Dear Anonymous Referee #1,

On behalf of all co-authors, first of all I would like to thank you for giving us very useful comments and suggestions. We really appreciate your detailed comments and suggestions, which certainly improve the quality of the paper. In the revision, we have carefully considered the suggested changes (grammar issues, extraneous figures, discussons of model error, forecast improvement and the background on the use of aircraft data for EnKF) and included them to the best of our ability.

In the following we will give our response to every comment. To make the changes easier to identify, we have numbered them.

Best regards, G. Fu

(The revised manuscript is in the latter part of this pdf.)

Reply to the comments and suggestions:

1. 2.10: This should be clarified to also acknowledge that some satellites provide very detailed vertical information on plumes (e.g., CALIOP) but are thus spatially sparse.

Response: It is included in lines 2.10-2.12:

"Note that, some satellites can provide very detailed vertical information on plumes (e.g., Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar measurements) but are thus spatially sparse (Winker et al., 2012)."

2. 2.16: Actually, LIDAR gives gives profiles of e.g. aerosol extinction. I think the point the authors are trying to make here is that optical measurements are only an indirect constraint on concentration, which require further modeling and assumptions to translate into concentration estimates.

Response: Yes, we agree. It is reformulated in lines 2.17–2.19:

"Whereas other measurements such as satellite data and LIDAR data observe optical properties that contain indirect information about concentrations and require further modeling and assumptions to translate into concentration estimates."

3. 2.21: "These kinds of observations" is rather vague and potentially overly broad, as there have been many previous studies that have used EnKF to assimilate aircraft observations other than the authors' previous work. It seems that the background on the use of EnKF with satellite data for air quality forecasting could be expanded.

Response: I agree that "These kinds of observations" is not clear here. "These kinds of observations" refers to aircraft ash measurements discussed in last paragraph. It is now changed to lines 2.22–2.23:

"Recently in the application of volcanic ash transport, the benefit of aircraft in-situ observations in an Ensemble Kalman Filter (EnKF) system has been studied (Fu et al., 2015).

4. 3.19: Splitting hairs on semantics, but this type of model can be evaluated but never really "validated", see Oreskes et al., Science, 1994.

Response: It is corrected in line 4.1:

,,

- "Recently, the LOTOS-EUROS model has been evaluated as an appropriate volcanic ash transport model (Fu et al., 2015)".
- 5. 3.22: It's not really clear how aerosols are treated based on this description. What does it mean "average particle size are implemented" Perhaps a brief summary using customary descriptions of aerosol modeling theory would be helpful (internal or external mixture? fixed size, modal or sectional scheme? etc.).

Response: I agree with you that it was not clearly described how aerosols are treated. In the revision, Table 1 is added to show the "average particle size" and customary descriptions of aerosol modeling are added in lines 3.21–4.11:

Table 1: Volcanic ash particle size distribution and ash bins property for LOTOS-EUROS model simulation.

LC)II.			
	Bins	Particle Diameter	Percent of Mass	Average Particle Size (μm)
	vash_1	250 to $2000~\mu\mathrm{m}$	29	1125.00
	$vash_2$	$63 \text{ to } 250 \ \mu\text{m}$	31	156.50
	$vash_3$	$30 \text{ to } 63 \ \mu\text{m}$	12	46.50
	$vash_4$	$10 \text{ to } 30 \ \mu\text{m}$	18	20.00
	$vash_5$	$2.5 \text{ to } 10 \ \mu\text{m}$	8	6.25
	$vash_{-}6$	0.0 to $2.5~\mu\mathrm{m}$	2	1.25

"The input parameter PH in LOTOS-EUROS is taken from hourly based Icelandic Meteorological Office (IMO) plume height detection and usually the uncertainty of PH is taken as 20 % (Bonadonna and Costa, 2013). For VMD, large explosive volcanic plumes have a typical 'umbrella' shaped vertical distribution (Sparks et al., 1997) and as such this 'umbrella' shaped VMD is adapted into LOTOS-EUROS (Fu et al., 2015; Lu et al., 2016). The PSD in LOTOS-EUROS is defined in the ESP type S2 (see definations in Mastin et al. (2009)), in which the mass fraction of erupted debris finer than 63 μ m is 0.4. For the S2 type eruption, Durant and Rose (2009) provides the base for the PSD from their analysis of the 1992 Crater Peak, Mount Spurr event. Hence, Table 1 provides the ash distribution based on their analysis and is used by LOTOS-EUROS. Another input parameter MER is very hard to measure for an explosive onsetting volcano. Usually it is calculated from the plume height. Mastin et al. (2009) studied the relation between these parameters and concluded that an empirical relationship between plume height PH(km) and mass eruption rate MER (kg s⁻¹) is

PH =
$$2.00V^{0.241}$$
, and $\frac{V}{MER} = \frac{1.5e^3}{4.0e^6}$. (1)

where V(m³ s⁻¹) represents the volumetric flow rate.

Recently, the LOTOS-EUROS model has been evaluated as an appropriate volcanic ash transport model (Fu et al., 2015) (Fig. 1a), where the volcanic ash concentrations are described by 6 aerosol tracers including PM₁₀ and PM_{2.5} (corresponding to vash₋5 and vash₋6 in Table 1) to model the transport process. The physical processes that are relevant for volcanic ash are similar as those that apply for mineral dust, e.g., advective transport and diffusion, deposition, coagulation, sedimentation, and resuspension (Langmann, 2013). Where the transport is determined by the wind fields that could be regarded as rather well known, the other processes deposition and sedimentation processes are rather uncertain. The parameterizations for the later processes involve assumptions on the particle distribution for example, which is difficult to summarize in a few numbers of bins. These processes act on the distribution of the total ash mass over the modes (particle sizes) and the total mass load; one could therefore state that almost everything in the description of an ash cloud is uncertain, except for the cloud shape and position. The processes included in this study are transport, sedimentation, and wet- and dry-deposition, where the relevant properties such as average particle size (Table 1) are implemented following Zhang (2001).

6. Section 2.3. Add numbers to equations. I think the apostrophe is being used to represent transpose, which is a bit odd. Superscript T would be much more standard.

Response: The apostrophe was used in some book on data assimilation, I agree that Superscript "T" is much more commonly used for the transpose. Numbers have been added to all equations and the transpose is represented with Superscript T.

7. Page 4, Line 1; Page 5, Lines 2 and 21: Comment: The 10% measurement uncertainty is repeated in three places. Consider including it only in Section 2.2 or 3.1. At the very least, the first time this is mentioned it should be explicitly stated that it includes an estimate of the model error as well. But later, in section 3.1, perhaps it needs to be explained further how the value of the model transport error is quantified. Usually this takes some work, and properly identifying the horizontal and vertical correlation in the model error is rather critical tuning of a robust EnKF.

Response: The 10% measurement uncertainty is now only included in Section 2.2 and 3.1.

As recommended, in section 2.2 (lines 4.22–4.23), it is explictly stated:

"The total measurement error not only contains the instrumental error, but also includes an estimate of the model representation error (Fu et al., 2015)."

In section 3.1, we added a detailed explanation about the model representation error in lines 6.14–6.27:

"To assimilate measurements in a simulation model, it is necessary to quantify the model representation error. The model representation error is the discrepancy between the quantity that instrument observes, and what the model value represents. Concentration values are defined on discrete grids with a finite resolution at discrete time steps. A measurement location usually does not coincide with the grid point where the concentration value is defined. The spatial resolution of the model used in this study is around 12 km \times 12 km \times 1 km, therefore the volume of one grid-box is about 150 km³. Through model processing, the concentration of one grid-box represents an average value for this grid-box, while

one aircraft in-situ measurement is a point value in a 3 dimensional field. In this study, we choose the in-situ measurement corresponding to the grid-box average value. This approximation makes sense only when two assimilated measurements are positioned in two different grid-boxes. Considering the aircraft speed of 100-200 km h⁻¹ and the LOTOS-EUROS horizontal and vertical resolution, a 10 minutes assimilation frequency is chosen to guarantee different assimilated measurements are in different grid-boxes. The observation therefore almost corresponds to one model state variable in a grid-box, which means the representation error of the model is probably small. For the moment we will therefore not explicitly specify a model representation error, but implicitly assume that it is zero. The total observation representation error, defined as the sum of the instrumental error and the model representation error, is taken as 10% in this study.

