How assumed composition affects the interpretation of satellite observations of volcanic ash

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ABSTRACT: The monitoring of volcanic ash in the atmosphere by satellite-borne instruments is highly important for generation of warnings of potential ash hazards to aviation, and to constraining model predictions of an ash cloud’s anticipated evolution. The high economic cost of flight restrictions creates a demand for precise monitoring and forecasting; however, no scientific product can be considered precise unless presented with a robust estimate of its associated uncertainty. Data from infrared sensors are focused on, as these monitor the atmosphere both day and night. Most methods for the detection of ash, and the retrieval of its properties, rely on forward modelling to estimate the ash signal at the satellite. This requires assumptions to be made about the ash composition in order to constrain its optical properties as represented in a radiative transfer model. Ash composition may change through the course of an eruption, and is often unknown for new eruptions. Even in cases where the composition of the ash can be sampled, it is unlikely that it is homogeneous enough to match the composition of any of the available optical property datasets exactly (which properties are required for radiative transfer modelling). This often necessary assumption can affect the observed ash signal by an amount that varies with cloud altitude, thickness, and concentration from a few percent to 17.7% for the highest ash concentration examined in this study. This has implications for methods that rely on forward modelling of ash observations, and for the interpretation of real ash observations when ash composition is unknown.

KEY WORDS remote sensing; hazards; modelling

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1. Introduction

It is well known that volcanic ash can pose a danger to aircraft (Casadevall, 1994), cause problems for human health (Horwell et al., 2003), and affect climate (Gow and Williamson, 1971; Robock, 2000; Schmidt et al., 2012), and therefore ash needs to be closely monitored in space and time. The April 2010 eruption of Eyjafjallajökull, Iceland, created a volcanic ash plume that was observed by aircraft (Marenco et al., 2011), by radiosonde (Harrison et al., 2010), and by ground-based instruments both near to the source and at locations across Europe (Ansmann et al., 2011). While these data are valuable, they are incapable of providing a complete depiction of the plume’s evolution in space and time, as radiosondes only provide information on the ash in the area where they are deployed, ground-based measurements generally cover only their immediate vicinity, and for safety reasons aircraft can only fly through the ash plume where ash concentrations are relatively low (Marenco et al., 2011). Only satellite-borne instruments have the spatial and temporal coverage needed for monitoring a volcanic plume that can travel large distances and persist for long periods of time. Satellite-borne sensors recording at infrared (IR) wavelengths are especially useful, as they allow continuous tracking of ash both day and night, and therefore IR wavelengths were concentrated on in this study. In a hazardous situation, information from ground-based instruments, aircraft, radiosondes, and satellite sensors should all be exploited, as each have different strengths and weaknesses and can be considered complimentary; however, these are not always all available. Any responsible data-based decision relies on the data being interpreted in light of their associated uncertainty, and this is particularly the case when data from a range of sources, which may not all agree, must be evaluated.

In recent years, much work has been done towards developing improved methods for detecting, and retrieving information about ash in satellite data. As new methods are put forward, it is important to consider some common assumptions and their likely implications for the certainty associated with ash products. This should both improve the usability of the products and help to focus future development efforts.

Most techniques for retrieving information from satellite data require the use of a radiative transfer model to forward-model anticipated observations of ash (see for example Prata, 1989; Wen and Rose, 1994; Clarisse et al., 2010; Pavolonis, 2010; Francis et al., 2012). Such forward-modelled observations are hereafter referred to as simulated observations. Simulated ash observations are useful in their own right (Kylling et al., 2012; Millington et al., 2012), but are more generally used for the interpretation of actual satellite observations in terms of the ash that they may or may not contain. One of the challenges of forward modelling volcanic ash is that what constitutes ‘volcanic ash’ is poorly constrained, as the term can describe a range of atmospheric states that are physically quite different. An ‘ashy’ atmospheric state can refer to an atmosphere containing ash clouds with a range of different optical and geometrical thicknesses, occurring at different altitudes, with different mass concentrations, composed of
particles with different shapes and sizes, composed of a range of different materials. Simulations of ash observations generally require assumptions for all of these. While previous work has looked at the sensitivity of retrieved ash properties to some of these assumptions (Francis et al., 2012; Millington et al., 2012) this has not followed from an examination of the uncertainty introduced to the simulated observations (which are then used to retrieve the ash properties), but rather from a comparison of the final product, as computed using different assumptions for these properties. It cannot be assumed that other methods of exploiting simulated observations would be subject to the same uncertainty, because this follows from how the simulations are used in the particular retrieval (or detection) method adopted. Furthermore, a large number of arguably equally appropriate choices exist for many of these assumptions, and the above studies, in looking at the effect of many assumptions, investigate only a couple of different choices for each. It is worthwhile to consider the uncertainty attributable to each specific assumption when it is made, so that it can be tracked through the detection and retrieval process and a robust assessment of the accuracy can be made for both detection and retrieved property products.

