Measuring Volcanic Ash With High-Spectral Resolution Infrared Sounders: Role of Refractive Indices

Alexandre Deguine, Lieven Clarisse, Hervé Herbin, and Denis Petitprez

***Abstract*— Airborne volcanic ash can be observed and quantified from hyperspectral infrared (IR) sounders. The retrieval process of physical quantities, such as particle radius and mass, depends critically on the assumed spectrally dependent complex refractive indices (CRIs) that are used. Traditionally, the Pollack et al. (1973) dataset was used almost exclusively. These indices are, however, based on measurements of rock slabs, and in recent years, two datasets have become available from laboratory measurements of ash in suspension, the Reed et al. (2018) and Deguine et al. (2020) datasets. Here, we compare the three datasets and quantify the extent to which each of them can be used to simulate satellite observed spectra of real volcanic ash plumes. We find that whereas the Pollack et al. indices perform worst throughout, the performance of the other two is comparable for andesitic and basaltic ash plumes. However, all three datasets have difficulty in reproducing the extinction minimum around 1250 cm^{-1}. The Reed et al. indices in addition yield inconsistent results in the 800–850 cm^{-1} range. The Deguine et al. dataset is the only one, which can be used to reproduce the very large spectral signatures often observed with rhyolitic ash across the entire thermal IR window 750–1250 cm^{-1}. In terms of retrieved quantities, the largest differences are seen for the radius, with the Deguine et al. dataset resulting in the smallest retrieved particle sizes.**

***Index Terms*— Refractive index (RI), remote sensing, volcanic ash.**

I. Introduction

IN view of the risks posed by volcanic eruptions and their impact on environment and climate, it is essential to monitor volcanoes and track and quantify eruptive plumes [1]. Volcanic ash can be measured with remote sensing techniques in the visible and ultraviolet [2] and in the infrared (IR) [3] spectral domain. IR observations have a good sensitivity to both small and larger particles, can be used to measure at night, and allow distinguishing volcanic ash from other aerosol. In addition, hyperspectral IR sounders, such as the infrared atmospheric sounding interferometer (IASI) [4], allow to quantify physical properties of ash, such as effective radii, optical depth (OD), mass, altitude, and even composition, by exploiting the spectral variations in absorption and scattering coefficients in the 7.5–12.5-μm spectral range [5], [6], [7], [8].

Most algorithms rely on the availability of accurate external information on the optical properties of volcanic ash in the form of spectrally dependent complex refractive indices (CRIs) [9]. The CRI of a material carries information on both the absorption and the scattering characteristics and can be used to characterize the optical properties as a function of particle size. In recent years, exploiting state-of-the-art experimental techniques, several new datasets of volcanic ash CRI have been published [10], [11], [12], covering a wide range of different ash types. Current available CRIs, even those measured on ash from the same eruption, exhibits large differences, as underlined in [11]. The goal of this letter is to characterize to what extend these differences impact the retrieval of physical parameters.

In the next section, we present the existing refractive indices that are the subject of this study and show how their derived extinction and scattering coefficients differ. In Section III, we perform a detailed comparison between satellite observed spectra of volcanic ash plume, with those simulated with a forward model using different sets of refractive indices. Going beyond individual observations, Section IV presents a case study of a large plume observed from the 2010 Eyjafjallajökull eruption. We end this letter with a short summary of the main findings.

II. Datasets

In this study, we use and compare three different datasets of refractive indices. The first one is the Pollack dataset [13] that has been used very widely in the past to retrieve physical parameters for volcanic plumes. Nowadays, its use is challenged, as these refractive indices are based on measurements on bulk materials (polished slabs of rock). The two other datasets, Reed et al. [10] and Deguine et al. [11], are

Manuscript received 20 September 2022; revised 8 February 2023; accepted 21 March 2023. Date of publication 23 March 2023; date of current version 5 April 2023. This work was supported in part by the French National Research Agency (ANR) through the Programme d'Investissement d'Avenir (PIA) for the Chemical and Physical Properties of the Atmosphere (CaPPA) Project under Contract ANR-11-LABX-0 0 05-01, in part by the Regional Council Nord Pas de Calais-Picardie, and in part by the European Funds for Regional Economic Development (FEDER). *(Corresponding author: Alexandre Deguine.)*

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Digital Object Identifier 10.1109/LGRS.2023.3261202