8. Figures 1b and 1c are unnecessary.

Response: I agree with you. In the revision, Figures 1b and Figures 1c have been deleted.

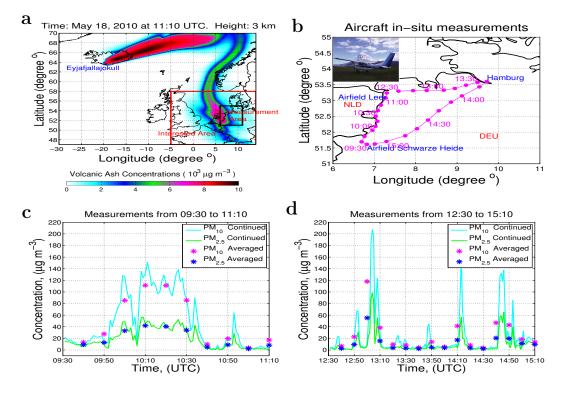


Figure 1: Aircraft in-situ measurements of distal volcanic ash plume. a, The LOTOS-EUROS simulation of volcanic ash plume at 11:10 (UTC), 18 May 2010. b, Measuring aircraft flight route on 18 May 2010. c, PM_{10} and $PM_{2.5}$ measurements from 09:30 to 11:10 (UTC). d, PM_{10} and $PM_{2.5}$ measurements from 12:30 to 15:10 (UTC). In c and d, the curves show the values of PM_{10} and $PM_{2.5}$ measured at a frequency of every 6 seconds. The values marked with a star are the averaged PM_{10} and $PM_{2.5}$ (average every 10 minutes) which are used in the LOTOS-EUROS model in accordance with the model simulation step (10 minutes).

9. 6.16 Bringing the analysis members into agreement in locations where there are no observations is only a sign of consistency, not accuracy. Independent measurements that are not assimilated are the only way to evaluate the forecasts away from the assimilation data.

Response: It is indeed not correct to say the accuracy in locations where there are no observations. It has been changed in lines 7.19–7.21:

"This tells us that assimilating aircraft measurements effectively reduce the ensemble spread of the whole distal ash plume, which is a sign of consistency to the measurement locations."

10. 7.26. This strikes me as a somewhat optimistic conclusion. It seems the evidence presented is that for some locations the forecast is improved, but in other it is not. Given the sparsity of the data used for assimilation, more often it is not. Rather, one might conclude that significantly more observations are required to completely constrain the ash plume across the domain. Extensive aircraft observations, or some combination of detailed aircraft measurements as used here with more broadly available remote sensing data may be useful.

Response: We agree that the conclusion was too optimistic. It is reformulated in lines 8.27–8.33:

"This shows that the forecast at these locations with assimilation is more accurate than the forecast without assimilation, so that the assimilated volcanic ash state (Fig. 3d) is a more accurate approximation to the real state of distal volcanic ash plume. In addition, we conclude that the assimilation process performs well in combining with the LOTOS-EUROS transport model with real-life measurements. Note that we find that for some locations the forecast is improved, but in other it is not. Thus, significantly more observations are required to completely constrain the ash plume across the domain. Extensive aircraft observations, or some combination of aircraft measurements as used here with remote sensing data may be useful."

11. 7.10. An approach that combines adjustments to state variables as well as eruption parameters may perform better.

Response: It has been added in lines 8.14–8.15:

"Note that the forecast for both cases may be performed better by combining adjustments to state variables as well as eruption parameters."

12. Section 5 and Figure 5. seem a bit flawed, in that they are presented in terms of an "Improvement rate", but all I see is a difference, without and with assimilation. Either a comparison to actual measurements needs to be included (or if that was actually the case, this needs to be made clear), or these need to be presented quite differently in terms of "impact rate", rather than "improvement rate".

Response: I agree that "impact rate" is the proper term to present the difference of the assimilation impact between downwind and upwind directions. As recommended, we changed Section 5 and Figure 5 in terms of "impact rate", see lines 9.3–9.18:

"Firstly, the assimilation impact in the downwind and upwind directions is considered. For this investigation, eight big cities around the measurement route are selected (see Fig.

5a). They are Dortmund, Düsseldorf, Cologne in the downwind direction and Amsterdam, Rotterdam, Antwerp, Brussels in the upwind direction. The evaluation height is chosen at 3 km for relevant to the continental commercial aircraft safety. This is because most national and maybe some continental passenger flights are around this altitude, while intercontinental flights are at much higher altitude (Fu et al., 2015). The evaluation time is chosen to be 11:10 UTC 18 May 2010 when the assimilation process finishes. The concentrations of two major distal volcanic ash components, i.e., PM_{10} and $PM_{2.5}$ (Webley et al., 2012; Fu et al., 2015), are evaluated. Fig. 5b shows that results with assimilation is lower for both PM_{10} and $PM_{2.5}$ in all the selected cities. To quantify this impact on estimation of both ash components, an impact rate (IR) is introduced for quantification. The IR is defined as:

$$(IR)_p(i) = \frac{(SimuNoAssimi)_p(i) - (SimuAssimi)_p(i)}{(SimuNoAssimi)_p(i)},$$
(2)

where p means either PM_{10} or $PM_{2.5}$, i means index of selected cities. Moreover, (SimuNoAssimi)_p and (SimuAssimi)_p represent two simulations without or with assimilation. Using this equation, we can get the IR of all cities (see Fig. 5b). Based on the IR values, we find the assimilation impact in the downwind direction (Dortmund, Düsseldorf, Cologne and Luxembourg) are much more significant than those in the upwind direction (Amsterdam, Rotterdam, Antwerp and Brussels). This means after assimilation, the most significant impact on ash clouds is in the downwind direction where in this study it is mainly Germany (see assimilation impact areas in Fig. 3f). "

13. 6.16: "This tells us that assimilating aircraft measurements effectively reduce the error of the whole distal ash plume, not only at measurement locations." Comment: There could be some confusion in using the phrase "error of the whole distal ash plume", because there are not measurements to extract the model residual at all locations. Ensemble "spread" and "variance" are reduced.

Response: As recommended, it is now changed to "spread", see lines 7.19–7.20: "This tells us that assimilating aircraft measurements effectively reduce the ensemble spread of the whole distal ash plume".

14. 9.17: This is mainly because. . ." Comment: Do you know that "this is mainly because" or is it "probably because" or "possibly because" There was no detailed process study to ascertain all sources of error.

Response: I agree that here it should be "probably because", see lines 10.19–10.20: "This is probably because model parameters (e.g., the plume height) are uncertain and some processes are missing, for example, coagulation, evaporation, and resuspension."

15. 9.18: "Whereas. . .approach, the model's deficiencies can be compensated." Comment: Although they can be compensated, underlying deficiencies remain. The evidence lies in the failure to match cross validation data far away from assimilated measurements.

Response: We agree and changed this in lines 10.20–10.22:

"Analysis of the results showed that data assimilation approach is able to compensate the

model's deficiencies. However, the failure to match cross validation data far away from assimilated measurements indicate that although they can be compensated, the underlying deficiencies remain."

Corrections:

- 1. As suggested, "It has been shown. . ." is corrected to "It was shown" in lines 2.23-2.25: "It was shown using so-called twin experiments that ensemble-based data assimilation is in principle able to combine the aircraft in-situ measurements with a volcanic ash transport and dispersion model (VATDM) to make improvements on volcanic ash estimation near to the eruption."
- 2. "In this study..." is corrected to "In that study" in lines 2.25–2.27:

 "In that study, the focus was on the near-volcano areas where the uncertainties on plume height and mass eruption rate turned out to have a large influence on the estimates."
- 3. "due to assimilation" is changed to "After the assimilation" in lines 7.25–7.27: "After the assimilation process, the calculated volcanic ash concentrations in continental Europe have a lower concentration level (lower than 3000 μg m⁻³) and the changes on volcanic ash state is shown in a wide area."
- 4. 8.18: "Note that areas with ash concentration higher than this value are classified as No Fly Zone (NFZ) . . ." Comment: It would be beneficial to mention this fact before Page 6, Line 21, where the concentration is mentioned. Give the context first.

