

Dear Editor,

Herewith we submit the revised manuscript. First we would like to thank you and two anonymous reviewers, and really appreciate the detailed comments and suggestions. We have carefully considered all the concerns and made changes accordingly in the revised paper. We believe the new version has been improved a lot compared to the previous version.

In the following we will give our answers and reactions to all of the comments. To make the changes easier to identify, we have numbered them.

Best regards,

G. Fu

on behalf of all co-authors

(The revised and marked manuscript is [in the latter part of this pdf.](#))

Reply to Anonymous Referee #1

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 2.14–2.15](#):

“ 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 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 2.21–2.23](#):

“ 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 for getting translated 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 2.28–2.29](#):

“ 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 4.7](#):

“ 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 cus- tomary*

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 3.28–4.17](#):

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 μm	29	1125.00
vash_2	63 to 250 μm	31	156.50
vash_3	30 to 63 μm	12	46.50
vash_4	10 to 30 μm	18	20.00
vash_5	2.5 to 10 μm	8	6.25
vash_6	0.0 to 2.5 μm	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 definitions in Mastin et al. (2009)), in which the mass fraction of erupted debris finer than 63 μm 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

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

where $\text{V}(\text{m}^3 \text{s}^{-1})$ 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 PM_{10} and $\text{PM}_{2.5}$ (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 4.28–4.29), it is explicitly 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 6.27–7.8:

“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 $100\text{-}200 \text{ km h}^{-1}$ 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.

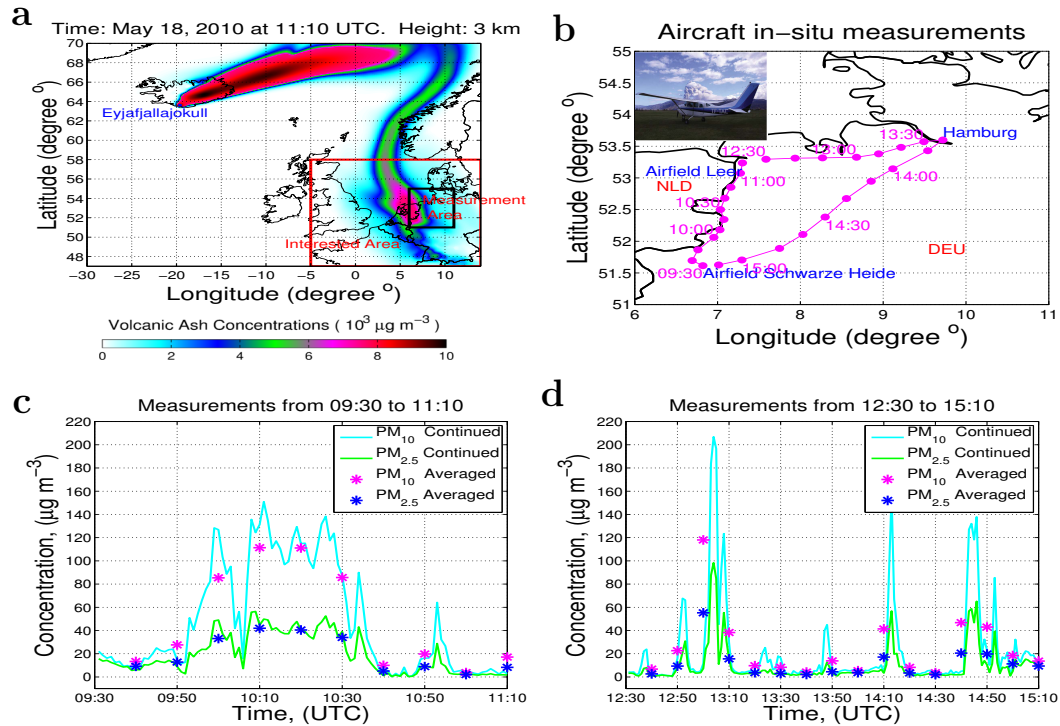


Figure 1: **Aircraft in-situ measurements of distal volcanic ash plume.** **a**, The LOTOS-EUROS simulation of volcanic ash plume at 11:10 (UTC), 18 May 2010. **b**, Measuring aircraft flight route on 18 May 2010. **c**, PM_{10} and $PM_{2.5}$ measurements from 09:30 to 11:10 (UTC). **d**, PM_{10} and $PM_{2.5}$ 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 7.34–8.2](#):

“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. With your comments and suggestions of Reviewer #2, we reconsider the validation locations after a careful selection of validation locations (see Fig. 2 in the marked pdf).

It is reformulated in [lines 9.11–9.15](#):

“Furthermore, we can also see that at each validation location, the forecast with assimilation is 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, therefore the assimilated volcanic ash state (Fig. 3d) is a more accurate approximation to the real state of distal volcanic ash plume.”

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 9.5–9.6](#):

“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 9.18–10.2](#):

“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, 5 big cities around the measurement route are selected (see Fig. 5a). They are Dortmund, Cologne, Luxembourg in the downwind direction and Amsterdam, Rotterdam in the upwind direction. The evaluation height is chosen at 3 km. For some continental or intercontinental passenger flights, 3 km might be of special interest regarding taking off and landing. 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:

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

where p means either PM_{10} or $\text{PM}_{2.5}$, i means index of selected cities. Moreover, $(\text{SimuNoAssimi})_p$ and $(\text{SimuAssimi})_p$ 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, Cologne and Luxembourg) are much more significant than those in the upwind direction (Amsterdam, Rotterdam). 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 7.34–8.1: “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 11.7–11.8: “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. After a careful reconsidering the validation locations by suggestions of Reviewer #2. We reformulated this in lines 11.7–11.9: “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.”

Corrections:

1. As suggested, “It has been shown. . .” is corrected to “It was shown” in line 2.29.

2. “In this study...” is corrected to “In that study” in [line 2.31](#).
3. “due to assimilation” is changed to “After the assimilation” in [line 8.7](#).
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 8.5–8.9](#):
“ Note that areas with ash concentration higher than $4000 \mu\text{g m}^{-3}$ 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 $3000 \mu\text{g m}^{-3}$) 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 8.24–8.25](#):
“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 9.13–9.15](#):
“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 9.18–9.20](#):

“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 9.23–9.25](#):

“The concentrations of two major distal volcanic ash components, i.e., PM_{10} and $\text{PM}_{2.5}$ (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 9.30–10.2](#).

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 10.3–10.4](#).
10. 8.20-23: *“This aviation. . .forbidden area.” Comment: This sentence has a lot of clauses and is confusing. Consider revising.*
11. “direction of distal” is corrected to “direction of the distal” in [lines 10.5– 10.6](#).

As suggested, the sentence is revised as in [lines 10.7–10.10](#):

“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 $\mu\text{g m}^{-3}$.”* Comment: *This sentence has a lot of clauses and is confusing. Consider revising.*

As suggested, the sentence is revised as in [lines 10.10–10.12](#):

“ 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 $\mu\text{g m}^{-3}$. ”

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 10.19–10.20](#):

“ 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 10.20](#).

15. Thanks for pointing out the difference between “detected” and “visible” or “noticeable”. It is corrected in [line 10.24](#).

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 10.24–10.25](#):

“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 10.31–11.5](#).

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 11.14–11.15](#)).

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 11.16–11.20](#):

“The computer experiment revealed that the time period of the improvement effect on the areas downwind to the assimilated observations 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 lines 11.22–11.23.