Most radiative transfer models require a fixed ash density, a particle size distribution (PSD) and a set of wavelength-dependent refractive indices in order to represent ash optical properties effectively. It is often necessary to make the further assumption that ash particles are spherical in order to model scattering effects. While this is very rarely likely to be a true reflection of reality, the effect of the sphericity assumption on radiative transfer calculations at IR wavelengths has been found to be insignificant (Yang et al., 2007). In reality, ash particles have been observed to have a range of densities and can be associated with a range of PSDs (see for example Hobbs et al. (1991) and Schumann et al. (2011)), and these assumptions are likely to contribute to the uncertainty for ash observations simulated using a radiative transfer model. When making a choice for any one of these model requirements, it is important that the uncertainty attributable to that single assumption be considered, so that the implications of changing just that one assumption are understood. While several studies have found specific retrieved ash properties, such as mass, to be more sensitive to the choice of PSD than to the choice of refractive indices (Francis et al., 2012), issues surrounding refractive index selection are also significant (Durant et al., 2009) and are shown here to affect significantly the appearance of ash clouds at IR wavelengths. This work looks specifically at the sensitivity of simulated ash observations to the choice of refractive indices, which follow from the assumed composition of the ash, i.e. what material the ash particles are assumed to be made of.

Too few far-field measurements are available for any consensus to be reached as to what constitutes an appropriate assumption for ash composition. In fact, the wide range of ash compositions that have been observed from different eruptions (see for example Mastin et al. (2009)), and from different stages of the same eruption (see for example, the aircraft-collected ash samples from Eyjafjallajökull examined in Newman et al. (2012)), suggests that ash composition is highly variable. It is therefore probable that the appropriateness of any assumed ash composition will vary with the source of the ash for which it is applied, and it is unlikely that any assumed composition can be considered to be universally appropriate. Previous eruptions of a volcano can provide clues as to the likely composition of ash from a new eruption, but this is not possible for volcanoes that do not have a history of observed eruptions, and can be misleading as different eruptions of the same volcano can produce different tephras (see for example Mullineaux (1986)). Observations of composition can be made by collecting fallout samples on the ground and by flying aircraft into the edge of the plume. However, the composition of ejected magma may change during the course of an eruption (Sigmarsson et al., 2011), and the applicability of measurements made during one stage of an eruption to ash produced during a later phase of the eruption is therefore questionable. Furthermore, such data are generally not available immediately after the onset of an eruption, when warnings may have to be issued to aviation.

An important consequence of the assumed ash composition for radiative transfer modelling is the wavelength-dependent refractive index, \( R \), which describes the optical properties of the material assumed to constitute the ash. \( R \) consists of both a real and an imaginary part, \( R = n + ik \), and is not trivial to measure (Grainger et al., 2012). A look-up table measured from either fine ground rocks and glasses, or from ash fallout samples collected from a historical eruption, is usually selected from the literature and applied to the eruption being studied. Several datasets exist and the estimated composition of the ash being studied is used to select the most appropriate dataset, i.e. the one which was measured for a substance most similar to the ash under scrutiny. It is highly unlikely that any dataset in the literature exactly matches the composition of any given ash cloud, and in cases where the ash composition is unknown, a dataset may be chosen from the literature for a material quite different to the ash cloud. While accepting that such a choice is necessary, it is important that it is made with a full understanding of the implications, and with an appreciation of the uncertainty thereby introduced to the detected ash mask and its retrieved properties.

### 1.1. Aim of this study

The aim of this study is to demonstrate the range in optical properties that results from different assumptions of ash composition, made by selecting different refractive index datasets. The breadth of this range is important, as it indicates the range of simulated observations that are possible for a single ash cloud, even if all other properties (e.g. height, geometric thickness, ash density, PSD) were to be known exactly. Understanding the breadth of this range is important for the manual interpretation of IR observations of ash, and is crucial to any meaningful estimate of the uncertainty associated with satellite-derived volcanic ash data following from any forward-modelling-based technique.

