Review of " Data Assimilation of Volcanic Aerosols using FALL3D+PDAF" by Mingari et al.

Overview: The authors apply an ensemble data assimilation method (LETKF) to obtain optimal estimates of volcanic ash/SO2 concentrations using the FALL3D dispersion model. For ash simulations, synthetic data are used while for SO2 simulations, satellite retrievals obtained during the 2019 eruption of Raikoke are used. Both experiments yielded significantly better results than reference experiments in which no data assimilation was employed. However, it was noted that during the first assimilation cycle in which the prior ensemble was based on straightforward sampling of model parameter uncertainty ranges, the probability distribution was non-Gaussian, resulting in unphysical (negative) optimal concentration values in the posterior ensemble. This was attributed to the Gaussian assumption underpinning the LETKF.

General comments:

This work is scientifically sound and will be of interest to the volcanic ash dispersion modelling community. I recommend that the authors consider the following issues in the manuscript prior to publication, which are mostly related to the presentation.

1. More clarity is needed about how the details of the algorithm. In my view Figure 1 is nice but doesn't really help the reader understand what is actually being done. For example, even after the reading the whole paper it was not 100% clear to me how the ensembles were generated at each cycle. Do you initialize an ensemble of dispersion models using prior uncertainty estimates at t0 and compute analysis at t1, then use analysis at t1 to re-initialize the dispersion model and propagate to t2 and so on? That would mean that the initialization at the first step (volcanic source?) is quite different from initialization at subsequent steps (distal?). Please provide more concrete details so the reader doesn't need to guess.

2. On reading, it feels like the appendix was originally part of an earlier chapter. I would suggest that the authors either perhaps shorten the appendix and then insert it at an earlier stage as part of the methodology section or make an effort to make sure that discussion in Section 2 is self-contained and does not require the reader to read the appendix first. Some of the specific comments below are related to this issue.

Specific Comments:

Introduction: There is a substantial body work on 'inverse modelling' methods using satellite retrievals of volcanic ash that has not been mentioned. See for example list of citations in Zidikheri, Meelis J., and Chris Lucas. "Improving Ensemble Volcanic Ash Forecasts by Direct Insertion of Satellite Data and Ensemble Filtering." *Atmosphere* 12.9 (2021): 1215. It would also be useful to mention what the DA method in this manuscript can do that these other approaches cannot do given that these methods also use observations to improve the forecasts.

Lines 100-105: "Background error covariance" is mentioned in Line 105 but it wouldn't be clear to readers unfamiliar with DA methods what the word "background" is referring to. It would be helpful to define "background forecast" (= " a priori forecast") earlier in the paragraph. Might also be useful to mention why the error covariance is important in DA.

Line 128: Last sentence of paragraph is hard to understand. What do you mean "filter operations are performed exclusively by ranks…". What "ranks"?

Line 134: Sentence stating that LETKF is "more realistic for volcanic ash" than ETKF might need a reference (or explain why you think this would be the case). Also, this statement is rather puzzling given that you state in the abstract that LETKF didn't work very well. I think a summary of the

differences between ETKF and LETKF might be needed here – including a brief discussion of the need for localisation in ensemble DA methods in general. Many readers will probably not have the time or inclination to read the appendix in detail even those details are available there. See also General Comment #2.

Line 146: "range" – is this is the localisation radius? "inflation factor" – needs explanation.

Lines 164-166: I didn't really understand this explanation for why the ensemble forecast prior PDF forecast would be skewed. Isn't the skewness just a consequence of the way the prior ensemble is constructed? Could not in principle the prior uncertainty be sampled in such a way so as to yield a more symmetric distribution?

Line 203: the overbar needs explanation (ensemble mean?)

Lines 212-216: Is there a reason for focussing on SO2 rather than volcanic ash retrievals here? Ash concentrations (rather than SO2) are of more interest in practical applications.