Authors response to reviewer 3:

We have marked our responses in blue.

The authors present an idealised study of a particle-filter-based inversion system to obtain height- and time-resolved volcanic ash emission estimates using ash column loads from satellite. The idealised study provides a good testing ground for their inversion method, without the complication of modelling errors and incomplete observation datasets which are encountered in real situations. I think it is suitable for publication but could benefit from some improvements to the readability and complexity of the manuscript. It is an interesting study and I'm keen to see future developments on this work.

We thank the reviewer for the encouraging comments and detailed suggestions helping to improve the manuscript. We give our responses below.

- I find the manuscript unnecessarily wordy in places and not the easiest to read. For example, the title is rather detailed - is 'sub-Plinian Eyjafjallajokull' and 'version 1.0' necessary here. Another example is the caption of Figure 2 – stating it shows the emission profile should be adequate – I would question the need to include what it is to be used for here. The authors use some complex words (e.g., 'pairwise distinct' (line 91) – does this mean independent? – 'investigated exemplary' (line 306) – what does this mean?). In places, sentences are very long. Section 3 is rather long – could it be split into subsections? Readability could be improved, I feel.
  Thank you very much for this helpful comment. We agree that the title is long. However, we have had discussed the title with the technical editor and came to the conclusion that the title should contain information about the eruption type („hypothetical sub-Plinian Eyjafjallajökull eruption") as well as the method characteristics („particle filter based"). Further, the naming of the model and model version is a requirement by GMD and cannot be removed. Unfortunately, this prerequisite almost doubles the length of the title. However, we have removed "the chemical component of" from the title and hope this improves the readability of the title.
  We agree that the caption of Fig. 2 contains unnecessary information. We have changed the caption to: „Hovmoeller-like plot of the nature run emission profile used in this study. Shown is the emission rate (colored) for a given time (x-axis) and height above the volcano (y-axis)."
  The mathematical phrase "pairwise distinct" implicitly means that no members of a set are equal to another. Here the phrase refers to the fact that every emission package, which is defined to be pairwise distinct, covers a unique time and height spot in the emission profile. We have added this information to the phrase: "The simulation is realized by an ensemble, in which each ensemble member simulates the dispersion of one single emission package. Thus, each emission package covers a unique time and height spot in the emission profile. We refer to this ensemble as ensemble of emission packages."
  We have removed the phrase "investigated exemplary". The respective sentence

now reads: “As an example, the analysis results using an assimilation window of 24 hours are investigated in more detail.”

We have revisited the full manuscript with special focus on long and complicated sentences as was also suggested by the other reviewers. Further, we have split Sect. 3 into subsections.

- It would help to have some connection between the theory (section 2) and the practice for this case study.

  We have added a table explaining the variables used in this manuscript. We have also carefully revisited Section 2.4. Here, we have added information about how the system is applied to volcanic ash eruptions and how the theory is used in practice.

  - Line 102: c is defined as a default mass of ash in the emission package but just previously on line 92 ‘a unit mass of ash’ is stated. I began to wonder why c was needed if it was one. Reading on, I realised that c is probably a first guess (e.g. from a prior) and that a is some adjustment factor. This could be made clearer. Again, on line 153 there is reference to ‘default mass’ without this ever being defined.

    Indeed, we have not been clear here. c is a default mass of ash, which can be arbitrarily chosen. It defines the resolution of the emission strength in our estimate. We have made this point clearer in the manuscript. The variable c is now consistently defined as “default mass of ash” throughout the manuscript, which is scaled by the factor **a**. This factor **a** is to be optimized in the DENM algorithm.

  - The use of the Nelder-Mead algorithm seems unnecessary since the problem here is linear and the resulting minimization problem is quadratic. The situation described on lines 114 and 115 (‘cases where the function to be minimized has discontinuities or the function values are noisy’) does not apply here. Again, this seems to overcomplicate the manuscript and is an example of the gap between the theory stated and the case study. Furthermore, the required restriction to only allow integer values for a for the method to be efficient (I presume the use of the word ‘effective’is a typo?) is unnecessary for such a simple minimization problem which could be solved explicitly and will no doubt introduce some uncertainty / errors. I take the authors point that the Nelder-Mean algorithm is more widely applicable but wider applications are not studied.