Quantified, robust, uncertainties are necessary in order to assess the reliability of forward-modelled ash observations, and of the detection and retrieval techniques that depend on such simulations. To illustrate this, various refractive index datasets are used to simulate observation spectra for 30 ash clouds, which are modelled identically in all respects except ash composition. Imagery is also simulated for a whole ash cloud from an historical eruption to show how the cloud would appear differently in an image if it had a different composition. Through comparison of these simulations, it is demonstrated that observations of ash are highly sensitive to ash composition.

### 2. Refractive index datasets

Ten different refractive index datasets, which could be taken to represent volcanic ash for the purposes of radiative transfer...
modelling, are listed in Table 1, and are compared in Figures 1 and 2. Volcanic ash can be thought of as a mixed mineral dust and some of the refractive indices considered here are those that have been measured for mineral dust composed of material that could reasonably be expected to be the main constituent of volcanic ash. Other datasets are refractive index data measured from finely ground rocks and glasses, or directly from fall out samples of volcanic ash.

The range of refractive indices that could be used to represent volcanic ash from these datasets is shown in Figure 1, and Figure 2 shows the minimum, maximum and mean taken from all the data. The point is not that any of these are inappropriate for ash, but that in some circumstances, e.g. in the absence of knowledge of the actual ash composition, all can be considered equally appropriate, despite their resulting in quite different simulated observations.

The Pollack datasets (Pollack et al., 1973) have been used extensively in simulations of volcanic ash observations (e.g. Wen and Rose, 1994; Prata and Grant, 2001; Yu et al., 2002; Pavolonis et al., 2006; Stohl et al., 2011; Francis et al., 2012; Kylling et al., 2012; Millington et al., 2012). Refractive index data from the Balkanski datasets have also been used as representative of volcanic ash. For example the 0.9% haematite samples were used in Millington et al. (2012) and Newman et al. (2012), and the 1.5% samples were used in Turnbull et al. (2012). The Volz dataset, measured from ash-fall samples collected from the eruption of Irazu volcano, is the basis of the default ‘volcanic ash’ aerosol optical properties used in the radiative transfer model, RTTOV v10 (Marco Matricardi, personal communication). In 2013, an additional alternative set of volcanic ash optical properties based on the Pollack dataset for andesite was released for use in RTTOV v11 (Saunders et al. (2013)).

The range of values in Figure 1 is striking, particularly at wavelengths between 9 and 10.5 μm. Volcanic ash is associated with a strong absorption feature in this wavelength region, which is exploited by many detection techniques. For example, the reverse absorption method discriminates ash on the basis of the anticipated stronger absorption around 10 μm for ash, in contrast to meteorological clouds, which are known to absorb more strongly around 12 μm (Prata, 1989). The imaginary part of the refractive index in this wavelength region indicates the strength of the expected absorption feature, largely determined by the silicate content of the ash, which many established ash detection techniques rely on exploiting. Figure 1 shows that the Volz dataset describes a much weaker absorption feature at 10 μm than any of the other datasets. The Balkanski 1.5 and

![Image](image_url)

**Figure 1.** The imaginary (a) and real (b) parts of the complex refractive indices measured for volcanic ash, as presented in the datasets described in the text.

![Image](image_url)

**Figure 2.** The mean, maximum, and minimum refractive indices for volcanic ash from all datasets plotted in Figure 1. Real and imaginary parts are shown as $n$ and $k$ respectively.

### Table 1. Refractive index datasets.

| Sample description          | References                        |
|-----------------------------|-----------------------------------|
| Andesite                    | Pollack et al. (1973)             |
| Basalt                      |                                   |
| Obsidian (sampled from      |                                   |
| Oregon)                     |                                   |
| Obsidian (sampled from      |                                   |
| California)                 |                                   |
| Mixed mineral dust containing 0.9% haematite | Balkanski et al. (2007) |
| Mixed mineral dust containing 1.5% haematite |                                   |
| Mixed mineral dust containing 2.7% haematite |                                   |
| Volcanic Dust sampled from  | Volz (1973)                       |
| Irazu Volcano, 1963         |                                   |
| Fallout sample from         | Peters et al. Oxford, ongoing work |
| Eyjafjalljökull, 2010       | (personal communication, 2012)    |
| Fallout sample from Aso, 2011 |                                   |