21. 9.30-31: “*In future work, . . . into account.*” *Comment: This sentence needs editing for grammar.*

It is corrected in lines 11.23–11.24:

“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. ”

Reply to Anonymous Referee #2

Reply to the specific comments:

1. 2.1: *It should be clarified why the influence of ESPs weakens with the distance to the volcano.*

Response: We added an explanation in [lines 1.23–2.1](#):

“ This is mainly because (1) compared to ESPs, the plume transport becomes more and more dominant as the distance to the volcano increases (Macedonio et al., 2016); (2) the small errors in ESPs can accumulate into large errors in predicted ash concentrations after a large transport distance (Webster et al., 2012). ”

2. 2.9: *The difficulty of satellite data assimilation should be discussed carefully. Satellite lidar instruments like CALIOP or CATS provide highly resolved aerosol profiles. The challenge of satellite data assimilation is that the observations are often no direct measurements of the quantity of interest, but optical property measurements. Therefore the aerosol quantity need to be derived by a retrieval process or a complex observation operator.*

Response: A discussion has now been added in [lines 2.11–2.15](#):

“ This is because satellite observations are often not direct measurements of the quantity of interest, but optical property measurements. Therefore the aerosol quantity needs to be derived by a retrieval process or a complex observation operator. Moreover, satellite data are often two-dimensional (2D), thus are lack of sufficient 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 spatially sparse (Winker et al., 2012). ”

3. 2.12: *Measurements “close to the eruption plume”, this formulation might be misleading, since the article focuses on distal ash clouds.*

Response: I agree. It is reformulated in [lines 2.17–2.18](#):

“ These aircraft-based measurements can be obtained in the boundaries of volcanic ash plume, which are probably the most direct volcanic ash observations possible. ”

4. 2.16: *Lidars do not provide vertical integrated column data.*

Response: It is corrected in [lines 2.21–2.23](#):

“ 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 for getting translated into concentration estimates. ”

5. 2.19: *It remains unclear how research flight operators know where to actually obtain the “most relevant” volcanic ash concentration.*

Response: It is added in [lines 2.24–2.27](#):

“ Third, an aircraft is flexible in flight route to follow the ash clouds. Although an aircraft measurement plan is usually beforehand designed for a region/altitude of interest, the aircraft operator can adjust the detailed route according to the real conditions (e.g., ash concentration level, wind direction) of the plume transport in order to always obtain appropriate volcanic ash concentrations (Weber et al., 2012). ”

6. 2.25: *It should be formulated what kind of estimates are highly influenced by “plume height and mass eruption rate”.*

Response: It is reformulated in [lines 2.31–2.33](#):

“ 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 of the forecasted ash concentrations. ”

7. 4.2: *“for this type of measurements in well calibrated cases.” To me it is unclear what “well calibrated cases” are and whether you have those during the performed flight.*

Response: Thanks for the question. The “for this type of measurements in well calibrated cases” means the aircraft data are directly calibrated with an independent reference method during the performed flight, as discussed in (Weber et al., 2010, 2012). We agree here “well calibrated cases” is a confusing term and has been reformulated in [lines 4.27–4.28](#):

“ which can be taken as the instrumental error for this type of measurements during the performed flight. ”

8. 4. 29 (and Chapter 2.3 in general): *To me it is not clear how the forecast error covariance matrix looks and especially how the localization is performed here.*

Response: We have added the following description in [lines 6.3–6.13](#):

“

$$\mathbf{K}(k) = (\mathbf{f} \circ \mathbf{P}^f(k))\mathbf{H}(k)^T [\mathbf{H}(k)(\mathbf{f} \circ \mathbf{P}^f(k))\mathbf{H}(k)^T + \mathbf{R}]^{-1}, \quad (3)$$

$$\xi_j^a(k) = \xi_j^f(k) + \mathbf{K}(k)[\mathbf{y}(k) - \mathbf{H}(k)\xi_j^f(k) + \mathbf{v}_j(k)], \quad (4)$$

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

In Eq. (3), a distance-based localization (Houtekamer and Mitchell, 1998, 2001). is obtained using a Schur product $\mathbf{f} \circ \mathbf{P}^f$ (i.e., element-wise multiplication) in order to reduce the spurious correlations caused by the finite ensemble size, which is a general problem in ensemble-based data assimilation. The correlation matrix \mathbf{f} is obtained by applying a correlation function to the Euclidean distance between two points. The correlations decrease to zero beyond a certain distance. Distance-based localization can be easily implemented for eliminating the spurious correlations outside the localized region. For some applications (e.g., ozone, CO₂, sulfur dioxide), this approach has achieved an acceptable performance

with a simple setup using a constant localization parameter of 50–100 km (Curier et al., 2012; Chatterjee et al., 2012; Barbu et al., 2009). In this study, 100 km is adopted as the localization parameter in volcanic ash assimilation of aircraft in-situ measurements. ”

9. 5.12: *You describe that there are no ash emissions prior to 9:00 UTC 14 April 2010, but it remains undiscussed how the volcanic ash is released during the first guess forecast (PH, MER, VMD)*

Response: We added the description in lines 6.18–6.19 which refers to lines 3.28–4.6 where a detailed description is added.

lines 6.18–6.19:

“ The volcanic ash is released during the first guess forecast based on the defined ESPs (PH, MER, VMD, PSD), as discussed in Section 2.1. ”

and lines 3.28–4.6:

“ The input parameter PH in LOTOS-EUROS is taken from hourly based Icelandic Meteorological Office (IMO) plume height detection (Gudmundsson et al., 2012) 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 as the ESP type S2 (see definitions in Mastin et al. (2009)), in which the mass fraction of erupted debris finer than 63 μm is 0.4. For the S2 type eruption, Durant and Rose (2009) provided the base for the PSD from their analysis of the 1992 Crater Peak, Mount Spurr event. Hence, Table 2 shows the ash distribution based on their analysis and is used in our experiments. 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

$$\text{PH} = 2.00V^{0.241}, \quad \text{and} \quad \frac{V}{\text{MER}} = \frac{1.5e^3}{4.0e^6}, \quad (5)$$

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

Table 2: 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 μm	29	1125.00
vash_2	63 to 250 μm	31	156.50
vash_3	30 to 63 μm	12	46.50
vash_4	10 to 30 μm	18	20.00
vash_5	2.5 to 10 μm	8	6.25
vash_6	0.0 to 2.5 μm	2	1.25

10. 5.17: *The “plume height detection data” has to be described more precisely.*

Response: A description is added in [lines 3.28–3.29](#):

“ The input parameter PH in LOTOS-EUROS is taken from the hourly based plume height detection by Icelandic Meteorological Office (IMO) (Gudmundsson et al., 2012) and usually the uncertainty of PH is taken as 20 % (Bonadonna and Costa, 2013). ”

11. 5.30: “... approximates the measurements with a high accuracy” → *If I did not see the corresponding figure, I would expect the analysis to be identically equal to the observations.*

Response: A note is added in [lines 7.17–7.18](#):

“ Note that, the high accuracy here doesn’t mean “identically equal”, but “very close”. ”

12. 6.21: “. . . continental Europe is all simulated in a low concentration level . . .” → *This sentence needs some reformulation, since the analysis does not apply to all continental Europe, e.g. in Northern Netherlands there still appear high ash concentration values.*

Response: It is now reformulated in [lines 8.7–8.9](#):

“ After the assimilation process, the calculated volcanic ash concentrations in Germany, Luxembourg and the Netherlands (except in Northern Netherlands) have a lower concentration level (lower than $3000 \mu\text{g m}^{-3}$) and the changes on volcanic ash state can be seen across a wide area. ”

13. 6.23: “Thus in a fairly large domain, the state change at measurement location also influences state variables in the surrounding areas.” → *To my understanding this is caused by the chosen influence radius, which is not discussed in this study. Further, the downwind direction includes influenced states due to the transport of earlier corrected ash concentrations, especially regarding forecasts later than the assimilated time steps.*

Response: As suggested, the discussion is added in [lines 8.12–8.15](#):

“ This is caused by the chosen localization radius (see Section 2.3) in the assimilation process. Further, the downwind direction includes influenced state variables due to the transport of earlier corrected ash concentrations, especially regarding forecasts later than the assimilated time steps. ”

14. 7.4: “Fig. 4 shows the difference of a number of . . .” → *There is no difference plot in Fig. 4. Please be more precise, e.g. “the comparison of Fig. 4a and 4b. . .”. And it remains unclear which “number” you refer here. Aren’t these just two forecasts?*

Response: You are right. It is corrected in [line 8.26](#):

“ Fig. 4a and Fig. 4b show the comparison of the forecasted volcanic ash plumes with and without assimilation. ”

15. 7.6: “initiated with Fig. 3c” → *Fig. 3c includes results of an assimilation process as well. Later, the forecasts using these initial values are always designated to be “without*

assimilation". Please resolve this issue more clearly.