Now this is mentioned before the first concentration value is discussed, as in lines 7.24–7.27:

- "Note that areas with ash concentration higher than 4000 μ g m⁻³ are classified as No Fly Zone (NFZ) (EASA, 2011; Fu et al., 2015), which means aviation in these areas is not allowed. After the assimilation process, the calculated volcanic ash concentrations in continental Europe have a lower concentration level (lower than 3000 μ g m⁻³) and the changes on volcanic ash state is shown in a wide area."
- 5. The grammar problem in the sentence has been corrected, as in lines 8.4–8.5: "Based on the analysis above, significant differences between volcanic ash simulations without and with assimilation have been revealed".
- 6. The sentence is revised, see lines 8.27–8.29: "This shows that the forecast at these locations with assimilation is more accurate than the forecast without assimilation, so that the assimilated volcanic ash state (Fig. 3d) is a more accurate approximation to the real state of distal volcanic ash plume.".
- 7. 7.30: Please correct to read as follows: "benefit has an effect. Firstly. . . .impact in the downwind. . . .directions is considered." Page 8, Line 2: "PM10 . . . evaluated." Comment: This sentence is grammatically confusing.

The description is modified in lines 9.2–9.4:

"Next it will be investigated what is the benefit of the improved forecasts of distal ash plume on aviation advice, and also how much and how long the benefit has an effect. Firstly, the assimilation impact in the downwind and upwind directions is considered."

And in lines 9.8–9.10:

"The concentrations of two major distal volcanic ash components, i.e., PM_{10} and $PM_{2.5}$ (Webley et al., 2012; Fu et al., 2015), are evaluated."

8. 8.9: "we can find" to "we find" 8.8-13: Comment: There are three sentences in a row about wind direction. Consider revising.

As suggested, corrections are made in lines 9.14–9.18:

- "Based on the IR values, we find the assimilation impact in the downwind direction (Dortmund, Düsseldorf, Cologne and Luxembourg) are much more significant than those in the upwind direction (Amsterdam, Rotterdam, Antwerp and Brussels). This means after assimilation, the most significant impact on ash clouds is in the downwind direction where in this study it is mainly Germany (see assimilation impact areas in Fig. 3f)."
- 9. "measurements have the ability" is corrected to "measurements has the ability" and "assimilation has impact" is corrected to "assimilation has an impact" in lines 9.19–9.21: "The analysis above demonstrates that assimilating aircraft in-situ measurements has the ability to impact on regional volcanic ash clouds, especially in the downwind direction of the measurement route. It is also shown that assimilation has an impact on aviation advice."
- 10. "direction of distal" is corrected to "direction of the distal" in lines 9.21–9.22: "If there is no assimilation employed (see Fig. 3b), the volcanic ash concentration in the main transport direction of the distal ash plume reaches over 4000 μ g m⁻³."
- 11. 8.20-23: "This aviation. . .forbidden area." Comment: This sentence has a lot of clauses and is confusing. Consider revising.

As suggested, the sentence is revised as in lines 9.23–9.26:

"This aviation advice would shutdown flights in a large area. Because the Netherlands and Germany are important aviation hubs in Europe, imposing such a no-fly zone will prohibit flights entering continental Europe in the Eastern direction and subsequently lead to a huge economic loss."

12. 8.23-25: "In contrast. . .all lower than $3000\mu g~m^{-3}$." Comment: This sentence has a lot of clauses and is confusing. Consider revising.

As suggested, the sentence is revised as in lines 9.26–9.28:

- "In contrast, if based on the improved simulation after a continuous assimilation (Fig. 3d), the aviation advice would have been changed. The sky in almost the whole of Europe is open for commercial flights, because except in small parts of the Netherlands ash concentrations all over Europe are lower than 3000 μ g m⁻³."
- 13. 8.31: "changes of the forecast differences between without and with assimilation" Comment: This phrase is very confusing. Consider revising.

As suggested, the sentence is revised as in lines 9.33–9.34:

"3 time snapshots in Fig. 6a - Fig. 6c are chosen to show the forecast differences between

without and with assimilation. "

- 14. "Since, there are" is corrected to "Since there are" in line 9.34–10.1: "Since there are clear differences between the two cases, the assimilation impact will last at least one day."
- 15. Thanks for pointing out the difference between "detected" and "visible" or "noticeable". It is corrected in lines 10.2–10.3:

 "Only small differences are visible in the Northern part of Italy."
- 16. 8.34: "Actually we also examined the assimilation impact in the forecast of the next day, the difference was shown to be very small." Comment: This is a run-on sentence. Either add a conjunction or appropriate punctuation.

As suggested, the sentence is corrected in lines 10.3–10.4:

- "Actually we also examined the assimilation impact in the forecast of the next day and observed only very small differences."
- 17. 9.7-15: Combine these two paragraphs as they both relate to assimilation accuracy. The two paragraphs are now combined together, see lines 10.10–10.17.
- 18. 9.24: "This phenomenon is due to the wind direction and the transport process during the continuous assimilation." Comment: This line is copied verbatim from Page 8, Line 12.

 The sentence is removed, it is now only mentioned in the conclusion (lines 10.27–10.28).
- 19. 9.26: Please correct to read as follows: "The computer experiment. . ." 9.26-28: "Based on this. . .regional measurement tasks." Comment: Clean this up a bit to distinguish whether "the aircraft campaign" is a specific plan or any in the future that might be planned.

The sentences are revised in lines 10.29–10.31:

- "The computer experiment revealed that the time period of the improvement effect can be taken as 24 hours. Based on this result, we suggest to schedule an aircraft measurement campaign at a frequency of once per day. This can be used to provide guidelines for planning future regional measurement tasks."
- 20. "Actually these data also contain uncertainties which have an influence on ash cloud transport." is corrected to "These data also contain uncertainties that influence ash cloud transport." in line 10.33.
- 21. 9.30-31: "In future work, . . . into account." Comment: This sentence needs editing for grammar.

It is corrected in lines 10.33–11.1:

"In future work, in order to further improve the accuracy of ash forecasting, uncertainties in the meteorological data such as wind speed should also be considered."

The revised manuscript starts from next page.

Model-based aviation advice on distal volcanic ash clouds by assimilating aircraft in-situ measurements

G. Fu¹, A.W. Heemink¹, S. Lu¹, A.J. Segers², K. Weber³, and H.X. Lin¹

Abstract. The forecast accuracy of distal volcanic ash clouds is important for providing valid aviation advice during volcanic ash eruption. However, because the distal part of volcanic ash plume is far from the volcano, the influence of eruption information on this part becomes rather indirect and uncertain, resulting in inaccurate volcanic ash forecasts in these distal areas. In our approach, we use real-life aircraft in-situ observations, measured in the North-West part of Germany during the 2010 Eyjafjallajökull eruption, in an ensemble-based data assimilation system combined with a volcanic ash transport model to investigate the potential improvement on the forecast accuracy with regard to the distal volcanic ash plume. We show that the error of the analyzed volcanic ash state can be significantly reduced through assimilating real-life in-situ measurements. After a continuous assimilation, it is shown that the aviation advice for Germany, the Netherlands and Belgium can be significantly improved. We suggest that with suitable aircrafts measuring once per day across the distal volcanic ash plume, the description and prediction of volcanic ash clouds in these areas can be greatly improved.

1 Introduction

Ash produced during explosive volcanic eruptions can cause serious impacts close to the volcano as well as at great distances (Melville, 1986). Turbine engines are particularly threatened by ingestion of airborne ash, and aircraft surfaces may be subject to abrasion and in the longer-term corrosion (Casadevall, 1994). For example, the sudden eruption of the ice-capped Eyjaf-jallajökull volcano at 1666 m height in Iceland from 14 April to 23 May 2010, had caused an unprecedented closure of the European and North Atlantic airspace resulting in a huge global economic loss (Bonadonna et al., 2012). Due to the huge impacts on aviation community, a lot of research has been initiated on how to efficiently reduce these aviation impacts starting with improving the accuracy of volcanic ash forecast after eruption onset (Gudmundsson et al., 2012; Eliasson et al., 2011; Schumann et al., 2011). Currently a lot of approaches, employing satellite-based (Prata and Prata, 2012; Stohl et al., 2011; Lu et al., 2016) or ground-based (Emeis et al., 2011) measurements, focus on improving the estimation of Eruption Source Parameters (ESPs) such as plume height and mass eruption rate. These are very important for a good estimation of volcanic ash emission. However, for the volcanic ash plume far from the volcano which could be very important for local aviation, more accurate ESPs alone will not be very useful. This is mainly because the influence of ESPs on the volcanic ash plume

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becomes weaker as the distance to the volcano increases. Therefore, additional observation data, e.g., direct observations of distal volcanic ash plume must be employed to improve the aviation advice over continental Europe.