    Basically, we fully agree with the reviewer that the minimization problem is quadratic, apart from the bounding due to positive semi-definiteness of all components (i.e. a bounded minimization problem). In fact, we started our study with quasi-Newton L-BFGS, where we always have had best success compared to other methods like CG (since the beginning of our chemistry data assimilation activities with 4D-var, Elbern and Schmidt, 1999). (The L-BFGS-B(ounded) version is very inefficient for parallelization, due to a Gauss-Seidel type problem.) To our experience, essential to the success of high dimensional quadratic minimization problems are two items in data assimilation: introduction of a background state reasonably close to the "truth" for a tangent-linear approximation to hold, (traditionally provided by a preceding forecast), and, to some extent linked with that, an efficient preconditioning (see e.g. Elbern et al., 2007, for a preconditioning technique by diffusion approach (Weaver and Courtier, 2001, in atmospheric science). With an increasing number of model levels and their (positive semi-definite) concentrations to be attributed, while column values as given data are single scalars only, the ill-conditioning of the minimization problem increases drastically and a much needed reasonable background information prior to the volcanic eruption is hardly available. Hence, this missing a priori knowledge cannot serve any preconditioning requirements other than highly speculative inferences from assumed eruption type and strength scenarios. Even simple smoothness assumptions of the vertical profile are often invalid for ash clouds. While we presently test to maintain positive semi-definiteness by assuming log-normal distribution underlying the least square minimization, we performed tests with the Nelder-Mead method, which performed clearly best, without getting lost in drastically elongated minima as introduced by underdetermined degrees of freedom through vertical level concentrations. Yet we agree that after an initial coarse Nelder-Mead minimization, a final least square step will have the potential to prove successful in the future. We add a modified discussion to this issue in the text.

- Line 133: What is 'the model state'? Presumably it is either the source emissions (a times s used earlier) or the model predictions of ash column loads (both H M[as] and x hat (equation 2) used earlier). Can you standardise the notation? Is x in equation 3 related to x hat in equation 2? If not, perhaps another variable other than x could be used for one? Also, whilst notation is consistent for x and y, the subscripts in equation 8 suggest to me derivatives or (x,y) coordinate components.
  We have added a table that explains the used variables. We have further extended equation 2 to show the link between M(as) and x. Equation 8 does not show the derivative but the deviation from the average. The subscripts do not refer to (x,y) coordinate components but to the modeled and observed volcanic ash detection. The notation using the prime symbol " ' " was introduced in Zidikheri et al. (2016). Thus, we suggest to keep the notation in Equation 8 as is.
- Lines 150 – 151. Does the mention of the capability of using ensemble meteorological members add anything to this paper? This capability isn't used, and its mention could cause confusion to the reader – what does 'ensemble' refer to hereinafter? Lines 191 - 192: 'EURAD-IM comes with the adjoint code of the chemical and aerosol modules for four–dimensional variational data assimilation.' – again, is this relevant to this study?
  We agree that our mentioning of the meteorological ensemble may cause confusion. However, it is an important feature of ESIAS-chem, which is used in subsequent analyses. Thus, we have rewritten the sentence:

“Further, it is capable to be coupled with ensembles of meteorological fields to account for additional uncertainties resulting from meteorological forecasts. However, this investigation focuses on the ability of the system to reconstruct the emission profile and its uncertainty under perfect meteorological conditions. Thus, no meteorological ensemble is used at this stage.”
We have removed the sentence concerning the adjoint of the EURAD-IM to avoid further confusion.

- Detail on the error covariance matrix B is thin. On one hand you say, ‘no fixed assumptions have been made for matrix B’ but later you state that ‘B is chosen as diagonal’. How is the optimal value found?
  We agree, we have discussed the B matrix insufficiently. We have added more detail to the estimate of the B matrix. Our point is, that the matrix B can be altered to include constraining information (e. g. correlations of different emission packages). In this study, for simplicity and lack of knowledge on height-time eruption parameters we have decided to not constrain the emission packages, leaving matrix B to be diagonal. The actual values were found by sensitivity runs. Thus, the sequence now reads: “In first tests without the regularization term, the emission rates have partly increased to unrealistic high values. Therefore, the B-matrix was chosen in a sequence of sensitivity tests, in which the influence of the regularization term on the emission profile was evaluated. Best results have been found by choosing B as diagonal matrix B=diag(10). Please note that the chosen diagonal form of the B-matrix led to reasonable results for the artificial emission profile used in this study. However, for realistic applications a more elaborated evaluation of a properly chosen B-matrix is required and straightforwardly applicable. In this performance test, the only purpose of the matrix serves to restrict the scaling factors **a** not to vary too strongly. In addition, the regularization term was chosen in order to maintain a suitable spread of the analysis ensemble.”
- Presumably the subscript 0 in line 169 is an iteration subscript?
  Yes, the subscript 0 indicates the first guess. However, we have removed the formula because it adds no further information to the manuscript and only causes confusion. We have changed the related sentence to make our point clearer: “The minimization is initialized with a set of arbitrarily varying scaling factors **a** for the pairwise distinct emission packages.”
- It would help for sections 2.3 and 2.4 to be linked better. For example, how does the weightings and likelihood relate to the cost function?
  We have carefully revised section 2.4 to better link the theory of the different methods.
- I’m presuming the ‘ensemble mean’ (line 242) is obtained from the ensemble members that are accepted by the particle filter, weighted according to the weights in equation 5? It would help the reader, to elaborate here. Also, I find the subsequent mention of ‘mean’ (line 249) and the ‘mean’ and the overbar (line 251 and equation 9) confusing. Also,

