The present study aims to estimate aerosol emissions from the SPEXone polarimeter of the upcoming NASA PACE mission, using Observing System Simulation Experiments (OSSEs) with the ECHAM-HAM global model and an ensemble Kalman Smoother. Two satellite observing systems are used to assimilate aerosol optical properties; the SPEXone and an idealized sensor with full spatial and temporal coverage. A significant improvement of prior emissions is found even with the SPEXone sensor with the limited coverage. In addition, several sensitivity experiments are performed, in order to investigate the role of biased meteorology, different emission inventories in synthetic observations and simulations, uncertainty in emissions, and days affected by assimilated observations, in emissions estimation. This is a well-written paper providing sufficient information on the different experiments setup and results. Given the limited number of studies on aerosol OSSEs, I find the paper to be a significant scientific contribution, as it provides information on the ability of the SPEXone instrument in the upcoming NASA PACE mission to be used for aerosol emission estimation, as well as on the benefits of an idealized instrument with full spatial and temporal coverage. I recommend publication of the paper after the following comments are addressed.

Comments:

- If I am not mistaken, the figures presenting the spatial distribution of the differences between the examined experiments present also as "mean" the mean difference (?) globally. Such an approach might result in masking of the error when positive and negative differences appear with similar frequency. I suggest also presenting the global mean of the absolute differences in order to have a more realistic overview of the differences between the experiments. Moreover, I suggest defining (in the text) the metrics used in the study (maybe as Appendix?).
- The NAT experiment represents the synthetic observations used for the assimilation. Do the authors have an estimate on how the NAT spatial variance is compared with that of the real observations? How does the comparison between the two might affect the
improvement of emissions estimation? This is something that needs to be discussed in the text.

Minor comments

- P2, L33 and where applicable: I suggest using the Oxford comma as "size, and absorption with...".
- P2, L42: Add a comma after "In addition".
- P2, L50: the have largest -> have the largest
- P4, L104: Add angle units.
- P4, L105: that it measures -> measuring
- P4, L106 and where applicable: Add space before nm in 700nm.
- P4, L107: Multiple sentences here starting with SPExOne. Please rephrase.
- P5, L139: taken into account -> are also taken into account
- P11, L330: I suggest replacing the first sentence with "Figure 4 shows that the differences between DAS and NAT (solid lines) reach a value close to zero after 26 days." In this case you can delete "(Figure 4)" in the nest sentence.
- P12, L357: “nature run” Please use the abbreviations (NAT here) throughout the manuscript.
- P12, L386: from NAT -> from NAT for AOD, AE, and AAOD.
- Figure 2: Maybe replace purple with green to better distinguish from blue.
- Figure 6 caption: Please indicate what the presented Mean value stands for.
- Captions of Figures 10 and 11: depict -> depicts
- Figure 13: The experiments name is missing from the DU and SS figures.
- Figure 18c: I suggest replacing orange rectangle with a blue one to better distinguish from the red one. Please also describe what the rectangles stand for in the respective caption.
- Figure A 1: There seem to be some color shade areas in the scatter plot. Please indicate in the caption what they stand for.