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2.7% haematite samples both exhibit a much weaker absorption feature than either the Aso or Eyjafjalljökull ash samples. This is interesting, given that the best radiative closure between aircraft-sampled ash from Eyjafjalljökull and the available refractive index datasets (for IR wavelengths) was found for the Balkanski 1.5% haematite dataset (Newman et al., 2012; Turnbull et al., 2012). Other studies of ash from Eyjafjalljökull found refractive indices from andesite to best match their observations (Francis et al., 2012; Millington et al., 2012). This may be interpreted as reflecting the difference between in situ point measurements and remotely sensed measurements of an area. An alternative explanation, however, is that it demonstrates how ash sampled from the same eruption can have a different composition, and so different optical properties, making selection of a truly representative refractive index dataset challenging, even for ash originating from the same source. The similarity between the Balkanski 1.5 and 2.7% haematite samples is also interesting; the maximum difference is 0.0260 and 0.0337 for the real and imaginary parts, respectively. Choosing between these two datasets will therefore have less impact on the forward modelling than a choice, for example, between one of these and the Pollack andesite dataset. The absorption feature (evidenced by the peak in the imaginary part of the refractive index) appears shifted to slightly shorter wavelengths for the haematite and obsidian datasets, which may have consequences for techniques that rely on finding this absorption feature at 10 µm in actual observations where ash is present.

In the case of a new eruption it is unlikely that the ash composition will be known from the outset, and a choice between the available datasets must be made. The appropriateness of this choice may vary throughout the eruption as the ash composition varies. The reasonably large differences between the available data suggest that a choice made at random, i.e. in the absence of any physical information about the ash, will introduce large uncertainties to the optical properties, and therefore to any retrieved ash properties that rely on these.

3. Optical properties

The optical properties generally required by a radiative transfer model were calculated and plotted to examine how they differ according to which refractive indices are used. The PSD used to represent volcanic ash in Millington et al. (2012) was assumed for all the simulated ash clouds, see Figure 3, and all ash was assumed to have a density of 2300 kg m$^{-3}$, following that same work and several other examples in the literature (such as Francis et al. (2012) and Johnson et al. (2012)). Ash particles were assumed to be spherical and Mie scattering calculations were used to represent scattering behaviour. These assumptions for PSD, ash density and particle shape are all likely to be further sources of uncertainty in simulated ash observations, as discussed in Section 1. These parameters are kept fixed here in order to focus on effects specifically attributable to assumed ash composition. Using these values means both that the simulations are realistic and are likely to be performed in an operational monitoring context, and that the difference between simulated observations can be wholly attributed to the difference in refractive indices. Figure 4 shows the range of optical properties calculated using the different refractive index datasets, and Figure 5 shows the mean from all the datasets.

Figure 5 shows that the maximum absolute spread in the calculated absorption cross-sections is 0.0066 µm$^2$, which occurs at a wavelength of 9.1 µm and corresponds to 91% of the mean absorption cross-section at this wavelength. The absolute spread in scattering cross-section is also high at this wavelength, 0.0033 µm$^2$, which is 123% of the mean
to the assumed ash composition. The calculated absorption cross-section at 10 µm is very different when refractive indices calculated from the Aso samples are used, which indicates how unrepresentative the other datasets would have been of those ash samples, and therefore how unsuitable for use in methods for detecting and measuring ash from that eruption (at least for the stage of the eruption that produced those samples, at the distance from source that those samples were collected). The position of the absorption peak in Figure 4 is also interesting, as it varies slightly for the different compositions, notably the absorption peak calculated from the Eyjafjallajökull ash sample is shifted to longer wavelengths than for the other datasets. The location of this absorption peak is known to correspond to longer wavelengths for ash with a lower silicate content (Prata et al., 2013), and it therefore seems likely that the Eyjafjallajökull ash sample was from the earlier more basaltic stages of the eruption.

4. Radiative transfer simulations

Ash clouds were added to two ECMWF atmospheric profiles: one over land (48.23°N, 17.60°E, surface elevation: 130 m, 1 January 2006), and one over sea (58.13°N, 3.20°W, 20 March 2006). These were taken from Chevallier et al. (2006) and ensure that the atmosphere in which the simulations are carried out is realistic. The fast radiative transfer model RTTOV v10.2 (Saunders et al., 1999; Hocking et al., 2011) was used to simulate observations for the infrared atmospheric sounding interferometer (IASI) from these data. RTTOV is the proprietary model used at the London Volcanic Ash Advisory Centre (VAAC) at the Met Office in the United Kingdom to interpret satellite radiance data, and so using it here ensures the replication of an operational process. It was decided to simulate observations from the IASI sensor because it has the highest spectral resolution in the IR of currently operational sensors, and is therefore appropriate for investigation of the effect of assumed ash composition on the whole IR spectrum. Ash clouds were added to the profiles with concentrations of 200, 300, 500, 1000 and 3000 µg m⁻³ at the altitudes detailed in Table 2. This range covers the minimum ash concentration for a volcanic ash advisory alert (Volcanic Ash Advisory Centre – London, 2011) up to a thick ash plume, where forward modelling sensitivity to the choice of refractive indices is expected to be weak (as the optical properties for all ash compositions tend towards optical saturation for all wavelengths).