Response: Thanks a lot for the careful check. This is a text error which should be "initiated with Fig. 3b". It is corrected in [line 8.28](#):

" [initiated with Fig. 3b and Fig. 3d.](#) "

16. *7.11: It is unclear where the area of "downwind direction" is, since the wind field is nowhere described or visualized.*

Response: A description has now been added in [lines 8.14–8.15](#):

" Note that after a careful check on the wind field around the aircraft route, the term "downwind" direction means the direction of "South-East", which will be used in the following discussions. "

17. *7.18 - 21: ". . . at times between 13:20 and 14:00 . . . are far away from the measurement track" Actually these observations are not in the area, which is influenced by the assimilation (see Fig. 3e/3f). Therefore, it is unclear how these observations can be utilized for validation. For validation independent data has to be chosen, but it must be selected carefully with respect to the influenced area. Additionally, these observations are in a much better agreement with the measurements than the observations taken before 13:20 UTC and after 14:00 UTC. This could lead to the interpretation that the first guess (without any assimilation) corresponds much better with the observations than the forecasts with assimilation. Please reconsider the choice of observations for validation.*

Response: We agree with you. The choice of observations is reconsidered to be from 14:10 UTC to 15:00 UTC, by a careful selection of validation locations.

The corresponding figure is re-plotted as the following figure.

The description is reformulated in [lines 9.11–9.15](#):

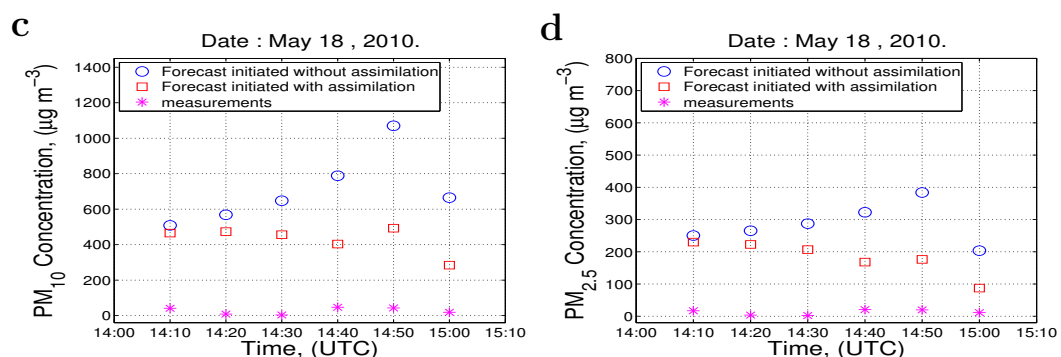
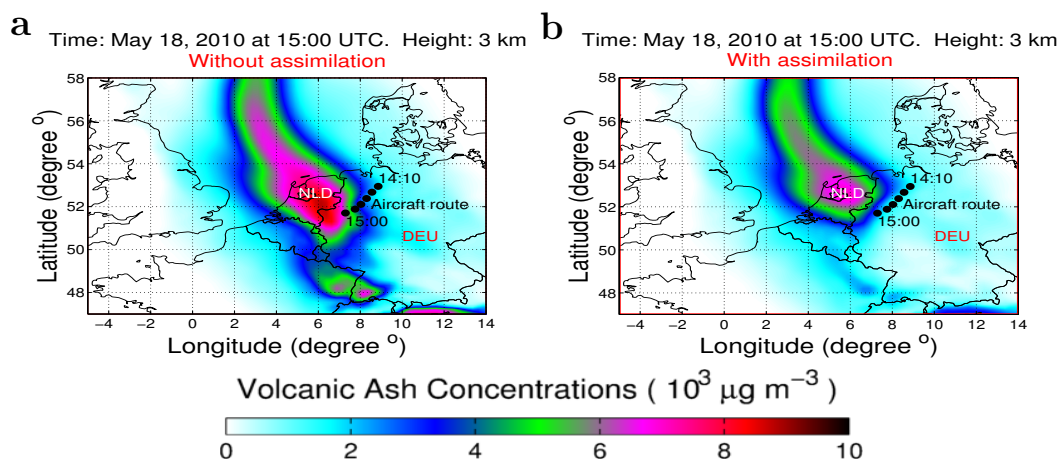
" Furthermore, we can also see that at each validation location, the forecast with assimilation is closer to the measurements than the forecast without assimilation, and also that the overestimation is significantly reduced using assimilation. This shows that the forecast at these locations with assimilation is more accurate than the forecast without assimilation, therefore the assimilated volcanic ash state (Fig. 3d) is a more accurate approximation to the real state of distal volcanic ash plume. "

18. *7.23: Declaring the forecast results with assimilation to be "much closer to the measurements than the forecast without assimilation" for the time steps between 12:30 UTC and 13:20 UTC might be too optimistic.*

Response: We agree. It is reformulated in [line 9.12](#):

" [at each validation location, the forecast with assimilation is closer to the measurements than the forecast without assimilation](#) "

19. *Comment on Chap. 4 and 5: It remains unclear to me how the length of integration time after the assimilation can influence the forecast results. It should be discussed how the area*



influenced by the assimilation temporally propagates.

Response: According to the suggestion, it is now discussed in [lines 8.28–8.30](#):

“ After the assimilation process, the assimilation influenced region temporally propagates to the downwind direction due to the meteorological drive (wind speed and direction). Thus the forecasted downwind ash concentrations are influenced along the length of integration time after assimilation. ”

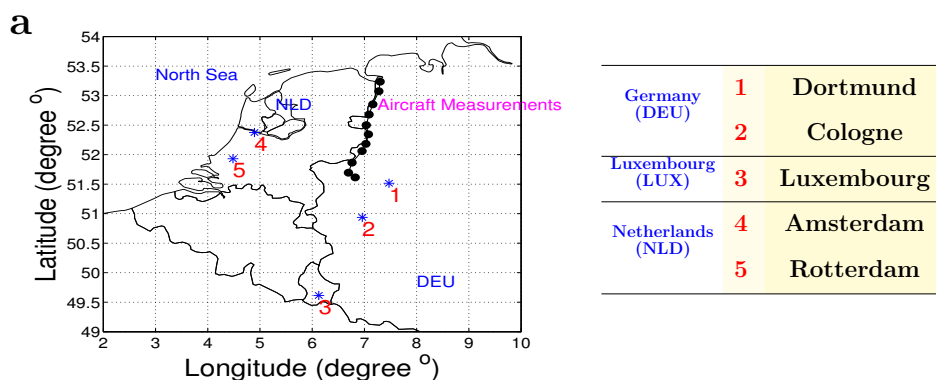
20. 7.32: *It has to be clarified if Antwerp and Brussels are even in the area, which is influenced by the assimilation.*

Response: After a careful check, Antwerp and Brussels are on the boundaries of the influenced area. For avoiding misleading, we removed two cities since the influence on them is not obvious. The new description is in [lines 9.20–9.21](#):

“ For this investigation, 5 big cities around the measurement route are selected (see Fig. 5a). They are Dortmund, Cologne, Luxembourg in the downwind direction and Amsterdam, Rotterdam in the upwind direction. ”

The corresponding figure is changed to:

21. 8.1: *In my opinion there are no continental passenger flights that operate in an altitude*



b

City Number		downwind			upwind	
		1	2	3	4	5
Simulation	Without assimilation ($\mu\text{g m}^{-3}$)	574.0	817.5	568.8	1055.3	863.4
	With assimilation ($\mu\text{g m}^{-3}$)	112.1 ↓	219.1 ↓	262.5 ↓	717.8 ↓	724.2 ↓
	Impact rate	80.5%	73.2%	53.9%	32.0%	16.1%
PM _{2.5}	Without assimilation ($\mu\text{g m}^{-3}$)	243.7	344.6	216.1	387.9	306.3
	With assimilation ($\mu\text{g m}^{-3}$)	49.3 ↓	90.5 ↓	97.3 ↓	259.3 ↓	256.6 ↓
	Impact rate	79.8%	73.7%	55.0%	33.2%	16.2%

of 3 km. 3 km might be of special interest regarding take off and landing.

