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Aircraft observations and model simulations of concentration and particle size distribution in the Eyjafjallajökull volcanic ash cloud

H. F. Dacre¹, A. L. M. Grant¹, and B. T. Johnson²

¹Department of Meteorology, University of Reading, UK

²Met Office, Exeter, UK

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Correspondence to: H. F. Dacre (h.f.dacre@reading.ac.uk)

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ACPD

12, 22587–22627, 2012

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

The Eyjafjallajökull volcano in Iceland emitted a cloud of ash into the atmosphere during April and May 2010. Over the UK the ash cloud was observed by the FAAM BAe-146 Atmospheric Research Aircraft which was equipped with in-situ probes measuring the concentration of volcanic ash carried by particles of varying sizes. The UK Met Office Numerical Atmospheric-dispersion Modelling Environment (NAME) has been used to simulate the evolution of the ash cloud emitted by the Eyjafjallajökull volcano during the period 4–18 May 2010. In the NAME simulations the processes controlling the evolution of the concentration and particle size distribution include sedimentation and deposition of particles, horizontal dispersion and vertical wind shear. For travel times between 24 and 72 h a $1/t$ relationship describes the evolution of the concentration at the centre of the ash cloud and the particle size distribution remains fairly constant. Although NAME does not represent the effects of microphysical processes it can capture the observed decrease in concentration with travel time in this period. This suggests that, for this eruption, microphysical processes play a small role in determining the evolution of the distal ash cloud. Quantitative comparison with observations shows that NAME can simulate the observed column integrated mass if around 4 % of the total emitted mass is assumed to be transported as far as the UK by small ($< 30 \mu\text{m}$ diameter) particles. NAME can also simulate the observed particle size distribution if a distal particle size distribution that contains a large fraction of $< 10 \mu\text{m}$ diameter particles is used, consistent with the idea that phraetomagmatic volcanoes, such as Eyjafjallajökull, emit very fine particles.

1 Introduction

The 2010 Eyjafjallajökull volcanic eruption affected air traffic over Europe for 13 days, leaving 10 million passengers stranded and leading to an estimated revenue loss of £1.5 billion to the airline industry due to the closure of European air space (European

ACPD

12, 22587–22627, 2012

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Commission, 2010). Thus accurate predictions of the evolution of volcanic ash clouds are important for the airline and tourist industries as well as policy makers and the general public. The aims of this paper are to evaluate model simulations of volcanic ash using in-situ measurements, to develop a new method for estimating the distal particle size distribution, and to explain the general dispersion characteristics of volcanic ash.

The characterisation of volcanic eruption source parameters such as the mass eruption rate and particle size distribution are crucial for accurately predicting volcanic ash dispersion. In this paper *Particles* are defined as the entities dispersing in the atmosphere, which may be either unaggregated single grains of ash or aggregates, i.e. clusters of grains of ash. As most volcanic ash dispersion models do not represent the complex dynamics which occur close to the volcano it is necessary to assume an effective 'distal' source describing the mass and characteristics of material remaining in the distal ash cloud after the near-source microphysical processes have occurred. In this paper *distal* refers to ash > 500 km or > 6 h travel time from the volcano. At present, methods exist for estimating the near-source mass eruption rate and particle size distribution. For example, as there is no direct method of measuring the mass eruption rate of eruption volcanoes, most volcanic ash dispersion models use empirical relationships to relate observed maximum plume height to the near-source mass eruption rate (Wilson et al., 1978; Sparks et al., 1997; Mastin et al., 2009). However, the fraction of the near-source mass that remains in the distal ash cloud (i.e. carried by small ash particles that have not fallen out close to the source) is not well known. Similarly, methods to estimate particle size distribution are frequently based on analysis of ash deposits close to the volcano vent as distal ash is often too widely dispersed to be sampled (Volentik et al., 2010; Rust and Cashman, 2011). Thus these calculated particle-size distributions, based on fall deposits, are representative of the near-source particle size distribution only. In addition, they are not available in real time and can depend strongly on the method used to analyse the deposits (Bonadonna and Houghton, 2005). In this paper we compare NAME simulations with in-situ observations of ash concentrations

Volcanic ash clouds

H. F. Dacre et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

and particle size distributions in the distal ash cloud with a view to determining the the distal fine ash fraction (DFAF) and distal particle size distribution (DPSD).

The characterisation of distal eruption source parameters is crucial for volcanic ash dispersion models as studies have shown that for individual case studies, variations in the assumed PSD can lead to dramatic variations in tephra dispersion results (Daniele et al., 2009; Scollo et al., 2008). Similarly varying the DFAF simply scales the modelled ash concentrations. It is possible, that these distal eruption source parameters may vary in time. For example, Kratzmann et al. (2010) suggested that the highly stratified nature of fall deposits they observed (fine ash layered with pumice lapilli) was likely to be the result of multiple events leading to variations in the dominant particle size being deposited. Other studies suggest that differential sedimentation and subsequent deposition of particles can lead to variations in the near-source observations. For example, Scasso et al. (1994) found that the mean and median particle sizes deposited at the surface from the Hudson volcano, decreased rapidly up to 270 km from the volcano vent, beyond that point they were more or less constant up to 550 km. Similarly, in a study combining ash fallout data observed up to 100 km from the volcano, for a range of volcanic eruptions, Horwell (2007) found that there was a strong linear relationship between 4 μm and 10 μm ash particles but a weaker non-linear relationship between 4 μm and 63 μm fractions. Both these results show that the coarse mode in the PSD shifts to finer sizes with distance from the volcano in the near-source region. However the equivalent relationships at distances greater than 500 km have not been studied. Furthermore, Horwell (2007) suggests that the effect of wind shear can result in particles of different diameters being transported in different directions. Similarly, aggregation processes, which may be size dependent, may affect the column integrated mass and PSD. High water content and electrostatic charging are believed to enhance deposition through particle aggregation (Ilyinskaya et al., 2011). In this paper we will determine the factors controlling the variability in concentration and PSD in the distal ash cloud.

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2 NAME simulations

NAME is a Lagrangian particle trajectory model designed for many dispersion applications, including the prediction of the dispersion and deposition of volcanic ash in the atmosphere (Jones et al., 2007). Emission of volcanic ash is modelled by releasing ash particles into the model atmosphere with each particle representing a mass of volcanic ash. The model ash particles are carried along by the wind with turbulent mixing represented by giving the trajectories a stochastic perturbation using semi-empirical turbulence profiles. NAME also includes treatments of sedimentation and dry and wet deposition (see Dacre et al., 2011; Webster et al., 2012 for further details).

In this paper, NAME III (version 6.0) is driven using the 3-D winds and thermodynamic fields from the UK Met Office global NWP model analysis fields, updated every 6 h and forecast fields updated every 3 h. Ash concentrations are computed by summing the mass of ash particles in areas of 0.375° latitude by 0.5625° longitude, averaged over 200 m in the vertical and over a time period of 1 hour. Plume height input is taken from measurements provided by the Icelandic Meteorological Office's C-band radar (Petersen and Arason, 2011) and ash is emitted over the top 1 km only. However, it should be noted that Stohl et al. (2011) found, using an inversion method, that the temporal evolution of the plume height may be significantly different from the radar observed values. All of the emitted mass in NAME is distributed among particles with a diameter drawn from DPSD 1, shown in table 1. This distribution is based on an average of measurements made in the plumes from explosive eruptions of Mount Redoubt on 8 January 1990, Mount St Helen's on 18 May 1980, and St Augustine on 8 February 1976 (Hobbs et al., 1991; Leadbetter and Hort, 2011). The ash density in NAME is assumed to be 2300 kg m^{-3} .

