Response to reviewer 2:

We have marked our responses in blue.

The manuscript presents an inversion method (particle filter based) to derive the volcanic ash emission profiles by converting two-dimensional ash loading data from, for e.g., geostationary satellites to three-dimensional emission data. Similar to previous studies the authors combine observations and ensemble simulations. The novel aspect of the method lays mainly in its ability to estimates the errors and uncertainties in the derived emissions. The authors use the inversion system for two notional sub-Plinian eruptions of the Eyjafjallajökull and show that the method's accuracy strongly depends on wind shear conditions.

The methods are valid and the results are interesting for the remote sensing and modeling communities. I have no major comments but few remarks that should be addressed before publication.

We very much appreciate the reviewer comments concerning the readability of our manuscript. We hope this helps to improve the text and to show our findings more clearly. Please see our response below.

General comments:

- The method quantifies the uncertainty with respect to the injection height and vertical wind profile but there is no hint of the uncertainty in the assimilated quantity, i.e. ash mass loading. The satellites do not measure this quantity directly. Rather, it is a retrieved parameter based on brightness temperature. The retrieval has its own limitations and uncertainties. Most importantly, in the first few hours of the eruption the umbrella cloud is quite large and thick so the ash retrievals are either missing or subjected to large uncertainties. Therefore, the authors should discuss the limitations of the method from this perspective.
  The reviewer is certainly right. We did not consider retrieval uncertainties of integrated column values as a possible obstacle to devaluate our concept. The observation error covariance matrix R (defined prior to eq. (3)) accounts for the impact of retrieval errors of the ash column loads and is considered diagonal. It can be made spatially and temporally dependent, to account for assumed increased retrieval errors due to water cloud influences, particularly thick umbrella ash clouds above or in the vicinity of the volcano or interference of other aerosols as e. g. mineral dust. In our study, we have made assumptions about the observation error (including retrieval error). Certainly, in applications to real volcanic eruptions, the use of retrieval errors provided by the observations is highly encouraged.
  Starting from a scalar column load value as exclusive data source we considered estimation uncertainties of the derived height profile presented here as an order of magnitude larger than retrieval errors. We are grateful for the reminder to consider columns sufficiently distant to the nascent umbrella cloud.

The observation error can also be incorporated in constructing the ensemble, as in general any ensemble data assimilation procedure can straightforwardly account for the retrieval uncertainty by artificially perturbing retrievals of column mass loads, where the random perturbation is scaled by the assumed statistics of retrieval errors. This perturbed observation approach is included in our analysis. Clearly, this must not be the only means to generate the ensemble, as this accounts only for a fraction of overall uncertainty, resulting in underdispersive ensembles. We included the remarks above in the formulation of the objectives and the data use description.

- The authors assume that the only parameter relevant for the ash transport is the wind. What about the particle size and aerosol dynamics? How does the method address the uncertainties with respect to processes like particle growth and sedimentation? Ash aggregation leads to particle growth and enhances the removal. Please discuss the limitations of the method from this perspective.
  Aerosol dynamics (nucleation, accumulation, deliquescence) and aerosol chemistry in EURAD-IM is based on MADE (Ackermann et al. 1998, with substantial update of the thermodynamical part by Friese and Ebel, 2010, both developed at our research group), which has been switched off for two reasons: Numerical efficiency in an ensemble context and specifics of volcanic ash properties cannot be expected to be reasonably well featured by a general pollutant aerosol module like MADE. Ideally, a full volcanic ash aerosol dynamics and chemistry as proposed by Schmidt, see e.g. https://www.springer.com/gp/book/9783642348389 would be in place, along with its not existing adjoint. Yet we consider the error to be negligible within the evolution time frame addressed in our idealized study. As requested, we added a discussion on the limitations of the method from this perspective in the text.
- The text is very difficult to read. It starts right in the title and then continues with the odd formulation of the first two sentences in abstract. In many places throughout the paper, the verb comes in a passive form at the end of a long sentence. This makes the text very difficult to follow. Please consider writing in an active form and avoid long sentences. Especially sections 1 and 2 contain lots of odd formulations and difficult passages. Section 3 is easier to follow but has some generic formulas related to validation processes. Please move all the formulas and their explanations to section 2.
  Thank you for the comments on the readability of the manuscript. We have revised the abstract and the first two sections, with emphasis to avoid long sentences, (and certainly also to follow the fellow reviewers' advices). We agree that the length of the title is poor. GMD guidelines request the model / model version in the title. So there was little space for optimization. Yet we now hope to present some reduced lengths.
  As requested, we have moved all formulas related to validation to section 2.