Response: It is corrected in [lines 9.21–9.23](#):

“ The evaluation height is chosen at 3 km. For some continental or intercontinental passenger flights, 3 km might be of special interest regarding taking off and landing. ”

22. *Comment on the aviation advice: Clarify that you give the aviation advice only on the strength of the results in 3 km height. Generally all model levels must be analyzed for real cases. And it should be pointed out for which exact area and which time frame the aviation advice is given.*

Response: It is now reformulated in [lines 10.13–10.15](#):

“ Note that we give the aviation advice only on the strength of the results at 3 km height. Generally all model levels must be analyzed for real cases. And the real aviation advice also includes for which exact area and which time frame the advice is given. ”

23. *8.21: “most of the flights in East direction” I think it is not a matter of the flight direction; it affects all flights in the ash penetrated area.*

Response: It is corrected in [lines 10.9–10.10](#):

“ imposing such a no-fly zone will affect all flights in the ash penetrated area and subsequently leads to a huge economic loss. ”

24. 8.24: *“the whole of Europe” This has to be reconsidered. Not all Europe is analyzed.*

Response: It is reformulated in [lines 10.11–10.12](#):

“ The sky in large parts of Europe is open for commercial flights, because except in small parts of the Netherlands ash concentrations all over the domain of interest are lower than $3000 \mu\text{g m}^{-3}$. ”

25. 8.32: *“. . . the assimilation impact will last at least one day.” Such conclusions have to be discussed carefully. This conclusion is only valid for the areas, that are influenced by the assimilation, which changes with time, and as long as there are no high concentrated plumes transported into the area of interest. You might have to reconsider this statement especially for regions upwind to the assimilated observations.*

Response: We agree. The statement has been reconsidered in [lines 10.20–10.25](#):

“ Since there are clear differences between the two cases, the assimilation impact can last one day. Note that this impact duration is only valid for the areas (especially for regions downwind to the assimilated observations) that are influenced by the assimilation, which changes with time. When forecasting 24 hours (Fig. 6c), differences still can be observed, but the impact of assimilation is obviously getting much smaller (compared to Fig. 6a and Fig. 6b). Small differences are visible at cloud boundaries. Actually we also examined the assimilation impact in the forecast of the next day and observed only very small differences. ”

26. 8.34: *“Only small differences are detected in the Northern part of Italy.” In general I recognize only small differences at all cloud edges.*

Response: It is corrected in [line 10.24](#):

“ Small differences are visible at cloud boundaries. ”

27. 9.10: *“using only one or two measurements can not produce accurate results.” How do you know that at the number of measurements you used for your analysis is sufficient? Using aircraft measurements the number of observations only influences the area which can be analyzed. This does not mean, that the assimilation of one or two measurements is not valuable for a certain region.*

Response: We agree that it is incorrect to state “using only one or two measurements can not produce accurate results.”. The sentence is changed in [line 11.1](#):

“ It was shown that all the assimilation steps contribute to the final result. ”

28. 9.25: *“aviation advice can significantly benefit from the ensemble-based data assimilation process” Here it should be pointed out again, that this is only true within the influence radius/the assimilation influenced area.*

Response: As suggested, it has been added in [line 11.15–11.17](#):

“ Investigation shows that the accuracy of aviation advice within the assimilation influenced area can significantly benefit from the ensemble-based data assimilation process. The

computer experiment revealed that the time period of the improvement effect on the areas downwind to the assimilated observations can be taken as 24 hours. ”

29. *9.27: “we suggest the frequency of the measurement campaign to be once per day.” This cannot be said without loss of generality. Be attentive with the area that is influenced by the assimilation and the temporal change due to wind induced transport.*

Response: It is reformulated in [lines 11.17–11.20](#):

“ 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. The suggested frequency should be adjusted by the temporal strength (due to wind induced transport) on the assimilation influenced area. ”

Reply to Comments on Figures:

1. *Fig. 1b and 1c: could be left out, since there is no special meaning to the study or their meaning should be pointed out in the text.*

Response: It has been changed as suggested, see the revised manuscript (latter part of this pdf).

2. *Fig. 2 and 3: Which level/height is shown? Why is this level chosen?*

Response: The height information of “3 km” is added in Fig. 2 and 3 (see the revised manuscript). The evaluation height is chosen to be 3 km because the measurements are taken at altitudes around this height, which is described in lines 4.31–4.32 (“ [The measurements took place at heights around 3 km \(Weber et al., 2012\)](#) ”).

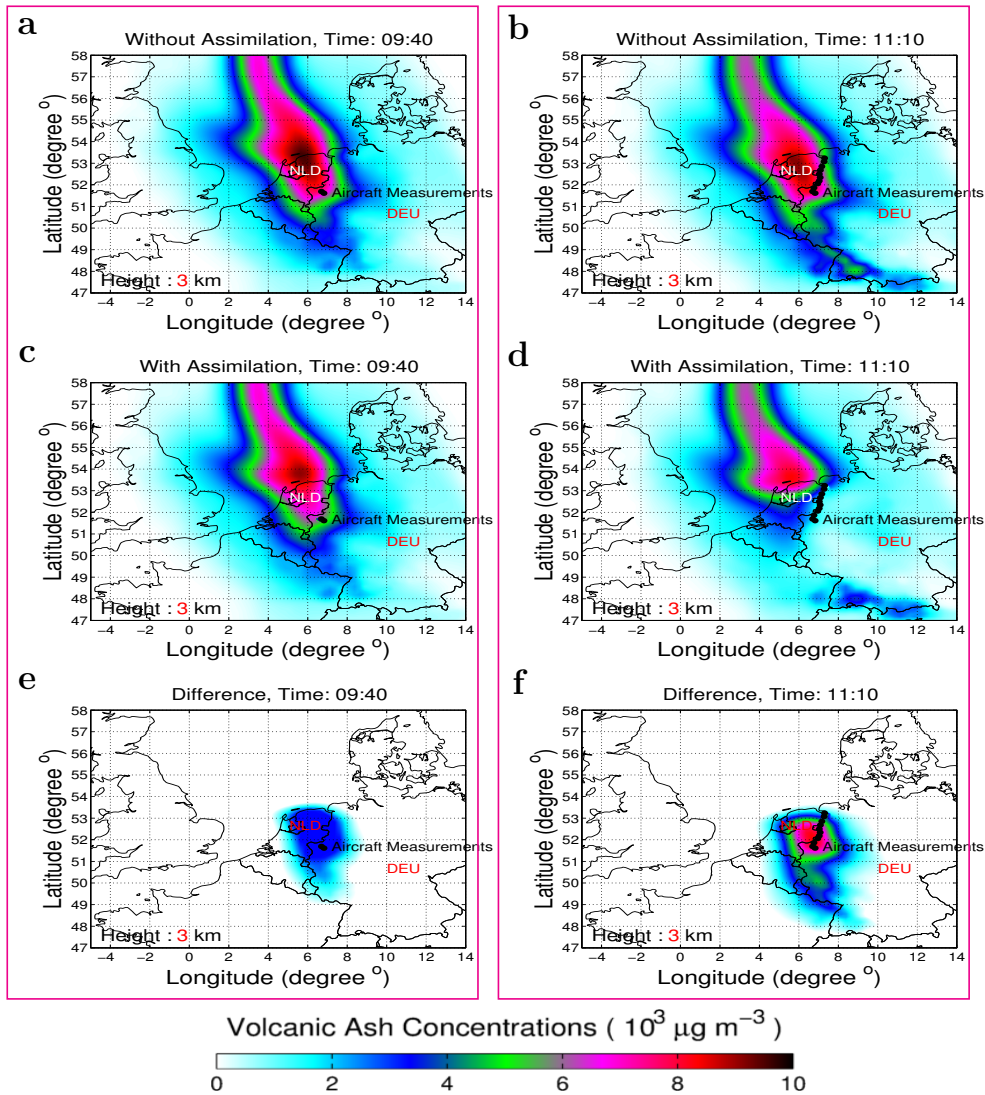
3. *Fig. 3e/3f: For me it is unclear how the differences of a-c and b-d can be a constant absolute value of 5. I would expect less difference between forecast and analysis at the edges of the influenced region. In addition, it is of special meaning if the assimilation induces reductions or increases of volcanic ash. Therefore, I suggest to define an extra color table for Figure 3e and 3f, which resolves the variation of the differences.*

Response: As suggested, we re-plotted Figure 3e and 3f.

4. *Fig. 4a/4b: The chosen colors of the aviation track might be misleading because they are included in the color table.*

Response: The color for aviation track is changed to black.

5. *Fig. 6: This figure shows a different domain compared to Fig. 1a,2,3,4. Be aware that not the whole domain was influenced by the assimilation.*



Response: Thank you for the careful check. It is explained in the caption of Fig. 6:
 “ A larger domain is chosen in this figure (compared to Fig. 2,3,4) to demonstrate the downwind change (with time) of the assimilation influenced area. ”

Reply to Technical corrections:

1. As suggested, we write UTC after every time statement.
2. “It has been shown. . .” is corrected to “It was shown. . .” in line 2.29.
3. “near to the eruption” is changed to “close to the eruption location” in line 2.31; “In this study” is changed to “in that study” in line 2.31.
4. “an Iceland eruption” is changed to “a volcanic eruption in Iceland” in lines 3.2–3.3.