ACPD

12, 22587–22627, 2012

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3 Qualitative comparison of NAME simulation and satellite observations

Figure 1 shows the ash cloud as measured by the Infrared Atmospheric Sounding Interferometer (IASI) on board METOP-A and the ash cloud simulated by NAME on the 5, 14 and 17 May. Volcanic ash has a very specific infrared signature, which makes it differentiable from other aerosols and clouds. The specific ash signature depends on the mineral composition and the particle size distribution. A method for detecting ash from high-resolution infrared sounders proposed by Clarisse et al. (2010) has been applied to produce this figure. Measurements have been quantified using an ash absorption index (brightness temperature difference between 1168 cm^{-1} and 1231.5 cm^{-1}) for measurements which pass the ash detection test (and 0 for those that do not). On the 5 May, Fig. 1a and b, a high pressure system was located to the west of the UK in the north Atlantic. Ash emitted from the volcano in Iceland was transported in an anticyclonic direction around the high pressure system. On this day the ash cloud was transported directly from Iceland to the UK. The location of the volcanic ash cloud over Ireland is a little west of the observable IASI detected ash cloud, consistent with the findings of Devenish et al. (2011), but in general is in good agreement with the observable IASI detected ash cloud. Note that on this day the widespread presence of cloud in the region prevented the detection of the majority of the ash cloud (there was a small gap in the cloud over Ireland). It is difficult to draw a conclusion about the accuracy of the ash cloud location based on the IASI measurements. However, assuming that the ash and SO_2 are co-located, the ash cloud position compares well with the GOME-2 SO_2 total column observations on the 5 May (http://atmos.caf.dlr.de/gome/product_so2.html).

Between the 12 and 14 May a low pressure system passed over Iceland travelling from the north-west. This resulted in ash transport to the north and west of Iceland on the 12 May as it was advected cyclonically around the low; towards Europe on the 13 May and finally to the west of Iceland on the 14 May. The location of the volcanic ash cloud both to the west of Iceland and extending in a south-easterly direction covering

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



western Scotland and north-west England is in good agreement with the IASI detected ash cloud, Figs. 1c and d. There was relatively little cloud cover at this time.

On the 17 May, Figs. 1e and f, a high pressure system moved north towards Iceland from the Atlantic resulting in ash transport to the east and then south of Iceland, travelling anticyclonically around the high pressure centre. The location of the volcanic ash cloud over the North Sea and covering the eastern part of Scotland and north-east England is in good agreement with the IASI detected ash cloud.

Overall the spatial correlation of the IASI ash clouds and the highest column loadings in the NAME simulations is very good suggesting that NAME correctly predicted the location of the ash clouds on these 3 days. However, for both 14 and 17 May the high column loadings in the NAME simulated ash cloud do not extend as far south as the IASI detected ash cloud. This may be the result of using a DPSD that contains too large a fraction of heavy particles (investigated further in Sect. 5.2) or due to errors in the plume height or the driving wind fields.

4 Aircraft observations

Over Europe the ash cloud from Eyjafjallajökull was observed by the Facility for Airborne Atmospheric Measurements (FAAM) BAE-146 aircraft. The aircraft was equipped with in-situ particle measuring probes including a Cloud Aerosol Precipitation Spectrometer (CAPS) probe operated by the University of Manchester. The Cloud Aerosol Spectrometer (CAS) component of CAPS was used to derive ash particle size distributions and mass concentrations. The CAS is a forward scattering (4–12 degrees) optical particle counter that measures particles in the nominal diameter range 0.6 – 50 μm . Johnson et al. (2012) showed that the size distribution of ash particles from the Eyjafjallajökull eruption was typically 0.6 – 35 μm and as such the mass concentration could be estimated, to within a factor of two, based on the CAS instrument. Figures 7a, c and e show the locations of three vertical profiles performed by the FAAM aircraft at 12:40 UTC on the 5th, 12:43 UTC on 14 and 14:52 UTC on the 17 May 2010

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



in relation to the Eyjafjallajökull volcano vent. Each of these profiles intersected a distinct ash layer that was free from water or ice clouds and well above the planetary boundary layer such that the aerosol mass was dominated by ash particles.

4.1 Observed particle size distributions

Figure 2a shows the observed column integrated PSD's measured on the FAAM aircraft. Both the magnitude and the shape of the distributions vary depending on the specific time and location at which the observations were taken. The column integrated mass loadings calculated from the CAS mass profiles were 197 mg m^{-2} , 969 mg m^{-2} and 1211 mg m^{-2} on 5, 14 and 17 May respectively. All of the PSD's have their mode at approximately $4 \mu\text{m}$ but the shape of the distributions varies. Figure 2b shows that on 5 and 14 May the percentage of the column integrated mass carried by particles with diameters less than $4 \mu\text{m}$ is very similar (35 %) but the percentage carried by particles with diameters greater than $10 \mu\text{m}$ is much larger on the 14th than on the 5th (20 % compared to 5 %). On 17 May the particle size distribution contains mainly fine ash, with 60 % of the column integrated mass carried by particles with diameters less than $4 \mu\text{m}$ and less than 5 % carried by particles with diameters greater than $10 \mu\text{m}$. The Aerosol Research Lidar Network (EARLINET) and Aerosol Robotic network (AERONET) Sun photometer observations of volcanic aerosol taken at Cabauw, Netherlands, and Hamburg, Munich and Leipzig, Germany during April and May 2010 (Ansmann et al., 2011) also have PSD's with a coarse mode between 1 and $3 \mu\text{m}$.

The aircraft observations provide an excellent opportunity to evaluate volcanic ash transport and dispersion models and to test the sensitivity of the simulated ash cloud to assumptions about the distal eruption source parameters. First it is important to determine whether NAME can simulate column integrated masses and PSD's that are consistent with the observations. If so, we can then use the NAME simulations to determine what controls the variability in both the column integrated mass and the column integrated PSD's seen in the observations.

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



4.2 Ash concentration profiles

When performing a quantitative comparison between the column integrated quantities in the NAME simulations and the aircraft observations it is important to ensure that the column integrated quantities are calculated over an appropriate depth of the atmosphere. Figure 3 shows the vertical profiles of volcanic ash as measured by the FAAM aircraft and simulated by NAME at 12:00 UTC on the 5th, 13:00 UTC on the 14th and 15:00 UTC on the 17 May. The observed and DPSD 1 concentration profiles are discussed in this section and the DPSD 2 concentration profiles are discussed in Sect. 5.3.

On 5 May, Fig. 3a, the ash cloud was observed at an altitude of 3 km and in a very thin layer of less than 1 km depth. The FAAM aircraft performed an ascent between 253 m and 6385 m a.s.l. However, the column integrated mass and particle size distribution, Fig. 2a, was only calculated over the part of the profile where there was a clearly defined ash layer (to minimise contamination of the size distribution with boundary layer or other background aerosol). This layer was between 2733 m and 3121 m. Therefore when comparing the observed and NAME simulated column integrated mass and PSD the NAME quantities should also only be calculated over the part of the profile where there is a clearly defined ash layer. Figure 3a also shows the ash layer from the NAME simulation using DPSD 1. A clearly defined layer of ash is simulated extending between 1 and 4.5 km. This layer is used to calculate the PSD and column integrated mass.

On 14 May, Fig. 3b, the ash layer was observed between 5.5 and 7.5 km. The FAAM aircraft performed a descent between 7312 m and 5491 m a.s.l. The column integrated mass and PSD, Fig. 2a, were calculated over the entire depth of the profile. Figure 3b also shows the ash layer from the NAME simulation using DPSD 1. A clearly defined layer of ash is simulated extending in a broad layer between 1.5 and 6.5 km and a second, lower concentration layer between 7.5 and 9.5 km. The FAAM aircraft did not take measurements above 7.3 km on this flight. Therefore the existence of this upper-

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



layer cannot be verified and only the layer between 1.5 and 6.5 km is used to calculate the PSD and column integrated mass.

On the 17 May, Fig. 3c, the ash layer was observed in a broad layer between 3.5 and 6.5 km with peak concentrations at 5.5 km. The FAAM aircraft performed a descent between 7944 m and 3081 m a.s.l. However, the column integrated mass and PSD, Fig. 2a, was only calculated over the part of the profile where there was a clearly defined ash layer, between 6473 m and 3266 m. Figure 3c also shows the ash layer from the NAME simulation using DPSD 1. The NAME simulation has low-levels of ash extending from the surface to 7 km with peak concentrations occurring at an altitude of 1.5–2.5 km. There is no clearly defined layer of ash in the NAME simulations. In this case the layer from the surface up to 6.5 km is used to calculate the PSD and column integrated mass.