on lines 263 – 264, the ensemble mean is denoted by an overbar. Can any improvements be made to help the reader negotiate these apparent different means?

We have added some information on how to calculate the mean in our analysis. As the ensemble members are resampled, the weights are not applied by calculating the ensemble mean. We have made clearer, that **va** is the binary volcanic ash detection vector for the analysis ensemble mean and that the overbar in line 49 denotes the spatial average of the volcanic ash detection vector **va**. We hope that this avoids further confusion.

- I would have liked the authors to state early on (perhaps in the abstract) that the study is an idealised study (I think the word 'idealised' is more commonly understood than 'identical twin') and therefore does not consider errors in the modelling (both in the input meteorology and in the model parametrizations) nor incomplete observations. I would also have liked to have seen more discussion of the implications of this work to real life situations (e.g., when errors will exist in the meteorological data and in the transport model and observations may be incomplete (perhaps due to the presence of meteorological cloud)). For example, the authors state that 'an assimilation window of 24 hours is sufficient in order to provide reliable forecasts', but this may not be true for a real case study.

  In the revised abstract, we have added that we demonstrate the system's validation in an idealized setup. Further, by introducing the term identical twin experiment we add this information again. We have also changed the headline of Section 3 from "Identical twin experiments" to "Validation of ESIAS-chem". We suggest leaving the term identical twin experiment as it refers to a special case of idealized studies, well defined in the data assimilation community (e.g. Daley 1991). We believe that our updated explanation of identical twin experiment is sufficient to avoid further confusion.

  We have revised the discussion and conclusion section and have added a discussion about the limitation of the current study to generalize the results.
- I suggest some thought is given to the use of the phrase 'data assimilation'. What is meant by the term 'data assimilation'? Some uses of the phrase I would refer to as source inversion methods, rather than data assimilation.

  Thank you for your suggestion. We agree that the term data assimilation is not precise enough. We used the term data assimilation to acknowledge the use of observations and model simulations to provide improved model analyses. Indeed, our main goal is the source inversion for volcanic ash emissions. We have revisited the manuscript and changed "data assimilation" to "source inversion" were appropriate.
- Lines 253 – 254: Why is the RMAE calculated over points where both the modelled and observed values are above the limit? If one was to compare model forecasts for a given set of observations, the RMAE may be obtained over a different set / number of (model, obs) pairs. If, however, one was to compare the RMAE calculate over points where the observed values are above a limit, one would have a consistent set. Why is the relative error (equation 11) normalised by the model ensemble mean (similarly equation 12)? The RMAE is normalised by the observations.

We thank the reviewer for this note. Indeed, the RMAE was calculated over different datasets. Our intention was to exclude low volcanic ash values from the comparison. However, this is also accomplished by taking all points, where the observation is above the threshold. We have changed the figure accordingly. The results are similar to the previous one.
We have chosen to normalize the relative error in Equation 11 by the modeled ensemble mean to be comparable by the relative standard deviation, for which the normalization with the model ensemble mean is more intuitive. To be consistent with the RMAE, we have normalized the relative error and relative standard deviation by the nature run emissions. As the maximum emissions of the nature run and analysis ensemble mean are comparable, the results remain valid.

- I would encourage the authors to add some model runtime information.
This has also been suggested by Nina Kristiansen (reviewer 1). As the core focus in this study is the reconstructability of the 3D ash field based on wind shear driven sequences of 2D column field imagery, numerical efficiency was not our primary concern. The run time of the ensemble system is an informative value about the applicability as early warning system. However, as for other methods in the literature, we have decided not to concentrate on the computational performance. Thus, we have adapted the simulations to the available compute resources (especially granted wall clock time). We run the ensemble of emission packages subdivided into chunks of 60. Further, we have increased the number of iterations in the DENM minimization to 15,000 (including restarts), which is not feasible in a realistic early warning scenario. However, we chose 15,000 iterations in order to track the performance of the minimization. We found that the costs reached the minimum value after ~1,000 iterations. With this setup, the run time of the system is not competitive with other algorithms.