The simulated cloud observations are grouped by ash concentration. A clear sky observation is also simulated from each of the atmospheric profiles, and the difference between this and the simulated ash observations is interpreted as the ‘ash signal’. The wavelength-dependent standard deviation of the ash signal for clouds of different thicknesses and at different altitudes is plotted for each group in Figures 6 and 7. Each standard deviation

Table 2. The altitudes at which ash clouds were simulated.

| Land/sea profile | Cloud base/top | Altitude (km) |
|------------------|----------------|---------------|
| Land             | Base           | 13.4          | 10.6          | 7.8           | 13.9         | 11.0         | 8.2          |
|                  | Top            | 15.1          | 12.2          | 9.4           | 14.7         | 11.8         | 9.0          |
| Sea              | Base           | 13.4          | 10.4          | 7.7           | 13.8         | 10.8         | 8.1          |
|                  | Top            | 15.1          | 12.1          | 9.3           | 14.6         | 11.7         | 8.9          |

Slightly different altitudes were used for the two profiles to allow the ash clouds to completely fill model layers in the radiative transfer model, which are defined at fixed pressures rather than altitudes.
How assumed composition affects interpretation of ash observations

5. Simulated IASI images

To illustrate how the sensitivity of ash spectra to ash composition affects IR images that might be used by forecasters for monitoring an ash hazard, such images were simulated for whole ash clouds from the Eyjafjallajökull eruption. The same ash cloud was used to simulate multiple images, changing only the assumed composition of the ash, as described below. The difference between the resulting images highlights the care that should be taken in interpreting such imagery when ash composition is unknown.

IASI data were simulated using the fast radiative transfer model RTTOV (as used above) and Met Office Numerical Weather Prediction data (Davies et al., 2005). The scattering and absorption co-efficients shown in Figure 4 were used in RTTOV in order to simulate the top of the atmosphere brightness temperatures for IASI. Ash concentration data from the Met Office’s atmospheric dispersion model for the dispersed ash cloud from the 2010 Eyjafjallajökull eruption were used as input data. The simulations follow the method of Millington et al. (2012) developed for the simulation of Spinning Enhanced Visible and Infrared Imager (SEVIRI) data. The IASI channels closest to the central wavelengths of the SEVIRI channels used were chosen. These were IASI channels 2019, 1125, and 754 at wavelengths of 8.7, 10.8, and 12.0 µm, respectively. The simulated brightness temperatures were used to create images of the Eyjafjallajökull ash cloud at 1200 UTC on 7
Figure 7. Standard deviation for the deviation of simulated ash observations from simulated clear-sky observations over sea. The key in (a) shows the vertical location and extent of the ash. Thick clouds are shown in blue and thin clouds in red. The simulated ash concentrations are 200, 300, 500, 1000, and 3000 µg m⁻³ for (a)–(e) respectively.

May 2010 (Figures 8 and 9). These are similar to those produced operationally at the London VAAC for SEVIRI to aid forecasters in forecasting the dispersion of volcanic ash, as described in Millington et al. (2012).

The 10.8–12.0 µm brightness temperature difference (BTD) image employs the reverse absorption method described earlier; here volcanic ash clouds have negative values and meteorological clouds have positive values (in general). The strongest BTD signals are observed for ash simulated using refractive indices for andesite, basalt, Aso and Eyjafjalljökull because these have the greatest gradient between 10.8 and 12.0 µm in the absorption cross-sections, as shown in Figure 4.

The dust RGB images were produced by assigning the 12.0–10.8 µm BTD to the red component of the image, the 10.8–8.7 µm BTD to the green component and the 10.8 µm brightness temperatures to the blue component. The variation in the gradient of the absorption cross-sections between 8.7 and 10.8 µm affects the green component of the images in addition to the variation in the red component owing to variations in the strength of the reverse absorption effect. This results in a yellowish colour for ash clouds simulated using the desert dust and obsidian refractive indices because of the relatively small gradient between the absorption cross-sections at 8.7 and 10.8 µm compared with the other datasets.