5 Distal eruption source parameters

As discussed in the introduction, in order to estimate the column integrated mass in the distal ash cloud, the important number to quantify is the fraction of total emitted mass that remains in the ash cloud after sedimentation of large particles and near-source microphysical processes have occurred. This distal fine ash fraction (DFAF) is used as a scaling factor that is applied to the model amounts to allow quantitative predictions of ash concentration at long range.

5.1 Distal fine ash fraction

In order to estimate the DFAF the ratio of the NAME column integrated mass, calculated over the depths determined in Sect. 4.2, to the observed column integrated mass is calculated using the method of Dacre et al. (2011).

On 5 May quantitative agreement between NAME and the observed column integrated mass is obtained by assuming that the DFAF is 2.1 %, i.e., the NAME column

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



integrated mass matches the observed column integrated mass if the modeled column integrated mass is scaled by 2.1 %. This suggests that 97.9 % of the total emitted mass falls out close to the source. On 14 May quantitative agreement between NAME and the observed column integrated mass is obtained by assuming that the DFAF is 2.9 %. On 17 May quantitative agreement between NAME and the observed column integrated masses is obtained by assuming that the DFAF is 6.9 %. Thus NAME can simulate column integrated masses that are consistent with the observations if a DFAF of between 2 and 7 % is used. There are of course uncertainties in these estimates arising from inaccuracies in the modeling. For example, Kaminski et al. (2011) show that mass eruption rates deduced from the height reached by the volcanic plume on the basis of scaling laws inferred from models of powerful Plinian plumes may be overestimated for less explosive basaltic eruptions.

5.2 Distal particle size distribution

Figure 4 shows the observed column integrated PSD's on 5, 14 and 17 May re-binned into particle size bins corresponding to the 6 size bins used in NAME (described in Table 1). The mode of the re-binned observed column integrated PSD occurs in the 3.0 and 10.0 μm diameter size bin for all three observations. Figure 4 also shows the NAME column integrated PSD's on the 5, 14 and 17 May at the same locations as their respective observations. The NAME column integrated mass has been re-scaled to match the corresponding observed column integrated mass using the DFAF's calculated in Sect. 5.1. For all 3 cases the NAME simulations using DPSD 1 result in a PSD at the observations location which underestimates the mass carried by particles with diameters between 1.0 and 10.0 μm and overestimates the mass carried by particles with diameters between 10.0 and 30.0 μm . This suggests that DPSD 1 contains a larger fraction of particles with diameters greater than 10 μm than actually remained in the distal ash cloud from Eyjafjallajökull.

In order to match the shape of the observed column integrated PSD in Fig. 4 it is necessary to use modified DPSD's as input for NAME. The measured DPSD's are

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



shown in Fig. 5. For all three cases the measured DPSD's contain a larger percentage of the distal eruption mass in the 1.0 – 3.0 μm and 3.0 – 10.0 μm diameter size bins and a smaller percentage of the distal eruption mass in the 10.0 – 30.0 μm diameter size bin. Note, that as there are no observed particles with diameters less than 0.3 μm or greater than 30.0 μm we therefore assume the mass in these bins is negligible in the distal ash cloud. This re-distribution of mass from larger to smaller particle diameter sizes is consistent with the results of Kristiansen et al. (2012) who compared NAME and FLEXPART simulations to observed PSD's. It is also supported by the idea that phraetomagmatic volcanoes, such as Eyjafjallajökull, emit very fine ash particles (Dellino et al., 2011).

Using an average of the 3 measured DPSD's as a modified input DPSD for NAME, i.e. DPSD 2 shown in Table 1, DFAF's of 5.0%, 2.6 % and 5.9 % are necessary to match 5, 14 and 17 May observed column integrated mass respectively (Fig. 6). Thus the calculated DFAF is sensitive to the DPSD used in the model simulation. These DFAF's are consistent with the DFAF found by Dacre et al. (2011) for an earlier stage of the eruption when comparing NAME simulations with ground based lidar observations, and with that found by Grant et al. (2012) when comparing with airborne lidar observations. Figure 6 shows that for all 3 cases there is a much better agreement between the observed and NAME simulated PSD's when using the modified DPSD.

5.3 Modified ash concentration profiles

In this section the ash concentration profiles for NAME simulations using DPSD's 1 and 2 are compared to the observed ash concentration profiles.

On 5 May, Fig. 3a, the ash cloud was observed at an altitude of 3 km and in a very thin layer of less than 1 km depth. The NAME simulation using DPSD 1 shows a layer of ash extending between 1 and 4.5 km. Although the mean height of the simulated ash cloud agrees with the observations, the NAME layer is 3 times thicker than the observed layer. Performing the NAME simulation but using DPSD 2 (Fig. 3a) results in a NAME ash layer that exhibits a stronger peak at an altitude of 4 km. There are

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



less large 10–30 μm particles in the DSPD and hence the vertical spreading of the ash cloud due to sedimentation is reduced. The shape of the ash cloud agrees better with the observations although the peak in concentrations is 1 km higher. This could be due to errors in the plume height, sedimentation rates or meteorology. However, since ash is emitted over the top 1 km of the plume only, meteorological errors are likely to have a small effect.

On the 14 May, Fig. 3b, the ash layer was observed between 5.5 and 7.5 km. The NAME simulation using DSPD 1 shows a broad layer between 1.5 and 6.5 km and a second, lower concentration layer between 7.5 and 9.5 km. As for 5 May, the structure of the ash layer becomes more peaked when DSPD 2 is used and the height of the peak concentrations shifts upwards by approximately 1 km. The resultant vertical distribution agrees better with the observations suggesting that DSPD 1 contains too large a fraction of heavy particles.

Finally, Fig. 3c shows the ash layer on 17 May. Ash was observed between 3.5 and 6.5 km with peak concentrations at 5.5 km. The NAME simulation using DSPD 1 has low-levels of ash throughout the depth of the troposphere with peak concentrations occurring at an altitude of 2–2.5 km. The simulation using DSPD 2 shifts the low-level peak up by approximately 1 km. The upper-level peak does not change altitude suggesting that it consists predominately of small particles.

Changing from DSPD 1 to DSPD 2 makes the ash layers thinner, more peaked and higher in altitude by approximately 1 km. However, the peak concentrations can be above or below the observed peak by 1 km reflecting the uncertainty in the plume height used in the simulations.

To summarise, NAME can simulate column integrated masses and PSD's that are consistent with the observations if a suitable DFAF and DSPD is used. In the remainder of the paper we use the NAME simulations to investigate what controls the spatial and temporal variability of the column integrated mass and PSD. For the remainder of this paper DSPD 2 and a DFAF of 4.5 % is used in the NAME simulations.

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



6 Column integrated mass

In this section we analyse the spatial and temporal variability of the column integrated mass. First we analyse the three cases for which we have in-situ observations then we use the NAME simulations to make more general conclusions.

6.1 Horizontal variability in column integrated mass

5 May: On the 5 May a high pressure system was located to the west of the UK in the north Atlantic. Ash emitted from the volcano in Iceland was transported in an anti-cyclonic direction around the high pressure system. Figure 7a shows the column integrated mass at the time at which the aircraft profile was taken, 12:00 UTC on 5 May. On this day the ash cloud was transported directly from Iceland to the UK. The horizontal extent of the ash cloud increases as the ash is transported away from the volcano. Generally the column integrated mass decreases both with distance from a maximum along the central ash cloud axis and with distance from the volcano, although there is a second peak in column integrated mass located over Ireland. Figure 7b shows the average age of the ash cloud at the height at which the maximum concentration in the column occurs. The age of the ash cloud represents the mean age of ash in a 12 km by 12 km grid box. The mean age of the ash cloud increases with distance traveled from the volcano as expected. In Figs. 7a and b the black cross shows the location of the aircraft vertical profile. On this day the aircraft observations were taken close to the central ash cloud axis. The NAME simulated ash at this location had an average age of 28 h and a column integrated mass of 197 mg m^{-2} . Figure 8 shows that the plume height 28 h prior to the observation was between 4.5 and 6 km a.s.l.