Minor points

- Lines 140-141: ‘no assumptions of the error statistics of the model state and the observations were made’. Is this true? The likelihood function commonly includes an error covariance term (as in equation 1 and equation 6), hence I would think there are some assumptions made in calculating the weights, even if the error covariance term is the identity matrix.
We see that we have not been clear on this point. The particle filter methodology is applicable to all kinds of probability density functions and is not restricted to Gaussian model and observation errors. We have added this to the text: “It is noted that in the particle filter method no assumptions of the statistical forecast error characteristics of the model state and the observation error were made (the errors do not need to be normally distributed and the model state does not need to be unbiased as other data assimilation methods require).”
- Line 26: ‘Chemistry transport models have limits in estimating the emission strength’. The emission strength is usually assumed in chemistry transport models. Models are not generally used to estimate emission strengths, except in the context of data assimilation / source estimation methods or by some simple

inference from observations.

You are right. We have changed this statement in the revised introduction. This first paragraph 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.”

- Line 31: ‘analysis error’ – it’s not clear whether this refers to the emission estimates or the predicted cloud.

  In principle, this statement is valid for both, the emissions estimate and the predicted volcanic ash cloud. We appended “of the emissions and the volcanic ash cloud” after “analysis error” to better illustrate this.
- Line 37: ‘and thus making’ should be ‘and thus make’ or ‘thus making’ or something similar.

  We have removed the full sentence in the revised introduction.
- Line 44: Satellite retrieval methods also usually retrieve an estimate of the cloud height.

  Thank you very much for pointing out that we need to be clearer at this point. We agree that there are many retrieval methods, exploiting infrared satellite measurements to obtain volcanic ash properties including retrieved cloud height information (e.g. Ventress et al., 2016 or Piontek et al., 2021). However, these only include cloud top height retrievals and give no information on the vertical extent of the ash cloud or the averaging kernel. In the sentence (line 44), we referred to data sets as used in Stohl et al. (2011) and Prata and Prata (2012). In these studies, an ash cloud height (and ash cloud thickness) has been roughly estimated according to the observed brightness temperatures. This cloud height then serves as retrieval input and is not included in the retrieved data sets. We now adjusted the related sentence to: “In contrast to lidar observations […], column mass loading observations rarely provide information about the vertical distribution of volcanic ash and are mostly limited to cloud top heights (e.g. Ventress et al., 2016 or Piontek et al., 2021).”
- Line 52: ‘remains to be solved’. I would probably dispute this. Established source inversion methods for volcanic ash use atmospheric wind shear to be able to determine the three-dimensional ash cloud information from two-dimensional observations.

  You are right. We give some examples of these inversion methods in the subsequent paragraph. Thus, we have removed this statement from the manuscript.
- Line 73: ‘but also may the’. Something is wrong here with the English – perhaps remove ‘may’?

  We have changed the sentence to: “They found that not only the ensemble statistics should be evaluated but also the single ensemble members, which may contribute significant information to the distribution of volcanic ash.”

- Lines 96-98: The work of Stohl et al and Kristiansen et al estimates the source emission profile (from which the volcanic ash column mass loading can be modelled) and the Bayesian method used does provide uncertainty information.
  Indeed, we are sorry for not have acknowledged their work in a pertinent way. We have added this information to the text. In addition to Stohl et al. (2011), in our approach the uncertainty estimation of the emission profile is used provide probabilistic estimates of the volcanic ash cloud extent. This has the potential to identify areas with high volcanic ash content that are not directly observed.
- Line 150: 'suitably' should be 'when suitably'?
  We have change "suitably" into "given"
- Line 187: 'on our case' should be 'in our case'?
  We have removed this half sentence in the revised version of the manuscript.
- Line 198: 'Spin' should be 'Spinning'
  Done
- Lines 223-224: 'Contrary, vertical and horizontal mixing of volcanic ash emitted may limit the benefit that is gained by increasing the assimilation window length.' I can't see why this would be the case – can the authors explain? I can see that additional observations from increasing the assimilation window length may not provide any further information (particularly in an idealised study) but why the reference to 'vertical and horizontal mixing'? Is it because the ash cloud may be below satellite detection limits when widely dispersed? Does the benefit differ depending on whether the study is an idealised study (i.e., no modelling errors and full view of the ash cloud) or a real case study (with modelling errors and missing observations)?
  This statement refers to the distinction of emission packages given vertically integrated ash column data. Once the volcanic ash emitted by different emission packages is well mixed due to vertical or horizontal mixing, it is impossible to attribute the volcanic ash to one or the other emission package. This effect is independent of our idealization in this study. We have added this example to the text: "For example, if volcanic ash emitted by two different emission packages is mixed, it is impossible to attribute the volcanic ash to one or the other emission package."
- Caption Figure 4: 'in approx. 5 km' should be 'at approx. 5 km'. This typo appears in a few other places in the manuscript (e.g., lines 326, 355, 396) – it should be 'at' a height, not 'in' a height.
  Thank you for this note. We have changed this typo throughout the manuscript.
- Line 229, Figure 4b. It's not clear whether this is height above the volcano (which probably doesn't make sense over a spatial region) or height about ground or height above sea level? How does one associate it with Figure 4a? Does one need to know the height of the volcano? Similarly, line 231. Lines 231 – 232 and Fig 4c require some units / labels for the variables stated / shown (e.g., label on the y axis, units for temperature and pressure). Similarly, for Figure 5 and associated text. Also, height information is not specific in Figures 9-12 and associated discussion.
  Thank you for mentioning this issue. The wind speed in Fig. 4b is actually plotted on pressure level 500 hPa and not at a height of 5 km. We apologize for the typo.