5. “volcanic ash simulations” is changed to “volcanic ash dispersion simulations” in line 3.24.
6. “All of the measurement flights were. . .” is changed to “ The measurement aircraft was...” in line 4.23.
7. “N state” is changed to “N states” in line 5.11; “N volcanic ash state” is changed to “N volcanic ash states” in line 5.16.
8. As suggested, the “transpose representation” is added in line 5.25: “ [where the superscript “T” represents the transpose of the matrix.](#) ”
9. The repeat of PH definition in Section 3.1 is deleted, which is only in Section 2.1.
10. “of some of the ensembles” is changed to “of selected ensemble members” in line 7.14.
11. “but the overestimation vanishes by the assimilation process” is changed to “ but the overestimation diminishes by the assimilation process significantly” in line 7.16.
12. “a better forecast will be. . .” is changed to “ a better forecast is expected . . . ” in line 8.31.
13. “can not” is changed to “ cannot” in line 11.26.
14. “in satellite data” is changed to “ in certain satellite data” in line 11.27.

The revised manuscript starts from next page.

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

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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 Luxembourg 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 Eyjafjallajökull 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 (1) compared to ESPs, the plume transport becomes more and more dominant as the distance to the volcano increases (Macedonio et al., 2016); (2) the small errors in ESPs can

accumulate into large errors in predicted ash concentrations after a large transport distance (Webster et al., 2012). 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 based on their large detection domain and long-time continuous output data, they are not directly suited for data assimilation systems. This is because satellite observations are often not direct measurements of the quantity of interest, but optical property measurements. Therefore the aerosol quantity needs to be derived by a retrieval process or a complex observation operator. Moreover, satellite data are often two-dimensional (2D), thus are lack of sufficient 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 spatially sparse (Winker et al., 2012). Fortunately, the in-situ volcanic ash state variables can be directly and accurately measured nowadays by means of airborne observations of volcanic ash (Weber et al., 2012). These aircraft-based measurements can be obtained in the boundaries of volcanic ash 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 for getting translated 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. Although an aircraft measurement plan is usually beforehand designed for a region/altitude of interest, the aircraft operator can adjust the detailed route according to the real conditions (e.g., ash concentration level, wind direction) of the plume transport in order to always obtain appropriate volcanic ash concentrations (Weber et al., 2012).

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 close to the eruption location. 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 of the forecasted ash concentrations. 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 [a volcanic eruption in Iceland](#). 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 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 dispersion 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 the hourly based plume height detection by Icelandic Meteorological Office (IMO) (Gudmundsson et al., 2012) 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 as the ESP type S2 (see definitions in Mastin et al. (2009)), in which the mass fraction of erupted debris finer than $63 \mu m$ is 0.4. For the S2 type eruption, Durant and Rose (2009) provided the base for the PSD from their

analysis of the 1992 Crater Peak, Mount Spurr event. Hence, Table 1 shows the ash distribution based on their analysis and is used in our experiments. 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

$$5 \quad \text{PH} = 2.00V^{0.241}, \quad \text{and} \quad \frac{V}{\text{MER}} = \frac{1.5e^3}{4.0e^6}. \quad (1)$$

where $V(\text{m}^3 \text{s}^{-1})$ 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 PM_{10} and $\text{PM}_{2.5}$ (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.

20 **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). The measurement aircraft (Fig. 1b) was 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 during the performed flight. The total measurement error not only contains the instrumental error, but also includes an estimate of the model representation error (Fu et al., 2015).

30 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 at heights around 3 km (Weber et al., 2012) and 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 $\text{PM}_{2.5}$ were measured, see Fig. 1c and 1d.

2.3 The Ensemble Kalman Filter

- 5 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.
- 10 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 states, $\xi_1, \xi_2, \dots, \xi_N$. 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
- 15 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 states $\xi^a(0)$ 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 :

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

- 20 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:

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

$$\mathbf{L}^f(k) = [\xi_1^f(k) - \mathbf{x}^f(k), \dots, \xi_q^f(k) - \mathbf{x}^f(k)], \quad (4)$$

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

- 25 where the superscript “ T ” represents the transpose of the matrix. The observational network is defined by the observation operator H that maps state vector \mathbf{x} to observation space \mathbf{y} :

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

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:

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

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

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

In Eq. (7), a distance-based localization (Houtekamer and Mitchell, 1998, 2001) is obtained using a Schur product $\mathbf{f} \circ \mathbf{P}^f$ (i.e., element-wise multiplication) in order to reduce the spurious correlations caused by the finite ensemble size, which is a general problem in ensemble-based data assimilation. The correlation matrix \mathbf{f} is obtained by applying a correlation function to the Euclidean distance between two points. The correlations decrease to zero beyond a certain distance. Distance-based localization can be easily implemented for eliminating the spurious correlations outside the localized region. For some applications (e.g., ozone, CO₂, sulfur dioxide), this approach has achieved an acceptable performance with a simple setup using a constant localization parameter of 50–100 km (Curier et al., 2012; Chatterjee et al., 2012; Barbu et al., 2009). In this study, 100 km is adopted as the localization parameter in volcanic ash assimilation of aircraft in-situ measurements.

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

15 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”. The volcanic ash is released during the first guess forecast based on the defined ESPs (PH, MER, VMD, PSD), as discussed in Section 2.1. 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 stochastic PH 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 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.

Since the real-life measurements of the PM₁₀ 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 selected ensemble members. From the estimation of both volcanic ash components PM₁₀ 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 diminishes by the assimilation process significantly. Instead, the analysis mean consistently approximates the measurements with a high accuracy. Note that, the high accuracy here doesn't mean "identically equal", but "very close". 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 UTC (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 UTC and 11:10 UTC 18 May with and without assimilating aircraft measurements. Compared to the case without assimilation at 11:10 UTC (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 $4000 \mu\text{g m}^{-3}$ 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 Germany, Luxembourg and the Netherlands (except in Northern Netherlands) have a lower concentration level (lower than $3000 \mu\text{g m}^{-3}$) and the changes on volcanic ash state can be seen across 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. This is caused by the chosen localization radius (see Section 2.3) in the assimilation process. Further, the downwind direction includes influenced state variables due to the transport of earlier corrected ash concentrations, especially regarding forecasts later than the assimilated time steps. Note that after a careful check on the wind field around the aircraft route, the term “downwind” direction means the direction of “South-East”, which will be used in the following discussions. Another note is 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 UTC to 11:10 UTC. This can be seen from the assimilation results at 09:40 UTC and 11:10 UTC (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 UTC is less pronounced than at 11:10 UTC. 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. 4a and Fig. 4b show the comparison of the 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. 3b and Fig. 3d. After the assimilation process, the assimilation influenced region temporally propagates to the downwind direction due to the meteorological drive (wind speed and direction). Thus the forecasted downwind ash concentrations are influenced along the length of integration time after assimilation. Because all the other settings in the system are the same, a better forecast is expected due to a more accurate initial state. We use the measurements from 09:30 UTC to 11:10 UTC 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 14:10 UTC to 15:00 UTC 18 May 2010 (see Fig. 1b and 1d). The validation data is selected carefully with respect to the influenced area (Fig. 3f).

With different initialization, the forecast of volcanic ash concentration at 15:00 UTC shows large differences. The forecast after assimilation (see Fig. 4b, lower than $3000 \mu\text{g m}^{-3}$ in the downwind direction of the measurement track) is much smaller than that without assimilation (Fig. 4a, higher than $4000 \mu\text{g m}^{-3}$ 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 each validation location, the forecast with assimilation is 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, therefore 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.

5 Assimilation benefit for aviation community

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, 5 big cities around the measurement route are selected (see Fig. 5a). They are Dortmund, Cologne, Luxembourg in the downwind direction and Amsterdam, Rotterdam in the upwind direction. The evaluation height is chosen at 3 km. For some continental or intercontinental passenger flights, 3 km might be of special interest regarding taking off and landing. 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 $\text{PM}_{2.5}$ (Webley et al., 2012; Fu et al., 2015), are evaluated. Fig. 5b shows that results with assimilation is lower for both PM_{10} and $\text{PM}_{2.5}$ 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:

$$(\text{IR})_p(i) = \frac{(\text{SimuNoAssimi})_p(i) - (\text{SimuAssimi})_p(i)}{(\text{SimuNoAssimi})_p(i)}, \quad (9)$$

where p means either PM_{10} or $\text{PM}_{2.5}$, i means index of selected cities. Moreover, $(\text{SimuNoAssimi})_p$ and $(\text{SimuAssimi})_p$ 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, Cologne and Luxembourg) are much more significant than those in the upwind direction (Amsterdam, Rotterdam). 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 $4000 \mu\text{g m}^{-3}$. 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 affect all flights in the ash penetrated area and subsequently leads 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 large parts of Europe is open for commercial flights, because except in small parts of the Netherlands ash concentrations all over the domain of interest are lower than $3000 \mu\text{g m}^{-3}$. This illustrates that the accuracy of aviation advice and the NFZ area can significantly benefit from the ensemble-based data assimilation process. Note that we give the aviation advice only on the strength of the results at 3 km height. Generally all model levels must be analyzed for real cases. And the real aviation advice also includes for which exact area and which time frame the advice is given.