14 May: Figure 7c shows the column integrated mass at 13:00 UTC on the 14 May. A low pressure system approached Iceland from the north-west on 12 May; on 13 May the low was situated directly over Iceland and on the 14 May the low moved to the south-east of Iceland. Thus ash released on the 12 May was advected cyclonically around the low and ended up north-west of Iceland on the 14th. Ash released on the

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



14 May is aged between 0 and 24 h and has high values of column integrated mass due to an increase in mass eruption rate, corresponding to an increase in plume height from 6.5 to 9 km, that occurred on 13 May (Fig. 8). The aircraft observations on 14 May were taken close to the central ash cloud axis in a region in which the ash had traveled for an average of 26 h from the volcano and a column integrated mass of 969 mg m^{-2} . The plume height 26 h prior to the observation time was between 6 and 6.5 km asl (Fig. 8).

17 May: Figure 7e shows the column integrated mass at 15 UTC on the 17 May. On 17 May ash was transported to the east and then south as it traveled anticyclonically around the high pressure system. This ash is aged between 0 and 48 h. The aircraft observations on 17 May were also taken close to the central ash cloud axis in a region in which the ash had traveled an average of 58 h from the volcano and the column integrated mass was 1211 mg m^{-2} . The plume height 58 h prior to the observation was approximately 6.5 km a.s.l. (Fig. 8).

6.2 Variation of concentration with travel time

Some of the spatial variability in column integrated mass shown in Figs. 7a, c and e is due to the fluctuating mass eruption rate. In this section the NAME simulated age and mass concentrations have been calculated every 6 h during a 14 day period of the eruption (4–18 May). A normalised concentration scale, related to the source properties at the time that the ash was emitted from the volcano, is defined. The normalised concentration is calculated by dividing the concentrations by their respective source concentrations. This should remove most of the variability due to the time varying mass eruption rate. The source concentration, Q , is in kilograms per metre cubed and is calculated using Eq. (1)

$$Q = M / (u \Delta y \Delta h), \quad (1)$$

where M is the mass eruption rate in kilograms per second, u is the horizontal wind-speed at the mean height of the emission in metres per second, Δy is the near-source

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



cross-plume width in metres, and Δh is the vertical distribution of ash at the source. In these NAME simulations Δy is taken to be 5000 m and Δh is set at 1000 m. An average of the normalised concentration in each column is then calculated over the depth of the layer. Finally, the maximum layer average normalised concentration is found for each age and 6 h time window. This represents the layer average normalised concentration at the centre of the ash cloud axis, hereafter termed *normalised concentration*.

For each travel time, the mean and upper and lower quartiles, evaluated over the collection of 6 hourly results were calculated and are shown in Fig. 9a along with the observed normalised concentrations as a function of age. The observed concentrations are normalised by dividing the observed concentrations by the source concentration at the time at which the ash was emitted from the volcano. For an assumed near-source cross-plume width of 5000 m, the concentration drops to 35 % of the source concentration in the first hour after emission from the volcano. This is a result of the rapid dispersion of ash by turbulent eddies represented in the model using parameterised diffusion, and due to the fact that concentrations in these NAME simulations are computed by summing the mass of ash particles in a 0.375° latitude by 0.5625° longitude box. Note, that a cross-plume width of 5000 m was chosen to provide a normalised concentration percentage between 2 and 6 %, in the time range of the observations, consistent with the DFAF values found in Sect. 5.2. Between 6 and 72 h after emission the normalised concentration decreases at a rate that decreases with time. Thus the time to half the concentration gets longer with each successive halving. From 72 h onwards however, the normalised concentration also decreases due to deposition and increased spreading (Fig. 9b).

Since a single layer average ash age is estimated from the NAME model simulation, for the in-situ measurements on 5, 14 and 17 May, the observed range corresponds roughly to the normalised (using the same procedure and with source time estimated using NAME) concentrations calculated using the maximum and minimum observed concentrations in the vertical profile measurements shown in Fig. 3. Figure 9a also shows observed normalised maximum concentrations from lidar observations of the

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ash cloud (Grant et al. (2012), their Fig. 11). For the lidar observations, the range corresponds to the distribution of normalised concentrations calculated using the observed column maximum concentrations measured by the airborne lidar as it flew above the ash cloud, thus these values are likely to be overestimates. Both the model and observations show that after approximately 24 h travel time, changes in ash concentration are small. This suggests that within the distal ash cloud processes leading to the rapid loss of ash from the column, such as microphysical processes or deposition of large particles (not represented in NAME), are not as important as in the near-source region.

Schumann et al. (2010) measured ash concentrations of aged volcanic ash in the distal ash cloud and found that concentrations follow roughly an exponential decay law with half times of order 20 to 22 h. However, their correlation was fairly weak, mainly because of the strong influence of the specific meteorological conditions. Rose et al. (2000) also found that masses of fine ash, as measured by satellite, decrease rapidly in the first 36 h and then decrease much more slowly. They concluded that as much as 75 – 90 % of the fine ash falls out of volcanic clouds in the first 36 h, and is deposited in ash blankets which they viewed as direct evidence for aggregation caused by interaction of volcanic ash particles with ice particles. However, direct evidence for ice in ash blankets is elusive. Due to the different nature of volcanic eruptions it is difficult to make a direct comparison with the Rose et al. (2000) study. However, given that NAME does not include aggregation processes but also simulates a rapid decrease in concentration in the first 36 hours, this suggests that aggregation processes do not play an important role in the distal ash cloud for this eruption.

Disparity between the model and observed concentrations can be due to a wide variety of reasons. For example, the model relationship represents the evolution of the concentrations at the centre of the ash cloud whereas the observations are from a variety of positions in the ash cloud. In addition, whilst the grey shading represents the models 25–75th percentile range, variability in the meteorological conditions can result in a much larger range of possible values. The observed values are very sensitive to the source concentration used to normalise the observed concentrations. A constant layer

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



averaged age was assumed and used to estimate the emission time, and hence the source concentration. During periods in which the source emission rate varied rapidly, small changes in ash emission time can greatly change the source concentration. For example, on 16 May, changing the emission time by 2 h resulted in a change in the range of normalised concentrations from $0.5 \times 10^{-5} - 3 \times 10^{-5} \%$ (off axis) to 0.24 – 1.28 % (dash-dot line). This small time shift is possible due to the uncertainty in the plume location on 16 May (Grant et al., 2012). Finally, if the observations are more patchy and in thinner layers, this will lead to generally higher maximum concentrations.

If we assume that there are no processes leading to the loss of ash from the ash cloud then the inverse of the normalised concentration is proportional to the cross-sectional area ($\Delta y \times \Delta z$) of the ash cloud. The instantaneous spread of the ash cloud, due to gridbox averaging, leads to an initial overestimation of the plume area and hence an underestimation of the normalised concentration. Between 1 and 72 h after emission the ash cloud area increases linearly with time. This is consistent with the following argument: For diffusive growth (on scales bigger than the eddies causing the diffusion) we expect Δy and Δz to increase like $\sqrt{\text{time}}$. Therefore the net effect will be an increase in area that is proportional to time, as shown for the first 24 h after emission. At later times, when the ash cloud gets larger, the range of eddy sizes that contribute to its spreading increases. Therefore we expect to see an acceleration in plume area with Δy and Δz increasing like time. Thus the net effect would be an increase in area that is proportional to time². However, after 24 h Δz has stopped increasing due to the lower ground boundary therefore the area continues to increase at a rate proportional to time until 72 h after emission. After this time, the assumption that loss of ash from the ash cloud is negligible is not valid (Fig. 9a). Thus it appears that the evolution of the normalised concentration during this period can be explained by the geometric spreading of the ash cloud.