Ensemble-based data assimilation, which refers to the (quasi-) continuous use of the direct measurements to create accurate initial conditions for model runs (Zehner, 2010), is one of the most commonly used approaches for real-time forecasting problems (Evensen, 2009). In each assimilation step, a forecast from the previous model simulation is used as a first guess, then this forecast is modified to be more in agreement with the available observations. This approach is very effective for regional forecasting. For employment of ensemble-based data assimilation, in-situ measurements are the optimal type of observations (Evensen, 2009). Although satellite measurements are considered as the most commonly used volcanic ash observations because of their large detection domain and long-time continuous output data, they are not directly suited for data assimilation systems due to their insufficient vertical resolution (Bocquet et al., 2015). Note that, some satellites can provide very detailed vertical information on plumes (e.g., Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar measurements) but are thus spatially sparse (Winker et al., 2012). Fortunately, the in-situ volcanic ash state variables can be accurately measured nowadays by means of airborne observations of volcanic ash (Weber et al., 2012). These aircraft-based measurements can be obtained close to the eruption plume, which are probably the most direct volcanic ash observations possible. They have some unique advantages. First, aircraft data is usually obtained from optical particle counters, thus real-time particle concentration observation can be directly measured (Weber et al., 2012). Second, the aircraft measurement is in-situ which can be compared directly to a 3-dimensional model state variable (e.g., concentration). Whereas other measurements such as satellite data and LIDAR data observe optical properties that contain indirect information about concentrations and require further modeling and assumptions to translate into concentration estimates. Note that in this study volcanic ash state refers to the whole volcanic ash plume, while volcanic ash state variables represent point-to-point volcanic ash concentrations inside the ash plume. Third, an aircraft is flexible in flight route to follow the ash clouds and to always obtain the most relevant volcanic ash concentration.

Recently in the application of volcanic ash transport, the benefit of aircraft in-situ observations in an Ensemble Kalman Filter (EnKF) system has been studied (Fu et al., 2015). It was shown using so-called twin experiments that ensemble-based data assimilation is in principle able to combine the aircraft in-situ measurements with a volcanic ash transport and dispersion model (VATDM) to make improvements on volcanic ash estimation near to the eruption. In that study, the focus was on the near-volcano areas where the uncertainties on plume height and mass eruption rate turned out to have a large influence on the estimates. However, for distal volcanic ash plume, these eruption parameters hardly improve the forecasts over a long distance. A larger mass eruption rate may cause the distal volcanic ash plume to spread stronger and wider after a long time period. But this potential effect can be significantly influenced or even canceled out by a combination of a number of elusive physical factors over a long time period such as wind speed and direction. Thus the results on near-volcano areas cannot be directly employed for far-volcano regions, e.g., central Europe in the case of an Iceland eruption. In addition, the aircraft in-situ measurements used in the previous studies were self-designed (artificial) based on model simulations from which actual conditions might differ significantly. For example, using data of a period of 10 hours by an aircraft gives accurate assimilation results. But in practical situations, a continuous aircraft measurement mission is at most 3 or 4 hours, thus it is still uncertain whether the assimilation can produce significant effect with a shorter measurement mission. Therefore, in case of real-life aircraft in-situ

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measurements, it remains unknown whether the ensemble-based data assimilation still has significant improvements on the distal part of volcanic ash clouds and how long the influence will last. The answers of these questions will lead us to a solution for evaluating distal volcanic ash clouds and further provide accurate aviation advice. This study aims at investigating these questions. Note that, the term real-life aircraft measurements in this study refer to authentic measurements obtained by real aircrafts. This is to distinguish the artificial aircraft measurements as used in (Fu et al., 2015). Another term distal volcanic ash plume is used to clarify the study focuses on volcanic ash forecasts far from the volcano, i.e. continental Europe in this study.

This paper is organized as follows. Section 2 introduces the LOTOS-EUROS model, aircraft in-situ measurements, and ensemble-based data assimilation methods used in this study. The assimilation experiments on distal volcanic ash clouds are specified in Section 3. Section 4 validates the performance of real-life data assimilation. Section 5 contains the benefit of the improved forecasts of distal ash plume on aviation advice, and also how much and how long the benefit has effect. Finally, the last section summarizes the concluding remarks of our research.

2 Materials and methods for volcanic ash assimilation

2.1 The LOTOS-EUROS model for volcanic ash transport

In this study, we use the LOTOS-EUROS model (Schaap et al., 2008) to simulate volcanic ash transport and dispersion, which is an operational air-quality model, used for daily air quality forecasts over Europe (Curier et al., 2012), focusing on ozone, nitrogen oxides, and particular matter. The model uses the off-line meteorological data produced by European Center for Medium-Range Weather Forecasts (ECMWF). The model is used to produce volcanic ash simulations in a timely and useful manner for forecasting. To describe a volcanic eruption in LOTOS-EUROS model, Eruption Source Parameters (ESP) such as plume height (PH), mass eruption rate (MER), vertical mass distribution (VMD) and particle size distribution (PSD) are needed. Typically ESPs for different volcanoes are provided as a look up table (Mastin et al., 2009).

The input parameter PH in LOTOS-EUROS is taken from hourly based Icelandic Meteorological Office (IMO) plume height detection and usually the uncertainty of PH is taken as 20 % (Bonadonna and Costa, 2013). For VMD, large explosive volcanic plumes have a typical 'umbrella' shaped vertical distribution (Sparks et al., 1997) and as such this 'umbrella' shaped VMD is adapted into LOTOS-EUROS (Fu et al., 2015; Lu et al., 2016). The PSD in LOTOS-EUROS is defined in the ESP type S2 (see definations in Mastin et al. (2009)), in which the mass fraction of erupted debris finer than 63 μ m is 0.4. For the S2 type eruption, Durant and Rose (2009) provides the base for the PSD from their analysis of the 1992 Crater Peak, Mount Spurr event. Hence, Table 1 provides the ash distribution based on their analysis and is used by LOTOS-EUROS. Another input parameter MER is very hard to measure for an explosive onsetting volcano. Usually it is calculated from the plume height. Mastin et al. (2009) studied the relation between these parameters and concluded that an empirical relationship between plume height PH(km) and mass eruption rate MER (kg s⁻¹) is

$$PH = 2.00V^{0.241}, \quad and \quad \frac{V}{MER} = \frac{1.5e^3}{4.0e^6}.$$
 (1)

where $V(m^3 s^{-1})$ represents the volumetric flow rate.

Recently, the LOTOS-EUROS model has been evaluated as an appropriate volcanic ash transport model (Fu et al., 2015) (Fig. 1a), where the volcanic ash concentrations are described by 6 aerosol tracers including PM₁₀ and PM_{2.5} (corresponding to vash_5 and vash_6 in Table 1) to model the transport process. The physical processes that are relevant for volcanic ash are similar as those that apply for mineral dust, e.g., advective transport and diffusion, deposition, coagulation, sedimentation, and resuspension (Langmann, 2013). Where the transport is determined by the wind fields that could be regarded as rather well known, the other processes deposition and sedimentation processes are rather uncertain. The parameterizations for the later processes involve assumptions on the particle distribution for example, which is difficult to summarize in a few numbers of bins. These processes act on the distribution of the total ash mass over the modes (particle sizes) and the total mass load; one could therefore state that almost everything in the description of an ash cloud is uncertain, except for the cloud shape and position. The processes included in this study are transport, sedimentation, and wet- and dry-deposition, where the relevant properties such as average particle size (Table 1) are implemented following Zhang (2001). Processes that are missing yet are for example coagulation, evaporation, and resuspension, which might be considered in future when appropriate observations are available to constrain them, for example sedimentation amounts.