6. Discussion

Different choices of assumed ash composition, and therefore of appropriate refractive indices, result in a large spread in the observations simulated for any single ash cloud over either land or sea. Figures 6 and 7 show that the spread is especially

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broad for geometrically thicker and denser clouds. This has significant implications for retrievals of ash mass from satellite data that rely on forward modelling observations of ash of a particular composition. The spread is particularly high to either side of the absorption feature at 10 \( \mu \text{m} \). This is important because the gradient of the slope between the radiances measured at this feature and at slightly longer wavelengths is important to calculations of ash mass (Wen and Rose, 1994; Prata and Prata, 2012). Francis et al. (2012) looked into the effect of using different refractive indices to calculate the absorption co-efficients for the retrieval of volcanic ash properties from SEVIRI data. There were significant differences in the retrieved values of ash column loading, for example of the order of a factor of 3 between values retrieved assuming mixed mineral dust containing 1.5% haematite (Balkanski et al., 2007) and volcanic dust sampled from Irazu volcano (Volz, 1973), (see figure 5 in Francis et al., 2012). Francis et al. (2012) also compared SEVIRI-retrieved ash heights and ash heights derived from CALIOP data and found that ash heights retrieved from SEVIRI data assuming a composition of andesite (Pollack et al., 1973) and volcanic dust (Volz, 1973) best matched the CALIOP data for the data available for the 2010 Eyjafjallajökull eruption.

It is unlikely that any operational scheme for detection and retrieval of ash mass will exploit refractive indices calculated for material that exactly matches the composition of the ash under analysis. At best, a close match exists between the observed ash composition and material in the literature for which refractive indices have been calculated; at worst, no observations of the ash composition exist, and no pertinent historical information is available and a refractive index dataset must be chosen from the literature at random.

The composition of the volcanic ash also has a significant impact on the strength of volcanic ash signals in commonly used satellite imagery products, as shown in Figure 8. This has implications for the visual interpretation of the imagery in an operational setting. The same quantity of ash can produce quite different signals when composed of different material, and thus
the coverage of volcanic ash and anticipated aviation hazard can be misinterpreted if an inappropriate composition is assumed. Figures 6 and 7 show that the amount by which an observed IR ash spectrum deviates from a clear sky spectrum varies with composition of the ash, and show this variation to be particularly high between 9 and 11 µm, which is the region most often exploited for detection of ash and retrieval of its properties. The maximum spread over land and sea is 17.4 and 17.7% respectively. In both cases this corresponds to a cloud with the highest simulated ash concentration (3000 µg m⁻³) with an altitude between 13.4 and 15.1 km. This peak in the sensitivity of the signal to assumed composition occurs at a wavelength of 9.1 µm for observations simulated over both land and sea. Some sensitivity is evident even for geometrically thin ash clouds with low ash concentration, for example ash between 13.8 and 14.6 km (13.9 and 14.7 km over land) with a concentration of 200 µg m⁻³ results in a signal that differs by 0.5% over both land and sea when different compositions are assumed. When the thickness of the cloud increases this sensitivity unsurprisingly also increases; for example a cloud with the same concentration extending from 13.4 to 15.1 km corresponds to a spread in sensitivity to the assumed composition of 1.0 and 1.1% for land and sea, respectively. At higher ash concentrations the sensitivity increases; for example, a cloud between 10.4 and 12.1 km (10.6 and 12.2 km over land) with a concentration of 1000 µg m⁻³ corresponds to a spread of 5.0 and 5.2% for land and sea, respectively, while a cloud at the same altitude with a concentration of 3000 µg m⁻³ corresponds to a spread of 15.9 and 16.4% for land and sea, respectively. The sensitivity to assumed composition also varies with the altitude of the ash; for example an ash cloud with a concentration of 500 µg m⁻³ corresponds to a spread of 1.2 and 1.3% over land and sea, respectively when it is between 8.1 and 8.9 km, whereas when it is between 10.8 and 11.7 km the spread is 1.9% for both land and sea, and when it is between 13.8 and 14.6 km the spread is 2.0 and 2.1% for land and sea, respectively. The wavelength at which the spread is greatest, i.e. at which the signal is most sensitive to the choice of assumed composition, is 9.1 µm for observations simulated over both land and sea in all cases, except for when the concentration is 500 µg m⁻³ in which case the peak sensitivity of some clouds is seen at 9.5 µm. The signal in this wavelength region is often interpreted to estimate the amount of ash present in an observation, without consideration of the effect of assumed composition. These results demonstrate that while this sensitivity is a source of uncertainty in the results, it may not be straightforward to account for or to correct for since the sensitivity varies with the ash altitude, concentration, and geometric thickness.