Figure 9c shows the normalised ash cloud depth. This is calculated by dividing the ash cloud depth (defined as the vertical extent of the concentrations with $> 1 \%$ of the peak column concentration), at the location of maximum normalised concentration,

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



by the height at which the ash was released at the source. For the first 24 h the normalised ash cloud depth increases from 30–70 % as the ash cloud spreads vertically via the sedimentation of large particles, large-scale ascent or descent and vertical diffusion. From 24 h onwards the normalised ash cloud depth remains fairly constant as the Earth's surface provides a lower bound to the plume vertical spreading.

7 Particle size distribution

In this section we analyse the spatial and temporal variability of the PSD at the location of the maximum column concentrations. First we analyse the PSD for the simplest meteorological case on 5 May. Then we extend this analysis to explain the variability on the 14 and 17 May.

7.1 Spatial variability in particle size distribution

5 May: Figure 10a shows the height of the maximum concentration in the column at 12:00 UTC on 5 May. For travel times between 12 and 24 h, Fig. 7b the ash cloud is sloped from a height of less than 2 km along the north-east edge of the ash cloud to a height of greater than 6 km along the south-west edge of the ash cloud. Further from the volcano (for ash with travel times greater than 36 h) the height of the ash cloud descends from 4 km to less than 2 km as it travels anticyclonically around the high pressure region in the north Atlantic.

A NAME simulation was performed in which a uniform DPSD was used (DPSD 3 in table 1). Figure 10b shows the mode of the PSD at the height of the maximum normalised concentration in the column at 12:00 UTC on 5 May. The PSD in the highest part of the sloped ash cloud has its mode between 0.3 and 3 μm diameter whereas in the lowest part of the sloped ash cloud the particle size distribution has its mode between 10 and 30 μm diameter. This is consistent with the idea that vertical wind shear combined with differential sedimentation of large and small ash particles has separated

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



out the large and small particles spatially leading to variability in the column integrated PSD. Further from the volcano vent the mode of the PSD's moves towards smaller diameter size bins reflecting the deposition of larger ash particles to the surface.

When the same analysis is performed for a simulation in which DPSD 2 is used at the source, Fig. 10c, most of the spatial variability in the mode of the PSD disappears. In the majority of the ash cloud the mode, influenced by the DPSD, remains between 3 and 10 μm and thus the effects of sedimentation and vertical wind shear are masked by the DPSD.

The aircraft profile was taken close to the centre of the ash cloud where the PSD has its mode in the 3–10 μm diameter particle size bin but in a region of vertical wind shear. If the observations had been taken further north-east the PSD would have contained a larger percentage of larger diameter particles due to the combined effects of sedimentation and wind shear. Similarly if the observations had been taken further south-west the PSD would have contained a larger percentage of small diameter particles.

5, 14 and 17 May: In NAME the sedimentation rates for different sized particles are prescribed. Thus we can calculate the average travel time from the volcano for different sized particles in DPSD 2 and can therefore estimate the effect that sedimentation, on its own, has on the PSD as it travels from the volcano to the observation location. Figure 11a shows the cumulative percentage of column integrated mass carried by different sized particles for the three observed cases when the effects of sedimentation only are taken into account. On 5 and 14 May the distributions are almost identical, as expected, since the average travel times are very similar, 28 h and 26 h respectively. The distribution on 17 May however contains a much smaller percentage of mass carried by particles with diameters greater than 10 μm (< 2 %) reflecting the longer average travel time of 58 h. Thus almost all of the large particles have been deposited to the surface. Figure 11b shows the percentage of mass carried by different sized particles for the three observed cases when the effects of sedimentation and transport by the 3-D winds are taken into account. The distribution on the 5th is now different. 3-D transport has a negligible effect on the particle size distribution on the 14 and 17 May.

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



However, on 5 May the effects of sedimentation and 3-D transport (namely vertical wind shear) are greater and result in a higher percentage of modelled mass carried by particles with diameters greater than 10 μm to the observation location. This result is inconsistent with the observations, Fig. 2b which show that the observations on 14 May contain a higher percentage of particles with diameters greater than 10 μm compared to 5 May observations. However, given the strong effect of vertical wind shear on 5 May (Fig. 10b), the NAME simulated PSD is very sensitive to the exact location of the ash cloud. Figure 11b shows that shifting the ash cloud 1 degree to the north(south) produces a PSD with a much larger(smaller) percentage of particles with diameters greater than 10 μm . Ash cloud location errors of this magnitude have been shown to exist by Dacre et al. (2011).

7.2 Temporal variability in particle size distribution

As for the concentration, much of the spatial variability in the column integrated PSD is due to the transport by the 3-D winds. In this section the NAME simulated average age and column integrated PSD's have been analysed in the whole domain every 6 h during a 14 day period of the eruption (4–18 May). Averaging over a range of synoptic conditions reduces the variability due to the 3-D transport. Figure 12a–d show how the total column integrated PSD changes with travel time. The percentage of the column integrated mass in the 0.3 – 1 μm , 1 – 3 μm and 3 – 10 μm diameter particle size bins increases with time balancing the decrease in the percentage of mass in the 10 – 30 μm diameter particle size bin. Initially, during the first 24 h after emission from the volcano, the percentage of the column integrated mass carried by 10 – 30 μm diameter particles decreases rapidly. This is a result of a higher range of sedimentation rates resulting in vertical spreading of these particles (Fig. 12e). The 10 – 30 μm diameter particles are distributed over a greater vertical depth than the 1 – 3 μm and 3 – 10 μm diameter particles and thus are subject to a greater degree of wind shear. Between 24 and 72 h after emission the PSD does not change much as it travels. After 72 h the 10 – 30 μm

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



diameter particles are deposited to the surface resulting in a shift in the particle size distribution to smaller particle sizes.

8 Conclusions

The Eyjafjallajökull volcano in Iceland erupted for 21 days during April and May 2010 emitting large amounts of ash into the atmosphere. Over the UK the ash cloud from Eyjafjallajökull was observed on several occasions by the FAAM BAE-146 aircraft. The aircraft was equipped with in-situ particle measuring probes which measured the concentration of volcanic ash carried by particles of varying sizes. These observations have been used to evaluate both the column integrated mass and PSD from the UK Met Office dispersion model, NAME. The NAME model has also been used to investigate the general dispersion characteristics of volcanic ash.

Quantitative comparison of the observed and NAME simulated column integrated masses at three different locations suggest that between 2 – 6 % of the total emitted mass is transported long distances by small ($< 30 \mu\text{m}$ diameter) ash particles. This is consistent with the results of Dacre et al. (2011) who compared NAME concentrations to ground based lidar estimates for an earlier stage of the eruption, and with the results of Grant et al. (2012) when compared with estimates from an airborne lidar. NAME is also able to simulate the observed column integrated particle size distribution (PSD) if a distal particle size distribution (DPSD, the size distribution of the particles remaining in the ash cloud $> 500 \text{ km}$ or $> 6 \text{ h}$ travel time from the volcano) containing a large fraction of $< 10 \mu\text{m}$ diameter particles is used. This suggests that Eyjafjallajökull emitted particles that were particularly fine possibly due to the interaction of volcanic ash with the ice cap.

By normalising temporal variations in the mass eruption rate and averaging over a range of synoptic conditions the factors controlling the evolution of the concentration and column integrated PSD at the centre of the distal ash cloud were determined.

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Figure 13 summarises the main processes occurring in the distal ash cloud for the Eyjafjallajökull tropospheric eruption.

