were corrected by n days of observations (n < ΔTa), while posterior for updated emissions that were corrected based on the full length of the smoother lag (n = ΔTa). This can be better understood visually in Figure 1. We realize that the sentence may have caused confusion so we have rephrased it:
“Note here that the term analysis is used to indicate the updated emissions affected by n days of observations (where n < ΔTa), while the term posterior is used to indicate updated emissions affected by ΔTa days of observations (Figure 1).”

L177: And what impact did you find?
The impact is discussed in detail on the result section and specifically on subsection 4.2 Emission estimation using SPEXone – Sensitivity experiments.
The impact on the optical properties: “SPX_W1 and SPX_W2 reduce the ΔTa length to 4 and 2 days (from 6), hence less observations are used to derive the analysis emissions in each assimilation cycle and only 2 and 1 assimilation cycles (instead of 3) are used to calculate the analysis emission perturbations. The results reveal that ΔTa=4 days (SPX_W1) is sufficient to constrain the AOD, AE and AAOD in a similar manner as a ΔTa=6 days (SPX) (Figure 11 a,b,c). In other words, under the current experimental setup, observations 5 to 6 days after the emissions probably hold very little information for the correction of these emissions, and their exclusion has a very limited impact on the data assimilation performance. Contrary the experiment SPX_W2 shows a degradation in performance over western Sahara and North Atlantic for AOD and AE (Figure 11 d,e,f), indicating that observation in subsequent days 3 and 4 hold useful information for the correct estimation of emissions at day 1 and 2 as discussed in the next paragraphs. Note that SPX_W1 and SPX_W2 need ~33% and ~66% less computational resources than SPX respectively, since the background step in each assimilation cycle is shorter.”

The impact on emissions: “Finally, SPX_W1 emission bias increases no more than 6 percent points in comparison to SPX in all species. However, dust emission error grows to 54% in SPX_W2 from 17% in SPX_W1, indicating that the information content of observations 3 and 4 days after the emissions is very rich and it should be used to correct these emissions, especially for Sahara dust plumes that extent over the Atlantic Ocean and last for several days. The emissions of OC, BC and SO2+SO4 are estimated very accurately by all of the data assimilation experiments, with relative MAE ranging from 0% to 5%, which indicates that in terms of the global mean emission estimation these emissions are unaffected by the sensor spatial coverage and observational uncertainty increase that were tested.”

L187: "[...] a unique distribution to drive the emissions[...]")" has a distinct prior emission distribution"
Changed as suggested.

L188: Should remove that sentence. This is confusing. What is the approach used to generate the prior error correlations? Please describe clearly.
Thank you for giving us the chance to clarify. We have rephrased the sentence and provided a reference to our previous paper where we describe this process in detail.
“Changes in neighboring grid cells of each member are not abrupt but smooth. This spatial correlation of the prior perturbations was generated using spatial smoothing, a method where data points are averaged with their neighbours. A step by step description on how our spatially correlated perturbations are created can be found at subsection 3.2 of our preceding work (Tsikerdekis et al. 2021).

L193: The mean and the standard deviation of the distribution (or the ensemble), not of the perturbations
Replaced “perturbations” with “distribution”, as suggested.

L195: Please better explain the rationale here.
Indeed a more detailed explanation was lacking at this point. Thus we have added:
“Furthermore, it is noted that the perturbations are uniquely defined every ΔTs=2days (different colors in the boxes of Figure 1). The rationale here is that the simulated observations and emissions at day D (where D is any integer number) will be more correlated than the simulated observations at day D+ΔTs and emissions at day D.
Consequently, changes in emissions caused by assimilated observations of day D will be stronger compare to changes in emissions by assimilated observations of day D+ΔTs. This design is based on the fact that observations on the day of the emissions carry more information about the emissions, than observations in subsequent days.”

Section 3.2: The whole section is not clear. It needs to be rewritten entirely. Please utilize equations rather than long sentences wherever appropriate. This will greatly facilitate the reading and the understanding of the so-called "prior correction" approach (currently unclear).

Thank you for noting this. We have adjusted subsection 3.2 making it more concise and adding equations where it was necessary. Further, we highlight that Figure 1 and Figure 2 improves the readability and comprehension of this subsection as they were designed with special attention to the details referred in the text.