2.2 Real-life aircraft in-situ measurements

During the period of the Eyjafjallajökull eruption in April – May, 2010, the outskirts of the eruption plume were entered directly by research flights, delivering most direct measurements within the eruption plume during this eruptive event (Weber et al., 2012). All of the measurement flights (Fig. 1b) were equipped with optical particle counters (OPC) for in-situ measurements. Real-time monitoring of the particle concentrations was possible during the flights and in-situ measurements from the eruption plume were obtained with high time- and spatial-resolution. Through a direct laboratory calibration experiment, in which the mass concentration obtained with the OPC was compared with the absolute mass concentration gathered on a gravimetric filter, the standard deviation between the gravimetric measurement and the OPC was estimated at 10% (Weber et al., 2010) which can be taken as the instrumental error for this type of measurements in well calibrated cases. The total measurement error not only contains the instrumental error, but also includes an estimate of the model representation error (Fu et al., 2015).

In this study we used the aircraft-based measurements taken by one measurement flight on 18 May, 2010 performed by the group Environmental Measurement Techniques at Duesseldorf university of Applied Sciences. The measurements took place in the North-West part of Germany including the border between the Netherlands and Germany, see Fig. 1b (the black rectangular area in Fig. 1a). The aircraft took off from the airfield "Schwarze Heide" in the Northern part of the Rhein-Ruhr area, headed along the Dutch border in the direction of the North Sea, continued towards Hamburg and then returned to the airfield. Along the route, concentrations of PM_{10} and $PM_{2.5}$ were measured, see Fig. 1c and 1d.

30 2.3 The Ensemble Kalman Filter

The ensemble-based data assimilation technique used in this study is an Ensemble Kalman Filter technique (EnKF). Apart from the original formulation (Evensen, 1994), other formulations have been introduced such as the Ensemble Kalman Smoother (EnKS) (Evensen and van Leeuwen, 2000), Ensemble Square Root Filter (EnSR) (Evensen, 2004), Reduced Rank Square Root

Filter (RRSQRT) (Verlaan and Heemink, 1997), etc. Ensemble-based assimilation is easy to implement, suitable for real-time estimation of concentrations and has a very general statistical formulation.

The Ensemble Kalman Filter essentially is a Monte Carlo ensemble-based method (Evensen, 2003), based on the representation of the probability density of the state estimate in an ensemble of N state, $\xi_1, \xi_2, \dots, \xi_N$. Each ensemble member is assumed to be a single sample out of a distribution of the true state. The number of required ensemble members depends on the complexity of the probability density function to be captured, which is usually determined by the nonlinearity of the model and the description of the involved uncertainties. For volcanic ash assimilation, an ensemble size of 50 is considered acceptable in terms of accuracy while keeping computation time within reach (Fu et al., 2015). For application of the filter algorithm to the LOTOS-EUROS model, in the first step of this algorithm an ensemble of N volcanic ash state $\xi^a(0)$ is generated to represent the uncertainty in the initial condition $\mathbf{x}(0)$. In the second step, the forecast step, the LOTOS-EUROS model (with stochastic plume height) propagates the ensemble members from the time k-1 to k:

$$\xi_i^f(k) = M_{k-1}(\xi_i^a(k-1)). \tag{2}$$

The state-space operator M_{k-1} describes the time evolution from the time k-1 to k of the state vector which contains the ash concentrations in the model grid boxes. The filter state is a stochastic distribution with mean \mathbf{x}^f and covariance \mathbf{P}^f following:

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$$\mathbf{x}^f(k) = [\sum_{j=1}^N \xi_j^f(k)]/N,$$
 (3)

$$\mathbf{L}^f(k) = [\xi_1^f(k) - \mathbf{x}^f(k), \cdots, \xi_n^f(k) - \mathbf{x}^f(k)],\tag{4}$$

$$\mathbf{P}^f(k) = [\mathbf{L}^f(k)\mathbf{L}^f(k)^T]/(N-1). \tag{5}$$

The observational network is defined by the observation operator H that maps state vector \mathbf{x} to observation space \mathbf{y} :

$$\mathbf{y}(k) = H_k(\mathbf{x}(k)) + \mathbf{v}(k), \quad \mathbf{v}(k) \sim N(0, \mathbf{R}),$$
 (6)

where the observation error \mathbf{v} is drawn from a Gaussian distribution with zero mean and covariance matrix \mathbf{R} . Here, \mathbf{y} contains aircraft in-situ measurements of ash concentration and \mathbf{R} is filled in a diagonal matrix with the square of the standard deviation of y. The operator H then selects the grid cell in x that corresponds to the observation location. When measurements become available, the ensemble members are updated in the analysis step using the Kalman gain:

$$\mathbf{K}(k) = \mathbf{P}^f(k)\mathbf{H}(k)^T[\mathbf{H}(k)\mathbf{P}^f(k)\mathbf{H}(k)^T + \mathbf{R}]^{-1},\tag{7}$$

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$$\xi_j^a(k) = \xi_j^f(k) + \mathbf{K}(k)[\mathbf{y}(k) - \mathbf{H}(k)\xi_j^f(k) + \mathbf{v}_j(k)],$$
 (8)

where \mathbf{v}_j represents realizations of the observation error v.

3 Sequentially assimilating real-life aircraft in-situ measurements for distal volcanic ash clouds

3.1 Experimental Setup

As described in Section 2, an Ensemble Kalman Filter (EnKF) is used in this study to assimilate real-life aircraft in-situ observations. The LOTOS-EUROS model run starts at 09:00 (UTC) 14 April 2010 by considering a zero initial condition, equivalent to an assumption of "no ash load yet". As the model state changes with the time in the numerical simulation (the time step of model run is 10 minutes (Fu et al., 2015)), the model result from the previous time step is taken as the initial state for the next time step. When the model run is at 09:40 (UTC) 18 May, the volcanic ash state gets continuously modified by the data assimilation process through combining real-life aircraft-based measurements taken along the Dutch border until the time 11:10 (UTC) 18 May. The specification of uncertainties is essential for a successful data assimilation in this study. The plume height (PH) is set to be the hourly plume height detection data and its uncertainty is estimated to be 20 % (Bonadonna and Costa, 2013). The stochastic plume height is assumed to be temporally correlated and the correlation parameter τ is set to be 1 hour (Fu et al., 2015). Thus, the PH noise (N_{ph}) at two times $(t_1$ and $t_2)$ has the relation (Evensen, 2009) of $\mathbb{E}[N_{ph}(t_1) \cdot N_{ph}(t_2)] = e^{\frac{-|t_1-t_2|}{\tau}}$, where \mathbb{E} represents the mathematical expectation.

To assimilate measurements in a simulation model, it is necessary to quantify the model representation error. The model representation error is the discrepancy between the quantity that instrument observes, and what the model value represents. Concentration values are defined on discrete grids with a finite resolution at discrete time steps. A measurement location usually does not coincide with the grid point where the concentration value is defined. The spatial resolution of the model used in this study is around $12 \text{ km} \times 12 \text{ km} \times 1 \text{ km}$, therefore the volume of one grid-box is about 150 km^3 . Through model processing, the concentration of one grid-box represents an average value for this grid-box, while one aircraft in-situ measurement is a point value in a 3 dimensional field. In this study, we choose the in-situ measurement corresponding to the grid-box average value. This approximation makes sense only when two assimilated measurements are positioned in two different grid-boxes. Considering the aircraft speed of $100\text{-}200 \text{ km} \text{ h}^{-1}$ and the LOTOS-EUROS horizontal and vertical resolution, a 10 minutes assimilation frequency is chosen to guarantee different assimilated measurements are in different grid-boxes. The observation therefore almost corresponds to one model state variable in a grid-box, which means the representation error of the model is probably small. For the moment we will therefore not explicitly specify a model representation error, but implicitly assume that it is zero. The total observation representation error, defined as the sum of the instrumental error and the model representation error, is taken as 10% in this study.