- Between 6 and 24 h after emission from the volcano the concentration at the centre of the ash cloud decreases due to horizontal dispersion and the depth of the ash cloud increases by 50 % due to sedimentation of 10 – 30 μm diameter particles. The column integrated PSD includes an increasing fraction of < 10 μm particles as the 10 – 30 μm particles are dispersed more widely due to their increased exposure to wind shear.
- Between 24 and 72 h after emission from the volcano the concentration at the centre of the ash cloud again decreases due to horizontal dispersion but the depth of the ash cloud only increases by 10 % due to the bounding limit of the Earth's surface. The PSD remains fairly constant.
- Between 72 and 120 h after emission from the volcano the concentration at the centre of the ash cloud decreases and the column integrated PSD includes an increasing fraction of < 10 μm particles due to the combined effect of horizontal dispersion and deposition of 10 – 30 μm diameter particles to the surface.

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Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



References

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ACPD

12, 22587–22627, 2012

Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. distal particle size distributions (DPSD's). DPSD's 1, 2 and 3 are used as input to the NAME model. Other DPSD's are derived from observations on 5, 14 and 17 May.

	Original DPSD 1	5 May DPSD	14 May DPSD	17 May DPSD	Average DPSD 2	Uniform DPSD 3
Particle diameter (μm)	fraction of mass (%)	fraction of mass (%)	fraction of mass (%)	fraction of mass (%)	fraction of mass (%)	fraction of mass (%)
0.1–0.3	0.1	–	–	–	0.0	16.7
0.3–1.0	0.5	0.7	0.6	0.9	0.7	16.7
1.0–3.0	5.0	29.5	23.5	41.4	31.4	16.7
3.0–10.0	20.0	68.6	53.5	54.6	58.9	16.7
10.0–30.0	70.0	1.2	22.5	3.2	9.0	16.7
30.0–100.0	4.4	–	–	–	0.0	16.7

Volcanic ash clouds

H. F. Dacre et al.

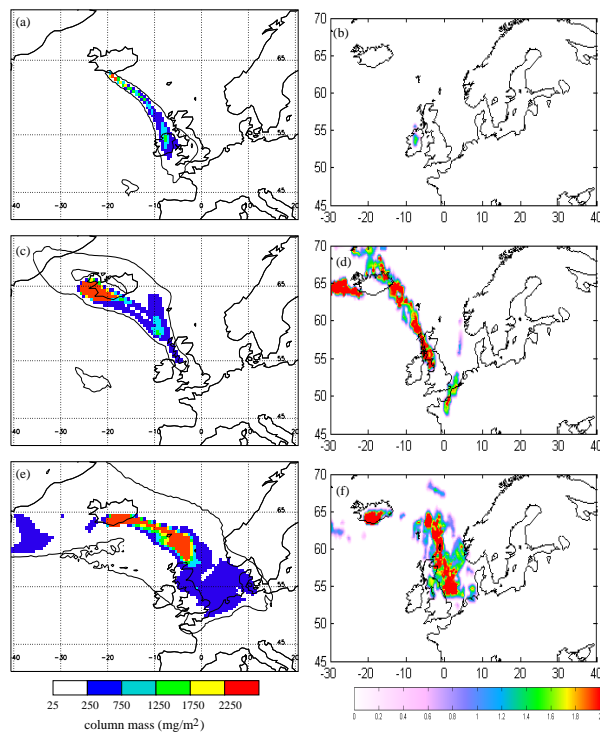


Fig. 1. (a), (c) and (e) NAME column integrated mass using distal particle size distribution 1 and a distal fine ash fraction of 3 %. (b), (d) and (f) IASI measured volcanic ash index (K). at (a) 10:00 UTC and (b) 10:35 UTC on 5 May at (c) 10:00 UTC and (d) 10:49 UTC on 14 May at (e) 10:00 UTC and (f) 09:46 UTC on 17 May.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Volcanic ash clouds

H. F. Dacre et al.

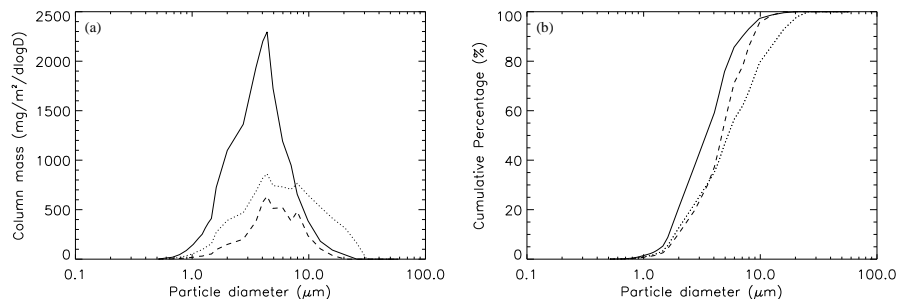


Fig. 2. (a) Observed column integrated mass and (b) Observed column integrated particle size distribution. 12:00 UTC on 5 May (dashed), 13:00 UTC on 14 May (dotted) and 15:00 UTC on 17 May (solid).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Volcanic ash clouds

H. F. Dacre et al.

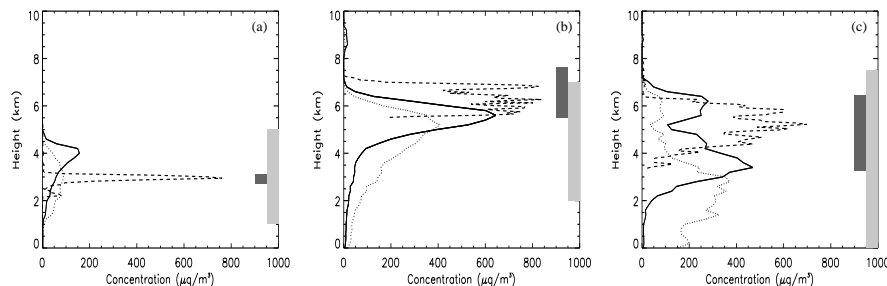


Fig. 3. Concentration profiles on the **(a)** 12:00 UTC on 5 May, **(b)** 13:00 UTC on 14 May and **(c)** 15:00 UTC on 17 May. Observations (dashed), NAME simulation using distal particle size distribution 1 (dotted) and distal particle size distribution 2 (solid). The dark grey bar shows the height range over which the observed particle size distributions are calculated. The light grey bar shows the height range over which the NAME simulated particle size distributions are calculated.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Volcanic ash clouds

H. F. Dacre et al.

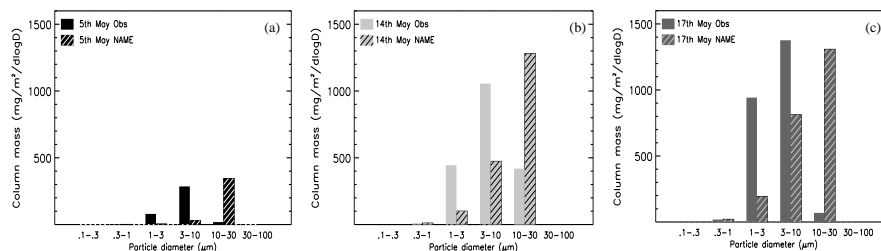


Fig. 4. Re-binned observed column integrated mass (solid fill) and NAME column integrated mass (striped fill) using distal particle size distribution 1. **(a)** 12:00 UTC on 5th May, **(b)** 13:00 UTC on 14 May and **(c)** 15:00 UTC on 17 May.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



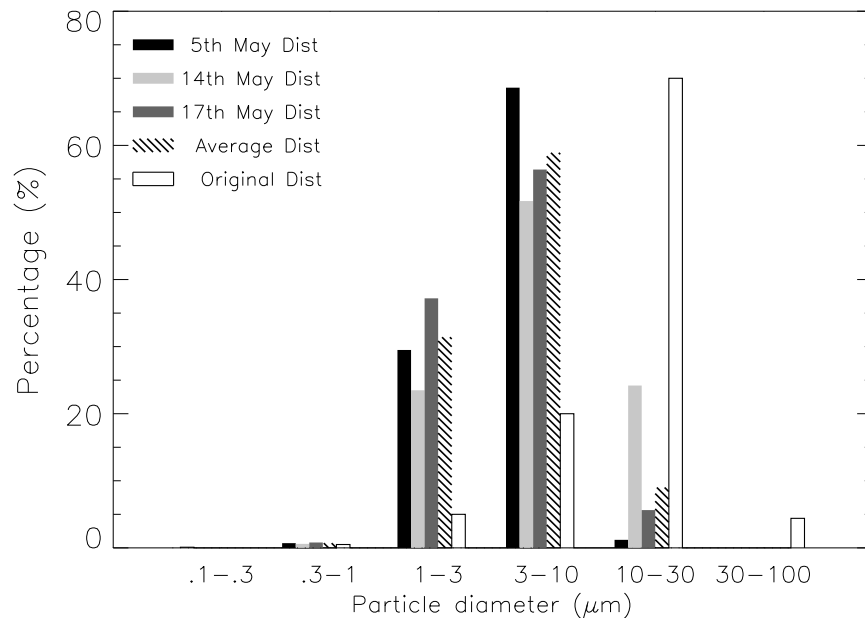


Fig. 5. Distribution of total column mass for distal particle size distribution 1 (white) and derived from observations on 5 May (black), 14 May (light grey) and 17 May (dark grey), average of observations (striped fill).