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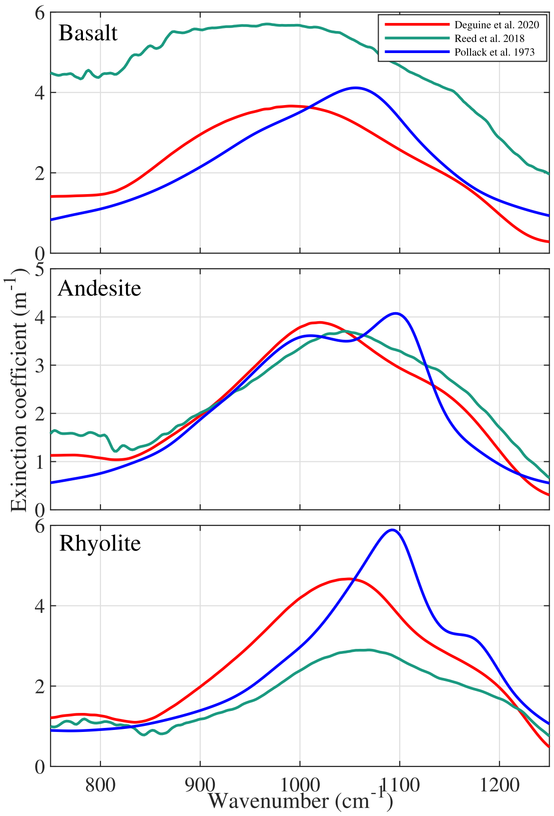

Fig. 1. Spectral variation of the extinction coefficient from the three sets of refractive indices and the three types of ash.

based on laboratory measurements from real volcanic ash, suspended in air. Even though both datasets were retrieved using similar experimental techniques, as we will see, they exhibit significant differences in the IR spectral region, most likely due to the difference in the aerosol model that was used to retrieve the refractive indices. For instance, in the case of Reed, the retrieval uses the Rayleigh continuous distribution of ellipsoids model, while Deguine used the more common Mie theory. It should be kept in mind that, even if the sample originates from the same volcano, the chemical composition, the size distribution, and/or the crystalline/amorphous fraction can significantly vary from a sample to another leading to differences in the CRI retrieval [14].

From each of the three datasets, we selected three refractive indices with contrasting chemical composition as follows.

1) *Basalt ($SiO_2 \leq 50\%$):* The Pollack refractive indices include a set derived from measurements of a non-glassy basaltic rock. The two other datasets both contain measurements of basaltic ash from the 2011 Grímsvötn eruption (Iceland) [6].
2) *Andesite ($55\% \leq SiO_2 \leq 60\%$):* The Pollack dataset has one andesitic rock sample. The two other datasets contain measurements of ash from the 2010 Eyjafjallajökull eruptions (Iceland), which released mostly trachy-andesitic ash [15].
3) *Rhyolite ($68\% \geq SiO_2$):* The refractive indices from the non-glassy obsidian rock sample are used from Pollack. From the Deguine dataset, we selected the refractive indices from ash of the 2011 Puyehue–Cordón Caulle eruption [16], and from the Reed dataset, we selected the CRI from ash of the Nisyros volcano. While, technically, the Nisyros sample is categorized as dacite because of its lower Alkali content, both samples are almost identical in terms of SiO_2 fraction (69.65% and 69.67%, respectively).

Fig. 1 compares the extinction coefficient for these three categories of volcanic ash as derived from the three datasets of refractive indices using Mie theory and assuming an effective radius of 1 μm. A well-known tendency [11], [17], which can also be observed here for the three datasets, is that the position of the maxima of the different coefficients shifts toward higher wavenumbers, as the silica fraction increases. In all three cases, however, this maxima is further to the right for the Pollack CRI compared with the other datasets. For the andesitic ash, the Pollack extinction coefficient also presents a double peak, not seen in the other datasets. The best agreement between the Reed and Deguine datasets is seen for andesitic ash. For the basaltic ash, the shape of the extinction coefficient is almost identical for the Deguine and Reed CRI, but the latter has a much higher intensity. The intensity of the extinction is much closer between the Reed and Pollack CRI for this ash type. For the rhyolitic ash, the indices of Deguine and Reed are, apart from a difference in intensity, again in reasonable agreement. All three sets exhibit a shoulder between 1500 and 1200 cm^{-1} for the rhyolitic ash, but it is most pronounced in the coefficient calculated from the Pollack CRI. The observed differences clearly demonstrate the impact of the refractive index (RI) on the extinction coefficients. In Sections III and IV, we analyze how this affects impact our capability to reproduce satellite observed spectra and satellite retrievals of ash physical properties.