Since the real-life measurements of the PM_{10} and $PM_{2.5}$ concentrations are available and the uncertainties of this type of measurements are approximately known, ensemble-based data assimilation can be used to combine them with the LOTOS-EUROS model to reconstruct optimal estimates.

3.2 Evaluation of real-life data assimilation

It is first examined how the data assimilation actually performs in the system. Fig. 2 shows the measurements, the mean of the ensemble members, as well as the forecast and the analysis of some of the ensembles. From the estimation of both volcanic ash

components PM_{10} and $PM_{2.5}$ (Fig. 2a and Fig. 2b), we find the forecast mean largely overestimates the measurement at every time step, but the overestimation vanishes by the assimilation process. Instead, the analysis mean consistently approximates the measurements with a high accuracy. This result illustrates that the assimilation at the measurement location is able to approximate the observed values and also solves the problem of overestimation. Moreover, at the measurement location, the spread in the analysis ensembles is much smaller than that in the forecast ensembles, which means the error variance of analysis value at the measurement locations is significantly reduced through the use of assimilation. This is because aircraft in-situ measurements are of high accuracy. Note that, Fig. 2a and Fig. 2b only show the ensemble mean and the spread of the ensembles at the measuring locations. However, we also want to know how much impact assimilation of aircraft in-situ measurements can have on a wide area of the distal volcanic ash plume. If the impact is only limited to the measurement locations or only a small nearby area of ash plume, there will be no significant improvement in terms of aviation advice because flights need a rather large domain for safety guarantee.

In order to further investigate this effect, we first show the ensembles of uncertain volcanic ash simulations (see Fig. 2c and Fig. 2d), which correspond to the ash distribution at 11:10 (UTC) 18 May 2010 before and after assimilation. Note that, this study focuses on the distal volcanic ash plume, thus only the area of the whole plume marked as red rectangular as shown in Fig. 1a is of interest. Without loss of generality, ensemble member 8 and 21 are chosen for illustrating the ensemble spread of the distal ash plume. Through comparing different ensemble members with respect to the forecast at 11:10 (Fig. 2c), the ensemble forecast member 21 is shown to be very different from the member 8 in almost all the complete distal plume, thus the large error of the forecast is not only at measurement locations, but also in a large area around the measurement. Compared to the forecast, ensembles of analysis state (Fig. 2d) show no large differences across the entire domain of interest. This tells us that assimilating aircraft measurements effectively reduce the ensemble spread of the whole distal ash plume, which is a sign of consistency to the measurement locations. Next, we investigate the assimilation impact on the ensemble mean over the distal volcanic ash plume. Fig. 3 shows examples of the mean at 09:40 and 11:10 (UTC) 18 May with and without assimilating aircraft measurements. Compared to the case without assimilation at 11:10 (Fig. 3b), large differences can be observed in the simulation results with a continuous assimilation (Fig. 3d). Note that areas with ash concentration higher than 4000 $\mu g \text{ m}^{-3}$ are classified as No Fly Zone (NFZ) (EASA, 2011; Fu et al., 2015), which means aviation in these areas is not allowed. After the assimilation process, the calculated volcanic ash concentrations in continental Europe have a lower concentration level (lower than 3000 $\mu g \text{ m}^{-3}$) and the changes on volcanic ash state is shown in a wide area. This is because the volcanic ash state variables become dependent and correlated due to the transport process (advection and diffusion) and the temporal correlation of emission (Fu et al., 2015). Thus in a fairly large domain, the state change at measurement location also influences state variables in surrounding areas. Note that the differences between with and without assimilation are not obtained in one-time, but step by step with assimilating measurements over a period of one and a half hours from 09:40 to 11:10. This can be seen from the assimilation results at 09:40 and 11:10 (Fig. 3c and Fig. 3d) where clear differences (Fig. 3e and Fig. 3f) between the two times can be observed and the effect of the assimilation at 09:40 is less pronounced than at 11:10. This shows that after a continuous assimilation of aircraft measurements, the differences with the original simulation are the result of an accumulation

of all previous assimilation effects. This analysis also tells us that all the assimilation steps are important for the final result and that only using one or two measurements does not produce accurate results.

4 Validation of assimilation performance

Based on the analysis above, significant differences between volcanic ash simulations without and with assimilation have been revealed. To examine whether the assimilated results are indeed more accurate than the model results, a further validation must be conducted. Fig. 4 shows the difference of a number of forecasted volcanic ash plumes with and without assimilation. The basic idea of this validation is to compare future in-situ measurements with the forecast of the volcanic ash plumes initiated with Fig. 3c and Fig. 3d. Because all the other settings in the system are the same, a better forecast will be due to a more accurate initial state. We use the measurements from 09:30 to 11:10 along the Dutch border to produce the assimilated results, then we validate the results using another set of aircraft in-situ measurements in the downwind direction taken from 12:30 to 15:00 UTC 18 May 2010 (see Fig. 1b and 1d). With different initialization, the forecast of volcanic ash concentration at 15:00 shows large differences. The forecast after assimilation (see Fig. 4b, lower than 3000 μ g m⁻³ in the downwind direction of the measurement track) is much smaller than that without assimilation (Fig. 4a, higher than 4000 μ g m⁻³ in some areas of continental Europe). Note that the forecast for both cases may be performed better by combining adjustments to state variables as well as eruption parameters.

The detailed ash concentrations of two forecasts are compared with measurements in Fig. 4c and 4d. Both forecasts are shown to overestimate the measurements. This is in accordance with practical experience that volcanic ash simulations often overestimate the truth to guarantee a safe aviation advice. This is because in practice, until carefully designed engine performance tests are conducted in realistic volcanic ash cloud conditions, a cautious approach (overestimation) to advising commercial jet operations in airspace affected by volcanic ash is recommended (Prata and Prata, 2012). Furthermore, we can also see that at some times between 13:20 and 14:00, there are no obvious differences between the two forecasts. This is because the validation measurements during this period (red points in Fig. 4b) are far away from the measurement track used in assimilation. Although assimilating aircraft in-situ measurements has impact on a large domain, the influence has still not reached the validation locations between 13:20 and 14:00. Therefore, at these locations, the volcanic ash state variables are not influenced by the assimilation process so that the two forecasts do not show clear differences. But in all the other time steps (from 12:30 to 13:20 and from 14:00 to 15:00), the forcast with assimilation is much closer to the measurements than the forecast without assimilation, and also that the overestimation is significantly reduced using assimilation. This shows that the forecast at these locations with assimilation is more accurate than the forecast without assimilation, so that the assimilated volcanic ash state (Fig. 3d) is a more accurate approximation to the real state of distal volcanic ash plume. In addition, we conclude that the assimilation process performs well in combining with the LOTOS-EUROS transport model with real-life measurements. Note that we find that for some locations the forecast is improved, but in other it is not. Thus, significantly more observations are required to completely constrain the ash plume across the domain. Extensive aircraft observations, or some combination of aircraft measurements as used here with remote sensing data may be useful.

5 Assimilation benefit for aviation community

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Next it will be investigated what is the benefit of the improved forecasts of distal ash plume on aviation advice, and also how much and how long the benefit has an effect. Firstly, the assimilation impact in the downwind and upwind directions is considered. For this investigation, eight big cities around the measurement route are selected (see Fig. 5a). They are Dortmund, Düsseldorf, Cologne in the downwind direction and Amsterdam, Rotterdam, Antwerp, Brussels in the upwind direction. The evaluation height is chosen at 3 km for relevant to the continental commercial aircraft safety. This is because most national and maybe some continental passenger flights are around this altitude, while intercontinental flights are at much higher altitude (Fu et al., 2015). The evaluation time is chosen to be 11:10 UTC 18 May 2010 when the assimilation process finishes. The concentrations of two major distal volcanic ash components, i.e., PM₁₀ and PM_{2.5} (Webley et al., 2012; Fu et al., 2015), are evaluated. Fig. 5b shows that results with assimilation is lower for both PM₁₀ and PM_{2.5} in all the selected cities. To quantify this impact on estimation of both ash components, an impact rate (IR) is introduced for quantification. The IR is defined as:

$$(IR)_{p}(i) = \frac{(SimuNoAssimi)_{p}(i) - (SimuAssimi)_{p}(i)}{(SimuNoAssimi)_{p}(i)},$$
(9)

where p means either PM_{10} or $PM_{2.5}$, i means index of selected cities. Moreover, $(SimuNoAssimi)_p$ and $(SimuAssimi)_p$ represent two simulations without or with assimilation. Using this equation, we can get the IR of all cities (see Fig. 5b). Based on the IR values, we find the assimilation impact in the downwind direction (Dortmund, Düsseldorf, Cologne and Luxembourg) are much more significant than those in the upwind direction (Amsterdam, Antwerp and Brussels). This means after assimilation, the most significant impact on ash clouds is in the downwind direction where in this study it is mainly Germany (see assimilation impact areas in Fig. 3f).