Volcanic ash clouds

H. F. Dacre et al.

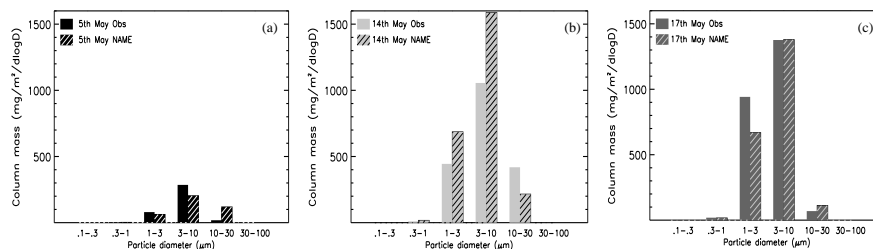


Fig. 6. Re-binned observed column integrated mass (solid fill) and NAME column integrated mass (striped fill) using distal particle size distribution 2. **(a)** 12:00 UTC on 5th May, **(b)** 13 UTC on 14 May and **(c)** 15:00 UTC on 17 May.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Volcanic ash clouds

H. F. Dacre et al.

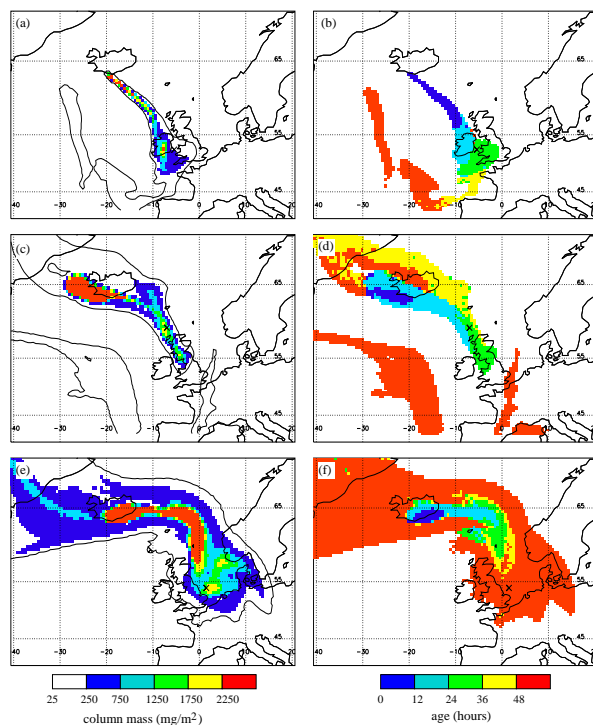


Fig. 7. (a), (c) and (e) Column integrated mass using distal particle size distribution 2 and a distal fine ash fraction of 4.5 %. (b), (d) and (f) average age of the ash cloud at the height at which the maximum concentration in the column occurs. (a) and (b) 12:00 UTC on 5th May, (c) and (d) 13:00 UTC on 14 May, (e) and (f) 15:00 UTC on 17 May. The cross shows the location of the aircraft observations.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I ◀

▶ I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



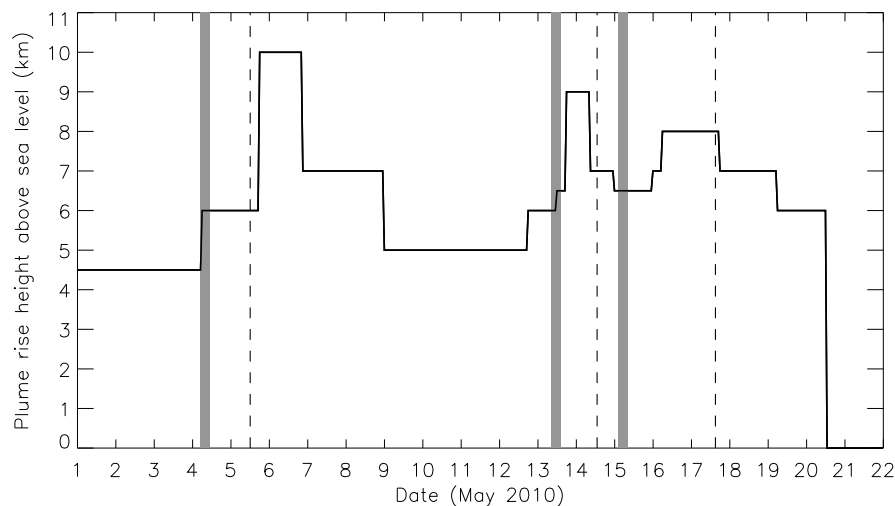


Fig. 8. Timeseries of the height of the eruption plume above sea level. The dashed lines represent the times at which the FAAM aircraft performed profiles through the ash cloud. The grey shading represents the approximate times that the observed ash particles were emitted from the volcano.

Volcanic ash clouds

H. F. Dacre et al.

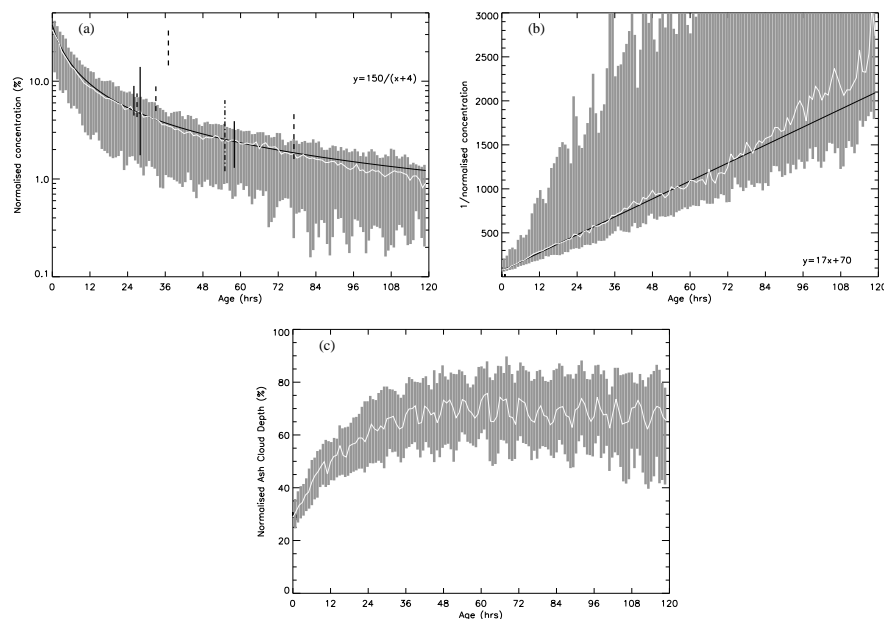


Fig. 9. Mean (solid white line) and 25–75 percentile (grey shading) of **(a)** NAME normalised concentration overlaid with observed normalised concentration as a function of travel time. In-situ measurements (solid), lidar measurements (dashed, dash-dot). Fit to mean (solid black line and equation), **(b)** 1/normalised concentration as a function of travel time. Fit to mean (solid black line and equation) and **(c)** normalised ash cloud depth as a function of travel time.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Volcanic ash clouds