III. Reproducing Observed Spectra

The method that we use here to simulate synthetic spectra and to fit to observed IASI spectra is presented in detail in [9] and [18]. In brief, the forward model combines line-by-line calculations for the trace gas compounds with a doubling-adding routine to account for the scattering and absorption of aerosols. The inverse model is based on the optimal estimation algorithm [19], which, by adjusting iteratively the trace gas and aerosol loadings, minimizes the difference between the observed and simulated spectra (the so-called residual), while taking into account a priori information on the state of the atmosphere and surface from EUMETSAT level 2. In what follows, we set the fitting range to 750–1250 cm^{-1}, corresponding to the entire atmospheric window in the longwave IR, and the range where the largest volcanic ash signatures are observed with IASI. The retrieved parameters are the volcanic ash loading (number concentrations), effective radius (assuming a lognormal distribution with a geometric standard deviation of 2), and the interfering trace gas compounds: carbon dioxide (CO_2), ozone (O_3), sulfur dioxide (SO_2), and water vapor (H_2O). From the number concentration and effective radius, we also calculate the aerosol mass density (g/m^2) and OD. One limitation of the algorithm is that it does

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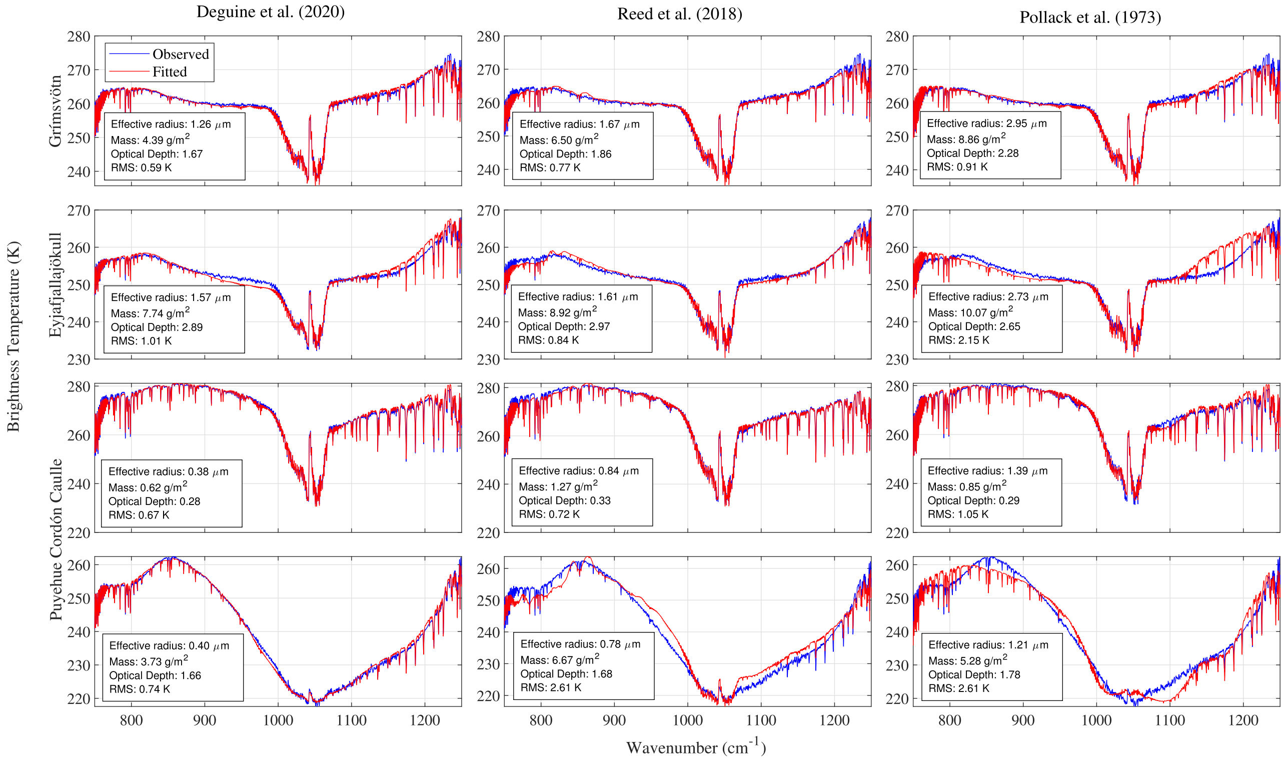