Specific comments:

L1-5 (Abstract): The sentences read odd and are difficult to follow. Please revise.

We have fully revised the abstract to increase the readability of the text. The first sentences now read: ”A particle filter based inversion system is presented, which enables to derive time- and altitude-resolved volcanic ash emission fluxes along with its uncertainty. The system assimilates observations of volcanic ash column mass loading as retrieved from geostationary satellites. It aims to estimate the temporally varying emission profile endowed with error margins. In addition, we analyze the dependency of our estimate on wind field characteristics, notably vertical shear, within variable observation intervals.”

L25-30: This is not an encouraging opening paragraph. The sentences read odd and are difficult to follow. Besides, there is no clear connection between the points. Please revise
We have revised the first paragraph of the introduction. It now reads: “Emission profiles of volcanic eruptions depend on multiple parameters, such as crater size or exit velocity of the emitted mass. Further, they depend on atmospheric stability and wind profile at the volcano. Many of these parameters are unknown or difficult to measure exactly. This renders the estimation of emission profiles of volcanic eruptions challenging for chemistry transport models in the context of data assimilation and inverse modelling for source estimation. Therefore, special methods for assessing the strength and vertical distribution of volcanic emissions are necessary. As volcanic eruptions contain enormous amounts of harmful trace gases and particulate matter, a detailed knowledge not only about the spatial and temporal variations of the emissions and its strength is needed but also accurate information about the analysis error of the emissions and the evolving volcanic ash cloud is required.”

L38: please add the specific uncertainties of these methods. Besides, add a
We have added the description of uncertainties of the methods to the text: “Statistical models base on observational data from historic volcanic eruptions, which are sparse and show a large variance in eruption rate for given plume heights. For example, Mastin et al. (2009) calculated an uncertainty by a factor of four in estimating the emission rate for a plume height of 25 km using their statistical model. Physical plume-scale models require orographic details of the volcano (e. g. crater size) but also meteorological fields and parameters (e. g. wind entrainment coefficients), which are often poorly known and render these models highly uncertain. Costa et al. (2016) identified the wind entrainment coefficient as main source of uncertainty leading to up to two orders of magnitude differences for the estimation of mass eruption rates for weak volcanic eruptions. In their analysis of the eruptions of the Eyjafjallajökull, Iceland, in 2010 and Grímsvötn, Iceland, in 2011, Woodhouse et al. (2015) found a comparable range of uncertainty depending on the choice of the wind entrainment coefficients.”

L108: you mean “It should be noted” or “we note”?
We have change “it is noticed” to “Please note”.

Figure 2: I do know that this is an idealized set-up. But is it physically realistic to have the same profiles and emission rates under two different atmospheric conditions (wind shear)?

Vertical wind shear and associated horizontal wind conditions are found to make the main difference of ash cloud analysability, given only estimated column mass loadings. We have chosen to analyze a special case of volcanic eruption with two short but strong emission pulses in a row. We chose this emission profile to test the method's ability to distinguish the two pulses under different wind conditions. Indeed, if we apply the same eruption parameters to a plume model we would expect differences in the emission profile and rates. However, the emission profile used in our analysis remains realistic to occur, even though differences in the source parameters (exit velocity, water content, etc.) could be expected.

Figure 4 and 5: I did not find the source of meteorological data in the text.
We have added the information about the meteorological data in the text: "As we consider the differences of feedbacks of the ash clouds on the meteorological evolution as not critical on the forecast time scale in our idealized tests, the EURAD-IM is offline coupled with the Weather Research and Forecasting (WRF) model version 3.7 (Skamarock et al., 2008). Meteorological boundary conditions are taken from the ECMWF (European Centre for Medium-Range Weather Forecasts) analysis."

Figure 8: what happens at 12 hours after the eruption. These is an spike in the error.
Figure 8 shows the relative mean absolute error of the (unobserved) volcanic ash concentrations. Thus, the volcanic ash concentration of the ensemble mean is compared to the nature run, from which observations have been extracted, in each grid cell. At 12 UTC, the spike in the RMAE results from the error in estimating the emission strength in the upper part of the eruption column. This error is smoothed out in the subsequent hours reducing the RMAE again. In Appendix A shows the emission profile for each assimilation window. Here you can see the errors of the test case using a 12 hour assimilation window in estimating the second eruption plume around 7 UTC, which leads to the large errors. We have added this information to the discussion of the figure.

Conclusion: Again very hard to follow. Please make it clear and concise.
We have revised the full conclusion to make it clearer.

Literature:

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