We have changed the caption of Figs. 4 and 5: “Meteorological conditions on 15 April 2010. (a) Wind speed above the volcano for the whole simulation period. (b) Wind speed at 500 hPa on 15 April 2010, 12 UTC, which corresponds to approx. 5 km above the volcano. (c) Vertical cross-section of isobars in [hPa] (red) and isotherms in [K] (grey) along the red line in b) on 15 April 2010, 12 UTC.” We have changed the text accordingly.
We added a label to the y-axis in Figure 4c. The units of isobars and isotherms are added to the caption. Similar changes have been made in Figure 5 and its caption.
In Figs. 9 and 10, the height is referred to the height above the volcano. Also, in Fig. 11 and 12, the height is referred to the height above the ground. We have changed the label of the y-axis accordingly.

- Lines 279-280: ‘Again, the pattern correlation coefficient does not account for deviations in the strength of volcanic ash column mass loading at locations in which the ensemble mean and the nature run differ in volcanic ash load’. It considers differences above and below the limit applied.
  You are right. We have changed the sentence to: “However, the pattern correlation coefficient is a measure for volcanic ash column mass loading above and below the chosen threshold. It does not measure differences in the strength of volcanic ash column mass loading above the threshold.”
- Line 316: Given the same emission profile is used in each nature runs, why are these total values different?
  Actually, the emission profiles slightly differ, which is not visible with the chosen colorbar. This difference results from the calculation of the emission profile in the underlying EURAD-IM model, in which the model layer depth is taken into account.
- Line 337: ‘the mixing of volcanic ash in the atmosphere is too effective’. This study is an idealised case study so the mixing of volcanic ash in the atmosphere is represented perfectly. My opinion is that the second case study does not enable the vertical distribution of the ash emissions to be determined by wind shear and hence the filtering method yields an emission profile which is widely distributed in the vertical compared to the nature run. The first case study has significant wind shear which allows the vertical distribution of emissions to be strongly constrained, but this is not possible for the second case study.
  We understand your interpretation of our results. The lack of vertical wind shear is one limitation for the estimation of the emission profile for the second test study. However, our results may also suggest alternative causes, which we would like to discuss briefly: Fig. 10b shows the mean emission profile of the analysis ensemble. Especially for the first eruption column around 3 UTC, the emission rates are underestimated in the full vertical column. Thus, the error is unlikely due to the vertical distribution of the emission rates. Compared to the nature run, lower emission rates in the eruption column are rather compensated by strong emissions at later hours. This leads to a temporally highly smoothed emission profile. In addition, Fig. 8b shows a low relative mean absolute error of the volcanic ash concentrations. Thus, we conclude that the dispersed volcanic ash cloud resulting from the temporally smoothed emission profile for the second

test case is similar to the volcanic ash cloud resulting from the nature run's emission profile.

- Line 381: It's not just the uncertainties in the meteorological fields which are neglected in this study, uncertainties in the model parametrizations (e.g., turbulent dispersion, washout, etc.) are also neglected.
  You are right. We have added this to the discussion section: "The analysis is idealized in different ways: The uncertainties in meteorological fields, especially in winds, in model parameters (e. g. deposition velocity), and parametrizations (e. g. clouds) have been neglected. Further, the amount of observational data is exceptionally large, with observations of the full domain every 6 hours. Thus, observations of ash-free areas allow for removing volcanic ash emissions from the analysis. The ability of ESIAS-chem to give reliable results for real volcanic eruption using non-idealized meteorology and incomplete observations needs to be addressed in another study."

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