Fig. 2. Retrieval of IASI spectra from the May 23, 2011 Grímsvötn eruption (first row −3-km altitude), May 6, 2010 Eyajfjallajokull eruption (second row −7-km altitude), and June 20, 2011 Puyehue–Cordón Caulle eruption (11-km altitude) using the three different CRI datasets.

not natively support retrieving aerosol altitude. To overcome this, in what follows, we either use external information on the altitude (e.g., from CALIPSO [20], as we do in this section) or repeat the retrieval with different altitude assumptions and keep the one, which results in the lowest residuals (as we do in Section IV). Throughout, we also fixed the surface temperature, based on the ERA5 model reanalysis, as, during initial tests, we found that often unrealistic low surface temperatures were retrieved, which together with lower ash loadings apparently resulted in better residuals.

To compare to what extent the three sets of refractive indices are able to reproduce spectra observed from space, we fit IASI spectra from the eruptions of Grimsvötn, Eyjafjallajökull, and Puyehue–Cordón Caulle. For Puyehue–Cordón Caulle, two spectra were fit, one with an ash OD (AOD) around 0.3 observed over 500 km downwind the volcano and one with an AOD above 1.5 observed near the volcano. Spectra were selected with a large spectral signature, as these pose the greatest challenges and, therefore, stringiest tests on the CRI. The results are presented in Fig. 2. Across the four different spectra, the Pollack CRI performs the worst, as seen from the higher root-mean-square (rms) values of the residuals. The differences with the other datasets are the largest for Eyjafjallajökull, with an rms of 2.2 K more than double that of the two other (0.8 K). It is also apparent from the figures that the fits with the Pollack CRI are especially poor in the 1075–1250 cm^{-1} range, where also the large differences in terms of extinction coefficients were found (see Fig. 1). With the rms values around 0.6–0.8 K, the forward model can largely reproduce the observed spectra of Grimsvötn and Eyjafjallajökull with the Reed and Deguine CRI, even though these rms values are still above the IASI noise of 0.2–0.3 K. A potential reason why the residual of the fit is above the instrumental noise level could be that the RI datasets were retrieved with two different experimental setups not using the same sample where the chemical composition and the size distribution can exhibit small variations. The largest discrepancies are found in the 1200–1250 cm^{-1}, where both sets apparently overestimate the extinction coefficient. The Reed CRI also notably struggles in the 750–825 cm^{-1} range across the four examples. For the Puyehue–Cordón Caulle spectra, the low AOD spectra are fit reasonably well using all the sets, but the large AOD spectrum can only be reconstructed with the Deguine CRI. With an rms of 0.74 K, compared with an ash absorption of 60 K, this is the first time that such a good fit has been reported in the literature for rhyolitic ash (e.g., compare with [19, Fig. 12]).

In terms of retrieved parameters, the largest differences are seen in the effective radius, with the Pollack indices yielding the largest and the Deguine indices yielding the smallest radii. These differences are especially pronounced for the observations of Puyehue–Cordón Caulle. In the next section, we have a closer look at the effect of the CRI on retrieved physical properties for a volcanic plume observed from Eyjafjallajökul.

IV. Case Study on May 7, 2010—Eyjafjallajökull Eruption

The case study presented in this section concerns the large ash plumes observed on May 7, 2010 from the Eyjafjallajökul volcano. To reduce the number of spectra that needed fitting, we ran first the ash detection algorithm from [5] to flag all observations with detectable quantities of ash. Next,