These results suggest a need for more in situ measurements to be made of volcanic ash in the far field. A dataset of such measurements, ideally including data from a wide range of eruptions and atmospheric conditions, would improve understanding of the range of spectral properties appropriate for ash and would be useful to the development of future detection and retrieval techniques. It is important that a wide range of atmospheric conditions be represented in such a dataset because observations of ash vary with ash composition, as shown in this study, and also with the composition of the atmosphere, notably with the amount of water vapour present (Prata et al., 2001). An observation of ash in the tropics may therefore be distinct enough not to be associated with a dataset of ash observations made in dry atmospheric conditions, even if the ash composition was comparable. These measurements could help us to move towards a consensus on which compositions are most representative of ash (this is likely to be dependent on location and eruption-type), and how to characterize the uncertainty associated with this assumption appropriately.

7. Conclusions

Simulations of ash observations depend on an assumed ash composition to determine the ash optical properties. There is often little to base such an assumption on, and it is practical to make it a choice from one of a range of compositions for which optical properties have been studied. For this reason, it is likely that in many, if not all, cases, the actual ash composition and optical properties will differ from those used to simulate the observation. This situation is unlikely to change in the future, meaning assumed ash composition is likely to remain a large source of uncertainty in simulated ash observations and in any technique that relies on them to detect ash or to retrieve properties such as mass. This work shows that the sensitivity of observations at IR wavelengths to ash varies with the composition assumed for the ash by up to 17.4 and 17.7% for high concentrations of ash over land and sea respectively. The results show that this uncertainty varies with cloud altitude and geometric thickness as well as with ash concentration. In this study, observations of higher altitude ash clouds are associated with greater sensitivity to assumed composition, and a slightly higher sensitivity is evident for observations over sea compared with those over land. This implies that an even greater spread in observations could follow from higher concentrations of ash, or for geometrically thicker ash clouds at higher altitudes. These uncertainties also have implications for the visual interpretation of IR satellite imagery for hazard monitoring, and this should be carried out with an appreciation of how the ash composition can affect the imagery.

Responsible interpretation of satellite-derived ash data and their evaluation alongside data from other sources relies on the data being presented with a robust and reliable estimate of its associated uncertainties. Further work into characterizing uncertainties attributable to assumed ash composition will make satellite-derived ash products more useable and allow decisions to be made based on a more informed evaluation of data from different sources.

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References

Ansmann A, Tesche M, Seifert P, Gross S, Freundenthaler V, Apituley A, Wilson KM, Serikov I, Linne H, Heinold B, Hiebsch A, Schneel F, Schmidt I, Mattis I, Wadinger R, Wiegner M. 2011. Ash and fine-mode particle mass profiles from EARLINET-AERONET observations over central Europe after the eruptions of the Eyjafjalljökullvolcano in 2010. J. Geophys. Res. 116: D00U02, DOI: 10.1029/2010JD015567.

Balkanski Y, Schulz M, Claquin T, Guibert S. 2007. Reevaluation of satellite and AERONET data. Atmos. Chem. Phys. 7: 81–95.

Casadevall TJ. 1994. The 1989–1990 eruption of Mount Redoubt Volcano, Alaska: impacts on aircraft operations. J. Volcanol. Geotherm. Res. 62: 301–316.

Chevallier F, Di Michele S, McNally AP. 2006. Diverse profile datasets from the ECMWF 91-level short-range forecasts. Document No. NWPSAF-EC-TR-010, EUMETSAT NWP SAF.
Clarisse L, Hurtmans D, Prata AJ, Karagulian F, Clerbaux C, De Maziere M, Coheur PF. 2010. Retrieval of physical properties of volcanic ash using Meteosat: a study case from the 2010 Eyjafjallajökull eruption. J. Geophys. Res. 117: D00039, DOI: 10.1029/2011JD016788.

Gow AJ, Williamson T. 1971. Volcanic ash in the Antarctic ice sheet and its possible climatic implications. Earth Planet. Sci. Lett. 13: 211–218.

Grainer RG, Peters DM, Thomas AJA, Siddans R, Carboni M, Davies T, Cullen MJP, Malcolm AJ, Mawson MH, Staniforth A, White AJA, Wood NN. 2005. A new dynamical core for the Met Office’s global and regional modelling of the atmosphere. Q. J. R. Meteorol. Soc. 131: 1759–1782, DOI: 10.1256/qj.04.101.

Davies T, Cullen MJP, Malcolm AJ, Mawson MH, Staniforth A, White AA, Wood NN. 2005. A new dynamical core for the Met Office’s global and regional modelling of the atmosphere. Q. J. R. Meteorol. Soc. 131: 1759–1782, DOI: 10.1256/qj.04.101.