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 can last one day. Note that this impact duration is only valid for the areas (especially for regions downwind to the assimilated observations), that are influenced by the assimilation, which changes with time. When forecasting 24 hours (Fig. 6c), differences still can be observed, but the impact of assimilation is obviously getting much smaller (compared to Fig. 6a and Fig. 6b). Small differences are visible at cloud boundaries. 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.

30 6 Conclusions

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 contribute to the final result. 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. 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. Investigation shows that the accuracy of aviation advice within the assimilation influenced area can significantly benefit from the ensemble-based data assimilation process. The computer experiment revealed that the time period of the improvement effect on the areas downwind to the assimilated observations 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. The suggested frequency should be adjusted by the temporal strength (due to wind induced transport) on the assimilation influenced area.

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 certain 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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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 μm	29	1125.00
vash_2	63 to 250 μm	31	156.50
vash_3	30 to 63 μm	12	46.50
vash_4	10 to 30 μm	18	20.00
vash_5	2.5 to 10 μm	8	6.25
vash_6	0.0 to 2.5 μm	2	1.25

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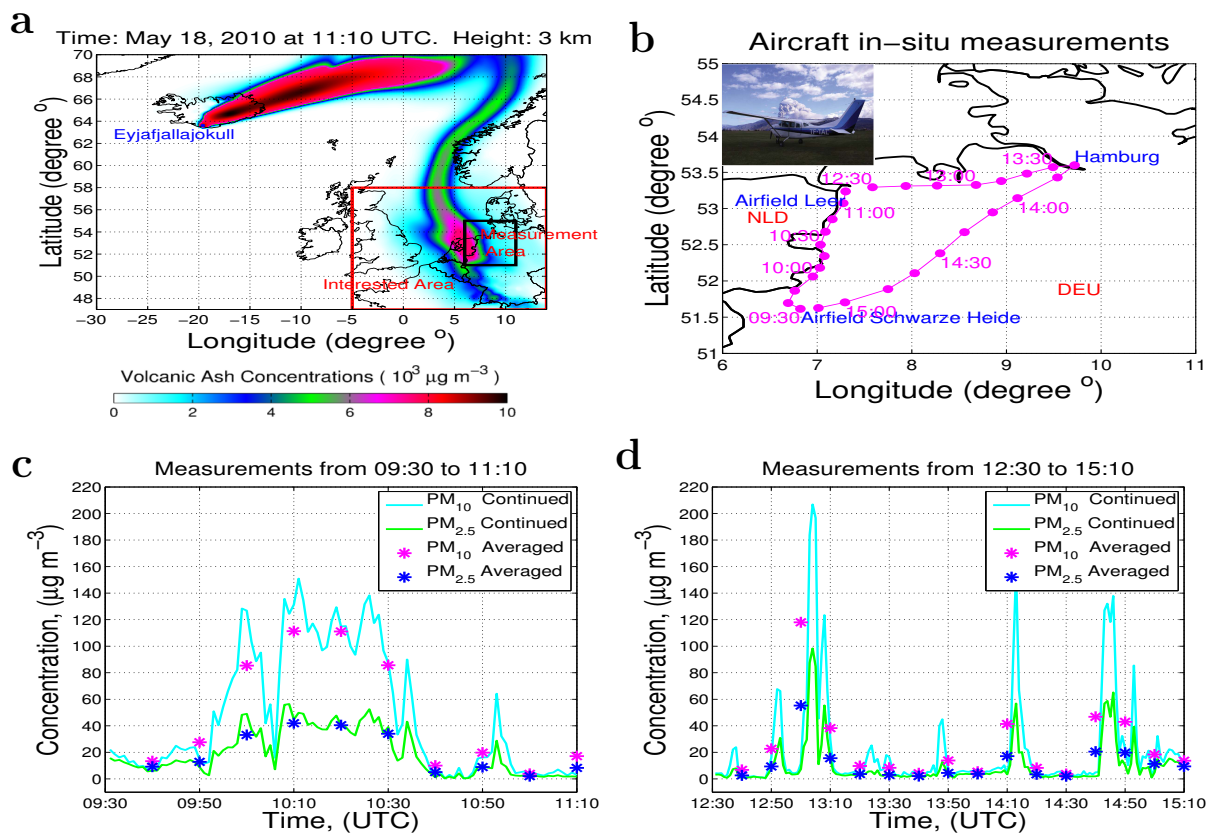


Figure 1. Aircraft in-situ measurements of distal volcanic ash plume. **a**, The LOTOS-EUROS simulation of volcanic ash plume at 11:10 UTC, 18 May 2010. **b**, Measuring aircraft flight route on 18 May 2010. **c**, PM₁₀ and PM_{2.5} measurements from 09:30 UTC to 11:10 UTC. **d**, PM₁₀ and PM_{2.5} measurements from 12:30 UTC to 15:10 UTC. In **c** and **d**, the curves show the values of PM₁₀ and PM_{2.5} measured at a frequency of every 6 seconds. The values marked with a star are the averaged PM₁₀ 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).