H. F. Dacre et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

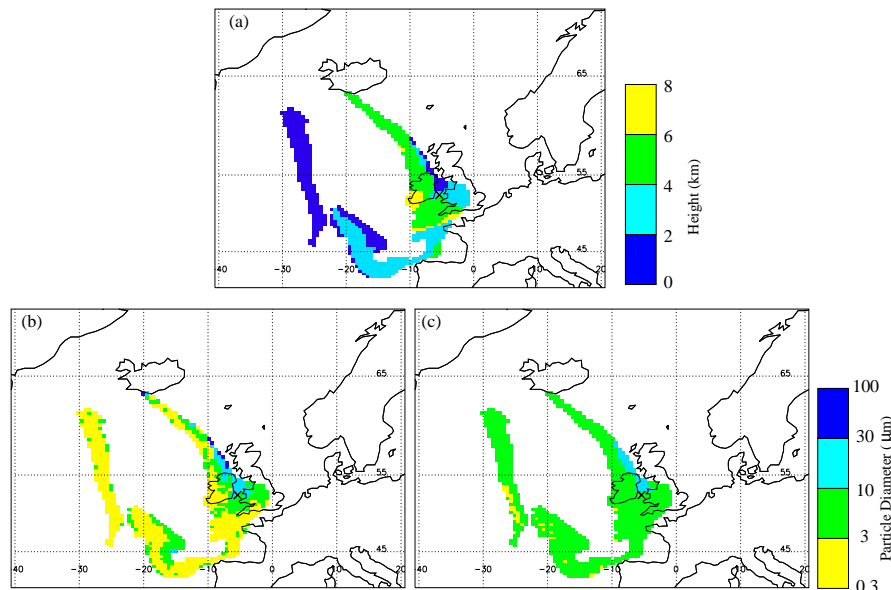


Fig. 10. (a) Height of the maximum mass concentration in each column. Mode of the particle size distribution at the height of the maximum mass concentration in NAME using (b) uniform distal particle size distribution 3 and (c) distal particle size distribution 2. 12:00 UTC on 5 May, the cross shows the location of the aircraft observations.

Volcanic ash clouds

H. F. Dacre et al.

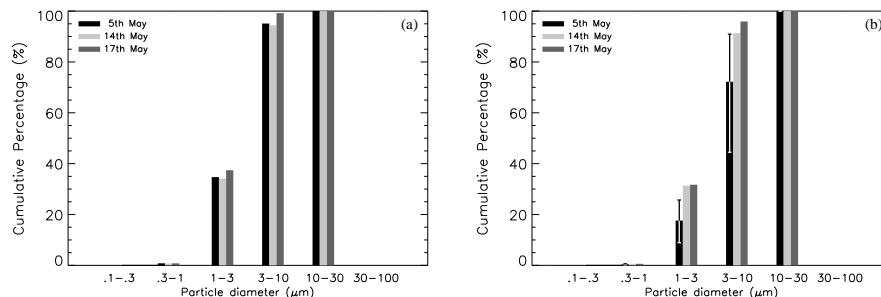


Fig. 11. Modelled column integrated particle size distribution. **(a)** Due to sedimentation of particles only from NAME travel times and simple sedimentation model and **(b)** due to sedimentation and 3-D transport from NAME. At the location of the aircraft observation at 12:00 UTC on 5th May (dashed), 13:00 UTC on 14 May (dotted) and 15:00 UTC on 17 May (solid). The error bars represent the range of particle size distributions simulated by NAME, at the location of the aircraft observation on 5 May, if the ash cloud is shifted 1 degree north (bottom range) or south (top range).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Volcanic ash clouds

H. F. Dacre et al.

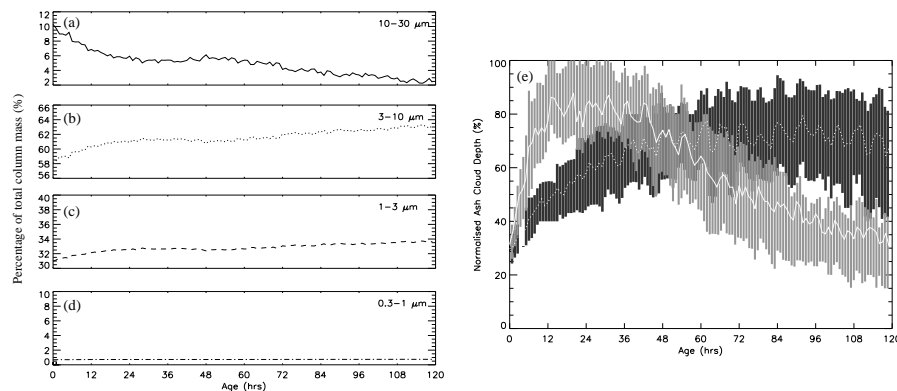


Fig. 12. (a) Mean percentage of total column integrated mass carried by (a) 10–30 μm diameter particles, (b) 3–10 μm particles, (c) 1–3 μm diameter particles and (d) 0.3–1 μm diameter particles. (e) Mean and 25–75th percentile of normalised ash cloud depth for 10–30 μm diameter particles (solid, light grey shading) and 1–3 μm diameter particles (dotted, dark grey shading)

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Volcanic ash clouds

H. F. Dacre et al.

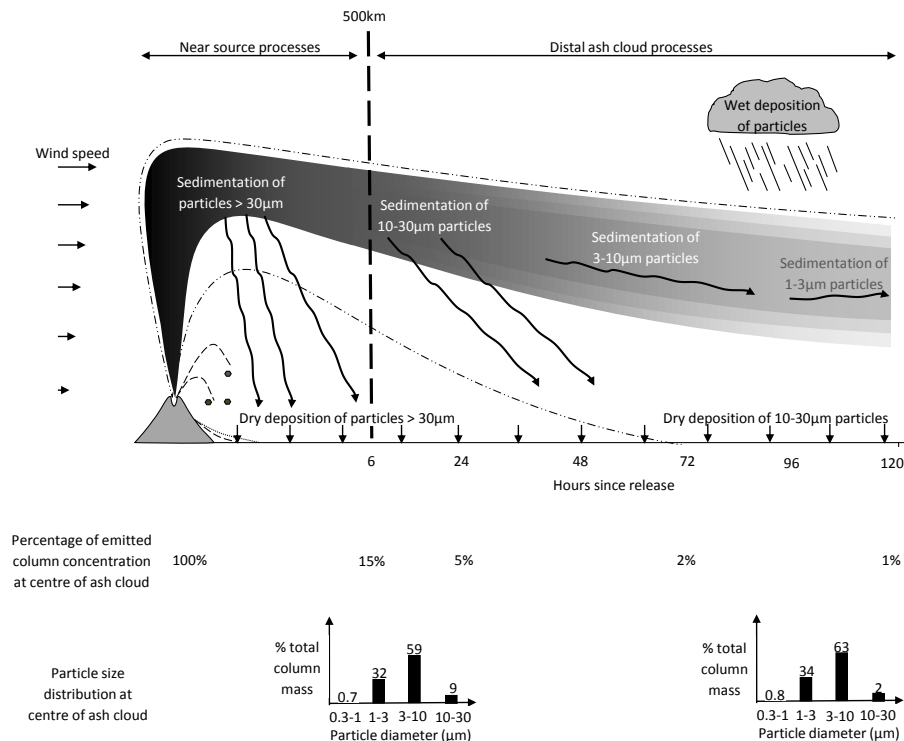


Fig. 13. Schematic showing the processes controlling the evolution of the column average ash concentration and particle size distribution in the distal ash cloud for the Eyjafjallajökull tropospheric eruption. The grey shadings represent the ash concentration, with 1 % peak column concentrations represented by dash-dot contour. Note that, for other eruptions, the column concentration and particle size distributions are likely to depend on the magnitude of the eruption.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