Durant A, Harrison SP, Watson IM, Balkansky S. 2009. Sensitivity of direct radiative forcing to dust particle characteristics. Prog. Phys. Geogr. 33: 80–102.

Francisco P, Cooley MC, Saunders RW. 2012. Retrieval of physical properties of volcanic ash using Meteosat: a study case from the 2010 Eyjafjallajökull eruption. J. Geophys. Res. 117: D00039, DOI: 10.1029/2011JD016788.

Prata AJ, Prata AT. 2012. Eyjafjallajökull volcanic ash concentrations determined using Spin Enhanced Visible and Infrared Imager measurements. J. Geophys. Res. 117: D00023, DOI: 10.1029/2011JD016800.

Prata AJ, Bluth GJS, Werner C, Realminu VI, Barn SA, Watson IM. 2013. Gas Emissions from Volcanoes. In Monitoring Volcanoes in the North Pacific: Observations from Space, Dean KG, Dehn J (eds). Springer-Praxis Books: Chichester, UK.

Robock A. 2000. Volcanic eruptions and climate. Rev. Geophys. 38: 191–219.

Saunders R, Matricardi M, Brunel P. 1999. An improved fast radiative transfer model for assimilation of satellite radiance observations. Q. J. R. Meteorol. Soc. 125: 1407–1429.

Saunders R, Hocking J, Runde D, Rayer P, Matricardi M, Geer A, Lupe C, Brunel P. Vidot J. 2013. RTTOV-11 Science and Validation Report. http://research.metoffice.gov.uk/research/interproj/nwpasi/rttm/docs_rttov11/users_guide_v1.5.pdf [accessed 26 November 2013].

Schmidt A, Carslaw KS, Mann GW, Rap A, Pringle KJ, Spracklen DV, Wilson M, Forster PM. 2012. Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate. Atmos. Chem. Phys. 12: 6009–6058.

Schumann U, Weinzierl B, Reitebuch O, Schlager H, Minikin A, Forster C, Baumber R, Sailer T, Graf K, Mannstein H, Voigt C, Rahm S, Simmet R, Scheibe M, Lichtenstein M, Stock P, Rüba H, Schäuble D, Tafferner A, Rautenhaus M, Gierz T, Ziereis H, Krautstrunk M, Mallaun C, Gayet JF, Liefke K, Kandler K, Ebert M, Weinbruch S, Stohl A, Gasteiger J, Groß S, Freudenthaler V, Weingärtner A, Ansmann A, Tesche M, Olafsson H, Sturm K. 2011. Airborne observations of the Eyjafjallajökull volcanic ash cloud over Europe during air space closure in April and May 2010. Atmos. Chem. Phys. 11: 2245–2279.

Sigmarsson O, Vlastic I, Andreassen R, Bindeman I, Deviadal JJ, Mone S, Keiding JK, Larsen G, Hoskuldsson A, Thordarson T. 2011. Remobilization of silicic intrusion by malic magmas during the 2010 Eyjafjallajökull eruption. Solid Earth 2: 271–281.

Turnbull K, Johnson B, Marrone F, Haywood J, Minikin A, Freudenthaler V, Ansmann A, Tesche M, Olafsson H, Sturm K. 2011. Determination of size and height resolved volcanic ash emissions and their use for quantitative ash dispersion modelling: the 2010 Eyjafjallajökull eruption. Atmos. Chem. Phys. 11: 4333–4351.

Turnbull K, Johnson B, Marrone F, Haywood J, Minikin A, Weinzierl B, Schlager H, Schumann U, Leadbetter S, Woolley A. 2012. A case study of observations of volcanic ash from the Eyjafjallajökull eruption: In situ airborne observations. J. Geophys. Res. 117: D0012, DOI: 10.1029/2011JD016688.

Völz FE. 1973. Infrared optical constants of ammonium sulphate, Sahara dust, volcanic pumice and flyash. Appl. Optics 12: 564–568.

Wen S, Rose W. 1994. Retrieval of sizes and total masses of particles in volcanic clouds using AVHRR bands 4 and 5. J. Geophys. Res. 99(D5): 5421–5431.

Yang P, Feng Q, Hong G, Kattawar GW, Wiscombe WJ, Mishchenko DO, Lasko I, Sokolik IN. 2007. Modeling of the scattering and radiative properties of nonspherical dust-like aerosols. Aerosol Sci. 38: 995–1014.

Yu T, Rose WI, Prata AJ. 2002. Atmospheric correction for satellite-based volcanic ash mapping and retrievals using “split window” IR data from GOES and AVHRR. J. Geophys. Res. 107: D16, DOI: 10.1029/2001JD000706.