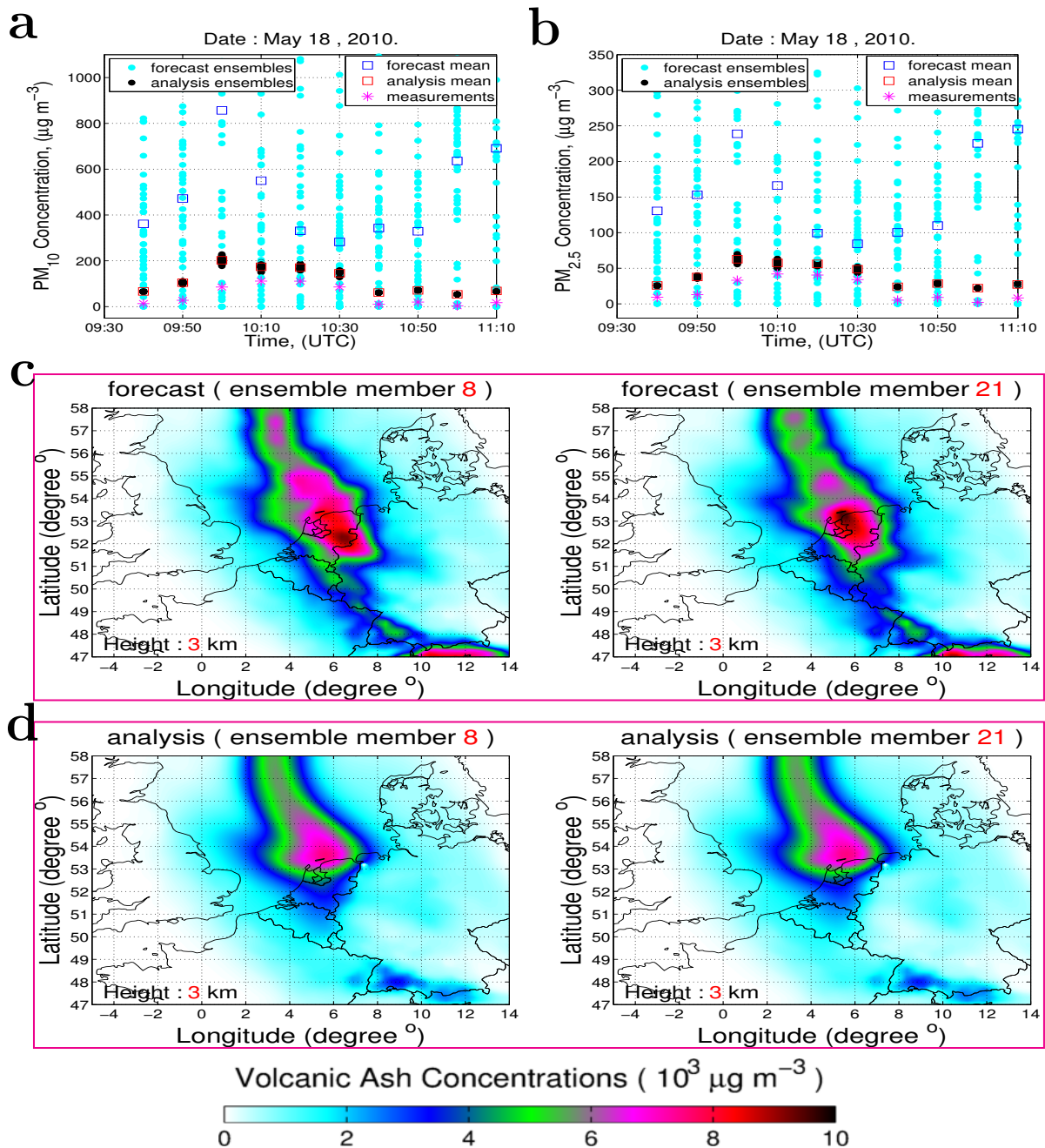


Figure 2. Assimilation effect from 09:40 UTC to 11:10 UTC, 18 May, 2010. **a** and **b**, Volcanic ash concentrations of PM₁₀ and PM_{2.5} at measurement locations. **c** and **d**, Volcanic ash ensemble state (ensemble member 8 and 21) of forecast and analysis at 11:10 UTC. The measurements, ensembles 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. The evaluation height is chosen at 3 km since the measurements are taken at altitudes around this height, see Section 2.2.

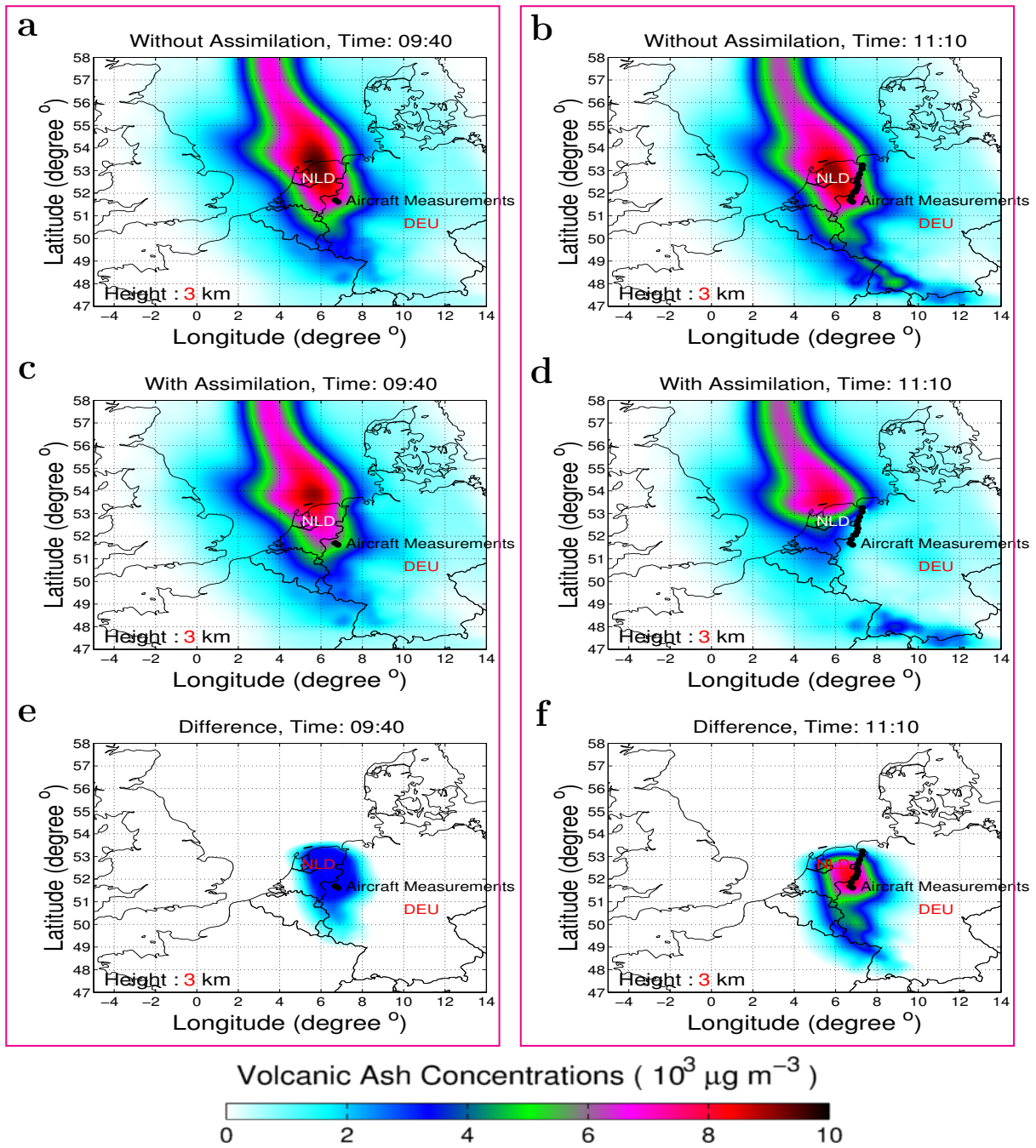


Figure 3. Comparison with and without assimilating aircraft in-situ measurements on 18 May 2010. **a** and **b**, Simulation results without assimilation at 09:40 UTC and 11:10 UTC. **c** and **d**, Simulation results with assimilation at 09:40 UTC and 11:10 UTC. **e**, Differences of **a** and **c**. **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.