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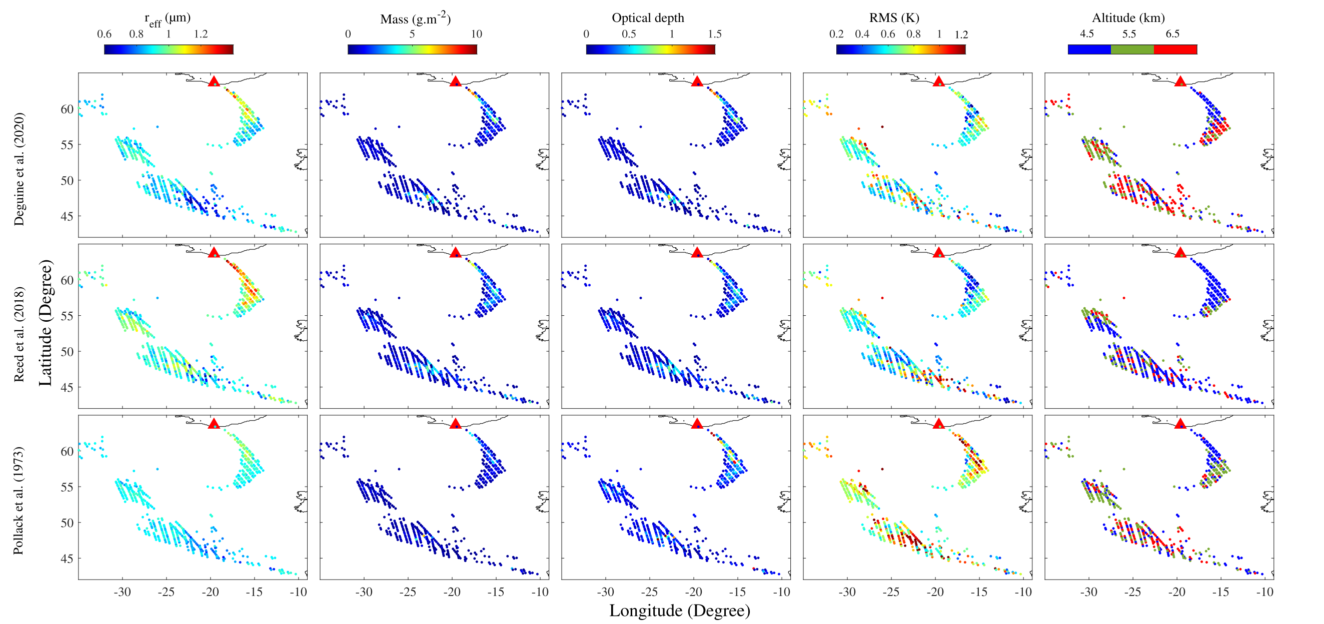

Fig. 3. Retrieval of physical parameters of the entire plume of Eyjafjallajökull on 7th May [post meridiem (PM)] using the different CRI datasets.

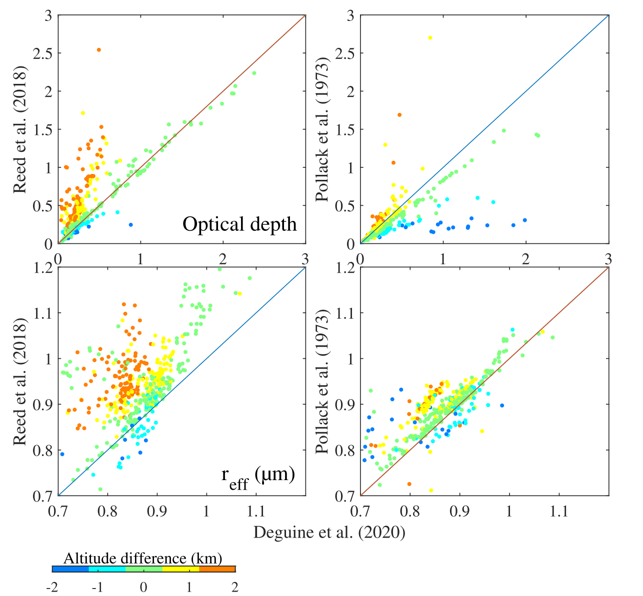

Fig. 4. Further analysis of the Eyjafjallajökull retrievals: correlation plots between the retrieved ODs, the effective radius retrieved using Deguine CRI and (left) Reed CRI and (right) Pollack CRI. The color bar represents the difference in retrieved altitude with respect to the Deguine dataset.

we retrieved mass, effective radius, and OD using the algorithm outlined in Section III. In order to account for variations in altitude, each fit was repeated three times with different altitude assumptions (4.5, 5.5, and 6.5 km), and the results of the fit with the lowest rms were retained. Convergence of the fit and removal of fits with a residual larger than 1.2 K led to 511, 461, and 419 successful retrievals, respectively, with the Deguine, Reed, and Pollack datasets. The results are summarized in Fig. 3, showing the distributions of the different parameters, and in Fig. 4, which shows correlation plots of the different parameters retrieved with the different datasets and as a function of the difference in retrieved altitude.