The analysis above demonstrates that assimilating aircraft in-situ measurements has the ability to impact on regional volcanic ash clouds, especially in the downwind direction of the measurement route. It is also shown that assimilation has an impact on aviation advice. If there is no assimilation employed (see Fig. 3b), the volcanic ash concentration in the main transport direction of the distal ash plume reaches over $4000 \mu g m^{-3}$. Thus, only relying on simulation results, the aviation advice on continental Europe is that the sky above the North Sea, the Netherlands and the western part of Germany is forbidden for flights. This aviation advice would shutdown flights in a large area. Because the Netherlands and Germany are important aviation hubs in Europe, imposing such a no-fly zone will prohibit flights entering continental Europe in the Eastern direction and subsequently lead to a huge economic loss. In contrast, if based on the improved simulation after a continuous assimilation (Fig. 3d), the aviation advice would have been changed. The sky in almost the whole of Europe is open for commercial flights, because except in small parts of the Netherlands ash concentrations all over Europe are lower than 3000 $\mu g m^{-3}$. This illustrates that the accuracy of aviation advice and the NFZ area can significantly benefit from the ensemble-based data assimilation process.

Another question is how long the effect of improvement by assimilating aircraft measurements will last? The answer of this will provide us guidance on how often aircraft measuring should be performed. For investigating the time period of the assimilation impact, the volcanic ash plume is forecasted one day (Fig. 6) starting at 11:10 (UTC) 18 May 2010. Without loss of generality, PM_{10} is chosen to analyze the forecast performance. 3 time snapshots in Fig. 6a – Fig. 6c are chosen to show the forecast differences between without and with assimilation. Since there are clear differences between the two cases,

the assimilation impact will last at least one day. When forecasting 24 hours (Fig. 6c), differences still can be observed, but the impact of assimilation is obviously getting much smaller (Fig. 6a and Fig. 6b). Only small differences are visible in the Northern part of Italy. Actually we also examined the assimilation impact in the forecast of the next day and observed only very small differences. Therefore, the time period of the assimilation impact of this case study can be taken as 24 hours. From this analysis, we suggest the frequency of the measurement campaign to be once per day. This study can be used to provide guidelines for an optimal flight schedule in regional measurement tasks. Note that the impact time investigated is based on the meteorological information in distal volcanic ash plume during the period considered in this study. For other cases, the duration of effective assimilation could be differed.

6 Conclusions

10 In this study, aircraft in-situ measurements in distal volcanic ash clouds were assimilated in the LOTOS-EUROS model. During a continuous assimilation, the error of the analyzed volcanic ash state was significantly reduced through assimilating real-life in-situ measurements. The improved volcanic ash state after assimilation are the result of an accumulation of all previous assimilation effects. It was shown that all the assimilation steps were important for the final result and using only one or two measurements can not produce accurate results. To examine whether the assimilated volcanic ash state were indeed more accurate than the conventional simulation, a validation with future in-situ measurements was conducted. The forecast with assimilation was shown more accurate than the conventional forecast without assimilation. It also concluded that the assimilation process performed well in combining with the LOTOS-EUROS transport model with real-life measurements.

The validation results also revealed that with the transport models alone, it is difficult to accurately model volcanic ash movements. This is probably because model parameters (e.g., the plume height) are uncertain and some processes are missing, for example, coagulation, evaporation, and resuspension. Analysis of the results showed that data assimilation approach is able to compensate the model's deficiencies. However, the failure to match cross validation data far away from assimilated measurements indicate that although they can be compensated, the underlying deficiencies remain. Aircraft in-situ measurements have a high accuracy and plays an important role to a successful data assimilation. The aircraft can enter the plume to selectively obtain observations, so that the measurements are in-situ and optimal for the ensemble-based data assimilation methodology.

Investigation was also carried out on the benefit of the improved forecasts of distal ash plume on aviation advice. We found that after assimilation, the most significant improvements on distal ash clouds are in the downwind direction where in this study it is mainly Germany. This phenomenon is due to the wind direction and the transport process during the continuous assimilation. We investigated the accuracy of aviation advice can significantly benefit from the ensemble-based data assimilation process. The computer experiment revealed that the time period of the improvement effect can be taken as 24 hours. Based on this result, we suggest to schedule an aircraft measurement campaign at a frequency of once per day. This can be used to provide guidelines for planning future regional measurement tasks.

In this study, we applied an off-line approach for model running and simply used the deterministic meteorological input data. These data also contain uncertainties that influence ash cloud transport. In future work, in order to further improve the accuracy of ash forecasting, uncertainties in the meteorological data such as wind speed should also be considered. In this study, only aircraft in-situ measurements are used in a data assimilation system. We may expect that with other types of measurements (e.g., satellite-based or LIDAR-based) together, the assimilation results will be more practical since the aircraft measurements cannot be always obtained. However, for this multi-observation data assimilation, other problems need to be first considered such as insufficient vertical resolution in satellite data. This is a difficult aspect for assimilating these data in a three dimensional model, and will be investigated in our future work.

Author contributions. All authors participated in the design and analysis of the assimilation experiment. G.F., A.J.S. and S.L. carried out the LOTOS-EUROS modeling volcanic ash transport. K.W. validated real-life aircraft in-situ measurements and provided them for the data assimilation experiment. G.F., H.X.L. and A.W.H. analyzed the results and wrote the paper with contributions from all co-authors.

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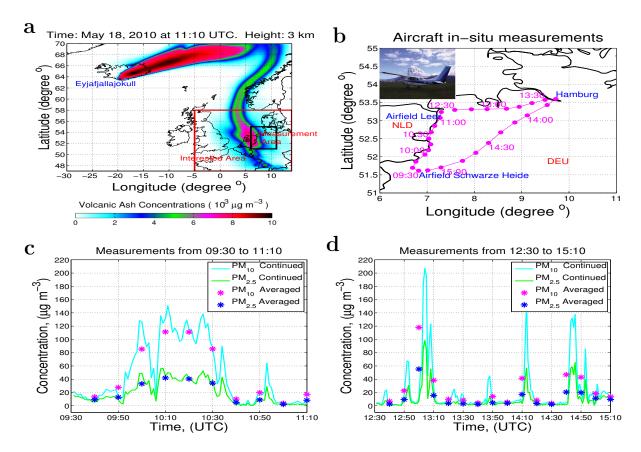


Figure 1. Aircraft in-situ measurements of distal volcanic ash plume. a, The LOTOS-EUROS simulation of volcanic ash plume at 11:10 (UTC), 18 May 2010. b, Measuring aircraft flight route on 18 May 2010. c, PM_{10} and $PM_{2.5}$ measurements from 09:30 to 11:10 (UTC). d, PM_{10} and $PM_{2.5}$ measurements from 12:30 to 15:10 (UTC). In c and d, the curves show the values of PM_{10} and $PM_{2.5}$ measured at a frequency of every 6 seconds. The values marked with a star are the averaged PM_{10} and $PM_{2.5}$ (average every 10 minutes) which are used in the LOTOS-EUROS model in accordance with the model simulation step (10 minutes).