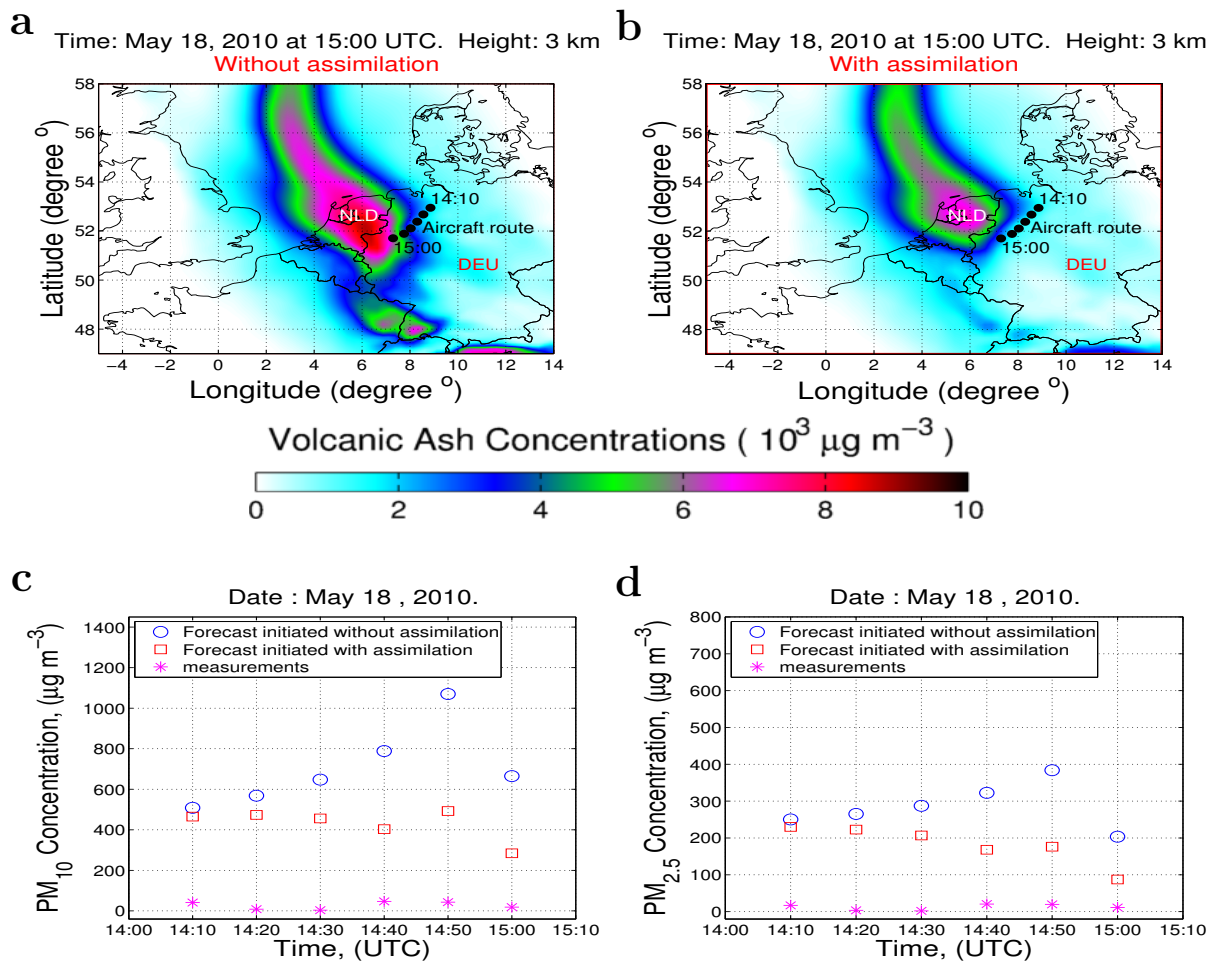


Figure 4. Forecast at 15:00 UTC 18 May 2010 with different initial conditions for the volcanic ash state. a, Forecast initiated with (Fig. 3b). b, Forecast initiated with (Fig. 3d). c, PM₁₀ concentration from 14:10 UTC to 15:00 UTC. d, PM_{2.5} concentration from 14:10 UTC to 15:00 UTC.

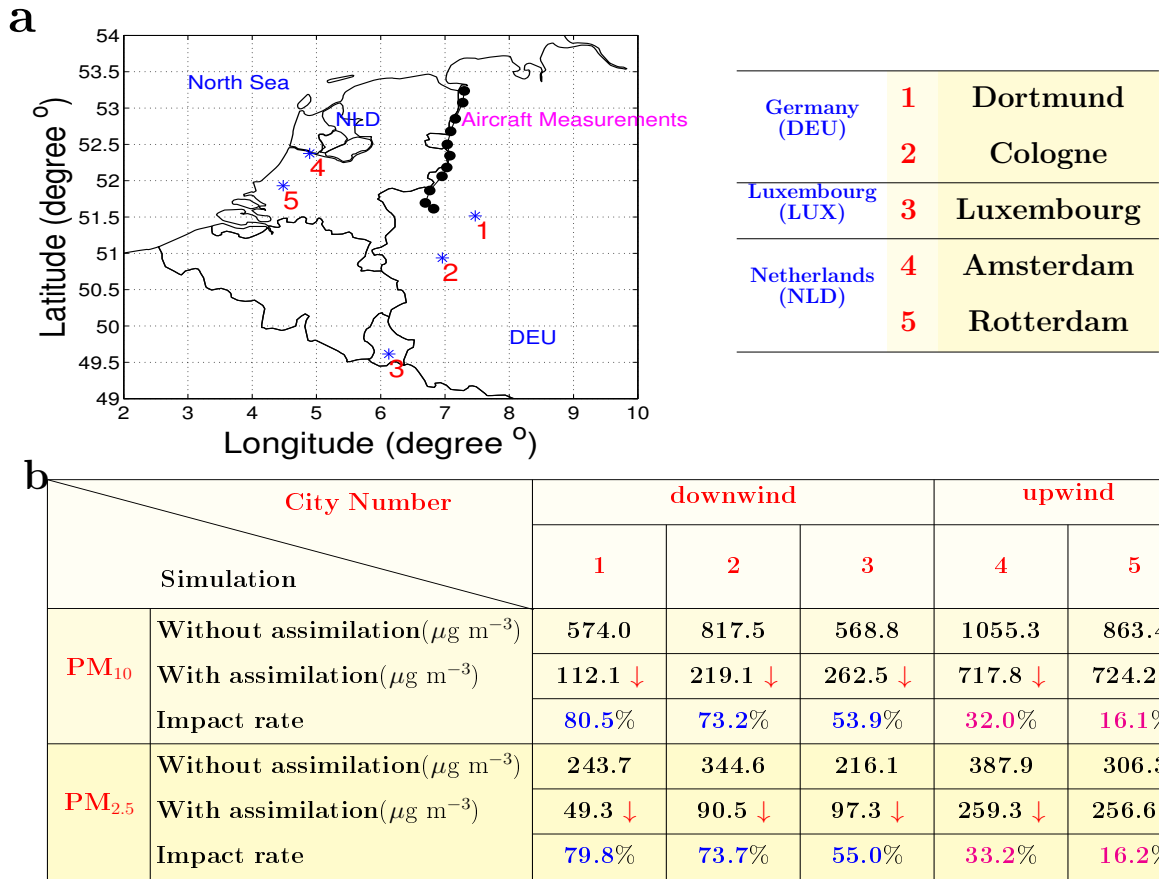


Figure 5. PM₁₀ and PM_{2.5} evaluation on selected cities with and without assimilation. **a**, Selected international cities around the aircraft measurement track. City 1 – 3 are in the downwind direction, while city 4 – 5 are in the upwind direction. **b**, Concentrations of PM₁₀ and PM_{2.5} and the 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.

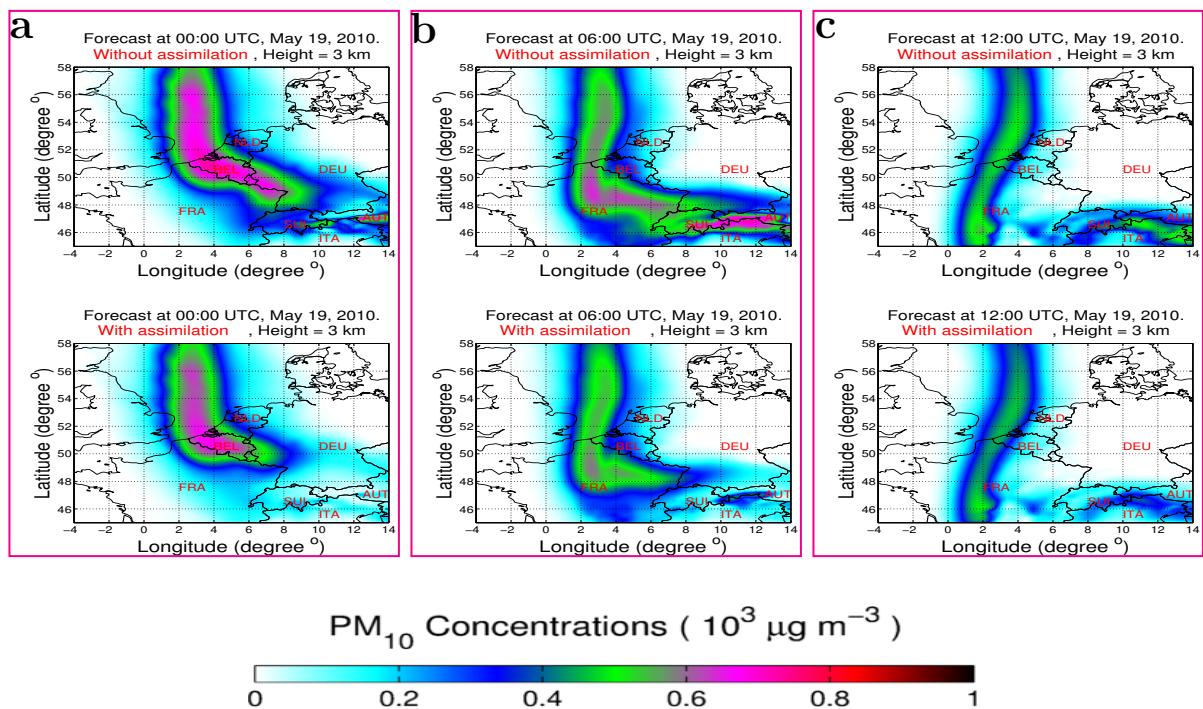


Figure 6. One-day forecast of PM_{10} concentration with and without assimilation. A larger domain is chosen in this figure (compared to Fig. 2, 3, 4) to demonstrate the downwind change (with time) of the assimilation influenced area. **a**, Forecast at 00:00 UTC 19 May 2010. **b**, Forecast at 06:00 UTC 19 May 2010. **c**, Forecast at 12:00 UTC 19 May 2010.