First, in terms of fit quality, as can be seen from the rms distribution in Fig. 3, the Reed CRI yields the best fits (apart from the 750–850 cm^{-1} range), consistent with what we observed in Fig. 2. The Deguine indices perform only slightly worse with rms values that are, on average, 10% higher. The Pollack indices provide consistently the worst fits, with rms values, on average, 25% larger than the other two sets. This confirms what was observed in the last section and demonstrates again the importance of using CRIs from suspended particles to retrieve physical parameters of volcanic ash plumes, as the CRI retrieved from bulk parameters are largely incompatible with what is observed from satellite.

OD τ characterizes the extent of the spectral extinction in the spectrum. However, in the thermal IR spectral range, this quantity also depends on the (thermal) emission from the ash layer, which, in turn, depends on the temperature and, thus, the altitude of the plume. This explains why the retrieved ODs for the three datasets correlate very well when the retrieved altitude corresponds. Similar observations can be made for the retrieved masses. Following the Mie theory, the ash mass satisfies $M \sim \tau r$. As the retrieved radii are close to one here for most of the plume, the distributions of mass and OD correlate very closely.

The dependence on altitude of both these parameters, irrespective of the chosen CRI, underlines its important role. As seen in Fig. 4, even differences of 1 km can result in biases as large as 50%. For ODs above one, it should be stressed that the retrieval with all three datasets yields an altitude of 4.5 km for the proximal ash plume, consistent with reported measurements from multi-angle imaging spectroradiometer (MISR) [21]. For smaller ODs [3], [8], retrieval of altitude from IR measurements is very challenging, and it is not surprising to observe significant scatter in the diffuse plumes further downwind from the volcano. Note that CALIPSO measurements indicate ash heights between 3 and 9 km for the plume around 50° latitude [22]. It is, therefore, advised

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to use external information on the altitude (in the form of modeled data, lidar observations, and so on) to constrain the retrieval as much as possible, especially when ODs are small.

Finally, as far the effective radius is concerned, we notice somewhat surprisingly that the scatter between the Reed and Deguine retrievals is larger than between the Pollack and Deguine retrievals. This is due to the fact that retrieval with the Reed indices seems to result in wider dynamic range of values, from 0.6 to 1.4 μm as opposed to from 0.7 to 1.2 μm for the other two datasets, and the maximum of the extinction coefficient from Reed (see Fig. 1) is shifted to lower wavenumbers. These values are in the ballpark of what was reported from earlier IASI analysis [7].

V. Conclusion

Hyperspectral IR sounders, such as IASI, are used to track and quantify volcanic ash in the atmosphere. The retrieved quantities, however, depend critically on the refractive indices that are employed in the retrieval. In this letter, we compare, for the first time, the three most important datasets of CRI with respect to the three most common ash types (basaltic, andesitic, and rhyolitic). When it comes to basaltic and andesitic ash, both the Deguine and Reed samples outperform Pollack in terms of able to reconstruct the satellite observed spectra. However, all datasets overestimate the extinction near 1250 cm^{-1}. The Reed indices also fail systematically at reproducing the 750–850 cm^{-1} range.

The Deguine dataset is the only dataset that can accurately fit observed spectra from the (rhyolitic) Puyehue–Cordón Caulle eruption across the entire thermal IR window 750–1250 cm^{-1}; however, it should be acknowledged that the other CRIs were not derived from ash from Puyehue–Cordón Caulle, but from ash with similar chemical composition. While this does not guarantee that the retrieved quantities are closer to the physical reality, being able to reconstruct the observed spectra is a prerequisite of constructing a consistent physical model. For both the rhyolitic and basalatic spectra, the retrieved radii with Deguine are smaller than those of Reed, which, in turn, are much smaller than those obtained with the Pollack indices. These differences could be due to the following: 1) the scattering theory used to retrieve the RI (continuous distribution of ellipsoids (CDE) or Mie) and 2) the variation of chemical composition of the sample analyzed. Finally, the gap in retrieved parameters is less pronounced for the andesitic ash, as confirmed by the case study presented in Section III on the May 7, 2010 plume of the Eyjafjallajökull eruption. For this case study, the (small) differences are found to be mostly related differences in retrieved altitudes.

It is clear from the above that while the availability of CRI based on ash suspended in air is an important milestone, a lot of further research is needed to strengthen the theoretical basis of IR retrievals of volcanic ash. A comprehensive database of reliable in situ measurements of volcanic clouds would, in this perspective, be most welcome.

Acknowledgment

Lieven Clarisse is a Research Associate with the Belgian Fonds de la Recherche Scientifique-Fonds National de la Recherche Scientifique (F.R.S.-FNRS).

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