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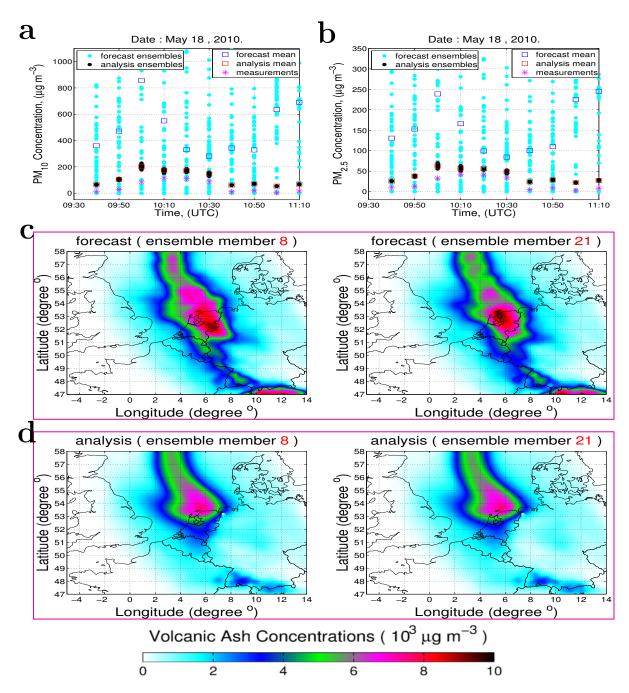


Figure 2. Assimilation effect at measurement locations from 09:40 to 11:10 (UTC), 18 May, 2010. a–b, Volcanic ash concentrations of PM₁₀ and PM_{2.5}. c–d, Volcanic ash ensemble state (ensemble member 8 and 21) of forecast and analysis at 11:10. The measurements, ensembles and mean of forecast and analysis are shown in a and b. In c and d, the area of interest is marked as red rectangular in Fig. 1a.

Table 1. Volcanic ash particle size distribution and ash bins property for LOTOS-EUROS model simulation.

Bins	Particle Diameter	Percent of Mass	Average Particle Size (μm)
vash_1	250 to 2000 $\mu \mathrm{m}$	29	1125.00
vash_2	63 to 250 $\mu \mathrm{m}$	31	156.50
vash_3	30 to 63 $\mu \mathrm{m}$	12	46.50
vash_4	10 to $30~\mu\mathrm{m}$	18	20.00
vash_5	2.5 to 10 $\mu \mathrm{m}$	8	6.25
vash_6	0.0 to 2.5 $\mu \mathrm{m}$	2	1.25

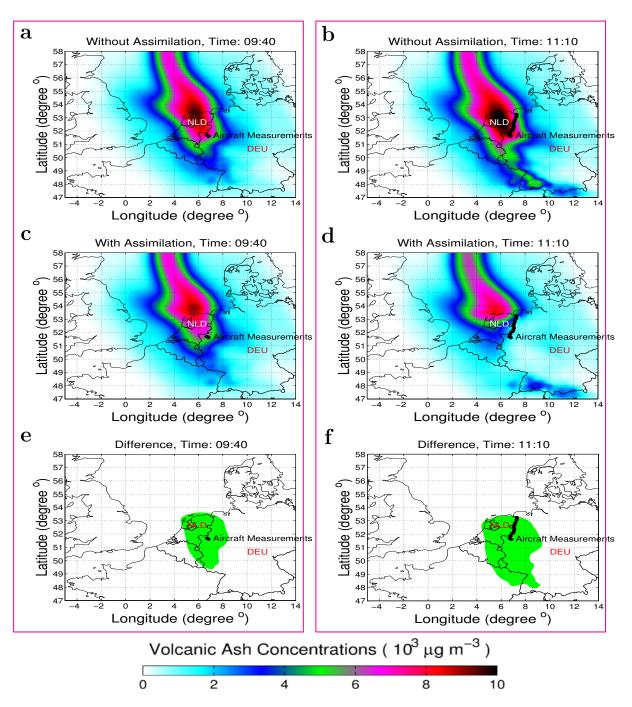


Figure 3. Comparison with and without assimilating aircraft in-situ measurements on 18 May 2010. a and b, Simulation results without assimilation at 09:40 and 11:10 UTC. c and d, Simulation results with assimilation at 09:40 and 11:10 UTC. c, Differences of a and c. f, Differences of b and d. The differences are in absolute values which are obtained by numerically subtracting the values between a and c, or b and d. e and f represent the areas where the assimilation has effect.

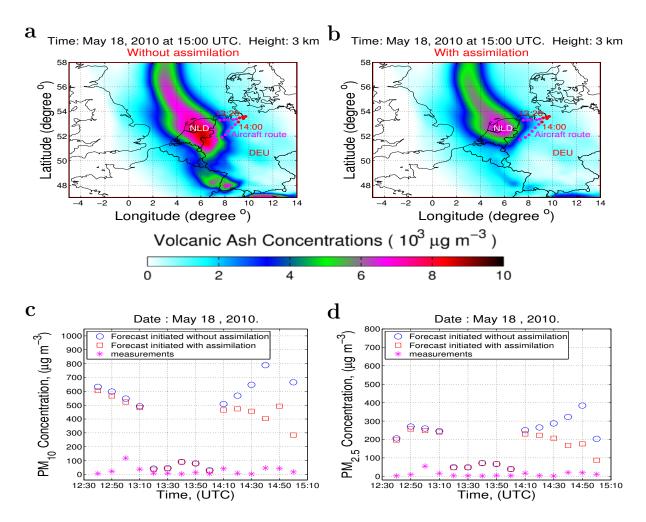


Figure 4. Forecast at 15:00 (UTC) 18 May 2010 with different initial conditions for the volcanic ash state. a, Forecast initiated with (Fig. 3c). b, Forecast initiated with (Fig. 3d). c, PM₁₀ concentration from 12:40 to 15:00. d, PM_{2.5} concentration from 12:40 to 15:00.

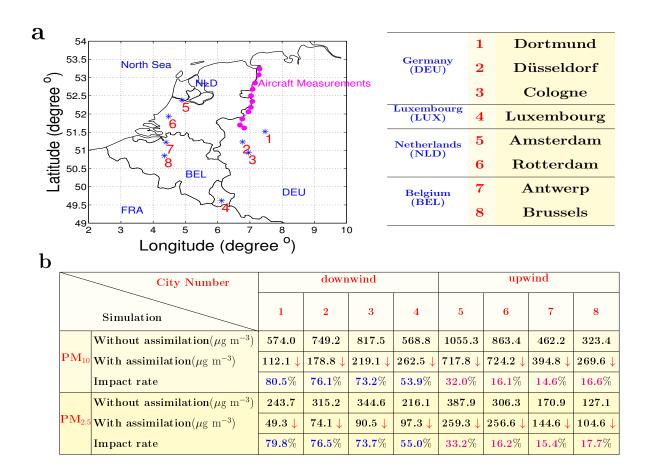


Figure 5. PM_{10} and $PM_{2.5}$ evaluation on selected cities with and without assimilation. a, Selected international cities around the aircraft measurement track. City 1 – 4 are in the downwind direction, while city 5 – 8 are in the upwind direction. b, Concentrations of PM_{10} and $PM_{2.5}$ and the quantified impact rates on selected cities. The height of interest is chosen at 3 km. The red arrow represents the trend of concentration values due to the assimilation process. The impact rates in downwind and upwind cases are distinguished by blue and magenta colors.

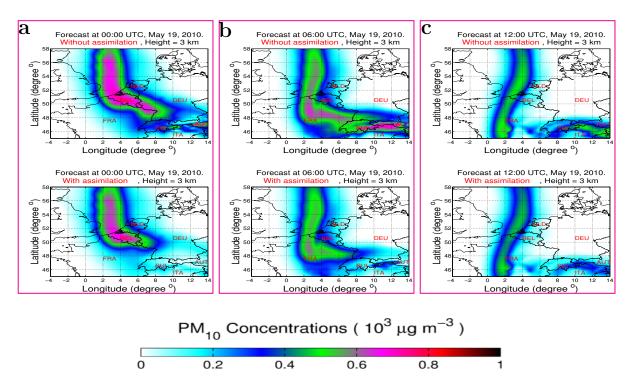


Figure 6. One-day forecast of PM₁₀ **concentration with and without assimilation. a**, Forecast at 00:00 UTC 19 May 2010. **b**, Forecast at 06:00 UTC 19 May 2010. **c**, Forecast at 12:00 UTC 19 May 2010.