

Optical Properties of Mineral Dust Aerosol in the Thermal Infrared

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Abstract. The optical properties of mineral dust and biomass burning aerosol in the thermal infrared (TIR) are examined by means of Fourier Transform Infrared Spectrometer (FTIR) measurements and radiative transfer (RT) simulations. The measurements were conducted within the scope of the Saharan Mineral Dust Experiment 2 (SAMUM-2) at Praia (Cape Verde) in January and February 2008. The aerosol radiative effect in the TIR atmospheric window region $800 - 1200 \text{ cm}^{-1}$ ($8 - 12 \text{ }\mu\text{m}$) is discussed in two case studies. The first case study employs a combination of IASI measurements and RT simulations to investigate a lofted optically thin biomass burning layer with emphasis on its potential influence on sea surface temperature (SST) retrieval. The second case study uses ground based measurements to establish the importance of particle shape and refractive index for benchmark RT simulations of dust optical properties in the TIR domain. Our research confirms earlier studies suggesting that spheroidal model particles lead to a significantly improved agreement between RT simulations and measurements compared to spheres. However, room for improvement remains, as the uncertainty originating from the refractive index data for many aerosol constituents prohibits more conclusive results.

INTRODUCTION

Mineral dust and biomass burning aerosols constitute two of the most abundant aerosol types in the Earth atmosphere. Knowledge of their respective optical properties is of paramount importance regarding the related impact on the radiation budget of our planet. Moreover, accurate representation of aerosol radiative properties is a prerequisite for the retrieval of high quality remote sensing products in the presence of aerosols. The SAMUM-2 field experiment [1] was dedicated to investigating the microphysical and optical properties of these two aerosol types. SAMUM-2 took place in January and February 2008 at Praia (Cape Verde). As explained by Haywood *et al.* [2], Saharan mineral dust and biomass burning aerosols from central Africa typically mix over the Cape Verde islands during the northern hemisphere winter months. The SAMUM-2 dataset is perfectly suited for column closure studies, because a broad variety of scientific instruments acquired all essential input quantities for RT simulations. In combination with the additionally conducted reference measurements of the up- and downwelling radiation at top and bottom of the atmosphere (TOA & BOA), the opportunity for a multitude of RT studies arises, as will be demonstrated in the following sections.

CASE STUDIES

This section discusses the TIR optical properties of mineral dust and biomass burning aerosols based on two radiative closure case studies. Case 1 features two optically thin lofted biomass burning layers and demonstrates the sensitivity of satellite based SST measurements to the presence of aerosols. Case 2 investigates the optical properties of a pure mineral dust layer with a focus on particle geometry and mineralogical composition. In both cases measured FTIR spectra are compared to RT simulations in the TIR atmospheric window region ($800 - 1200 \text{ cm}^{-1}$). All simulations were obtained with the PIRATES model [described in 3, chap. 3], which ingested the following input quantities determined experimentally within the scope of SAMUM-2:

- radiosonde profiles of temperature, pressure and relative humidity [see 4]

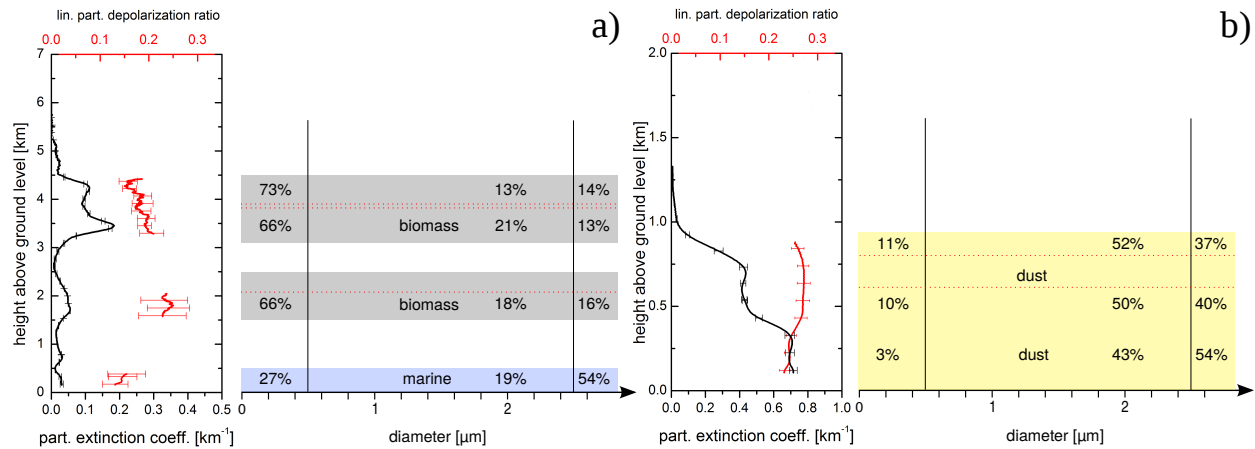


FIGURE 1. Measured profiles of extinction coefficient and depolarization ratio vs. modelled aerosol layers for 6 February (a) and 29 January (b). Percentage values indicate the fraction of total surface area contributed by each of the size bins indicated by the vertical black lines. Red dotted lines symbolise altitudes of airborne particle sampling for size distribution and aerosol composition measurements. Note the different scales for height and extinction coefficient used in (a) and (b). (LIDAR plots courtesy of S. Groß)

- vertical aerosol structure derived from LIDAR data by Tesche *et al.* [5] and Groß *et al.* [6]
- aerosol composition inferred from airborne and ground based in-situ samples analysed in various laboratory experiments by Kandler *et al.* [7] & Lieke *et al.* [8]
- airborne and ground based aerosol size distribution measurements performed by Weinzierl *et al.* [9] & Kandler *et al.* [10]

Note that no measurements of the ozone profile were available for our simulations. Thus, a scaled a priori profile had to be used instead. This may lead to pronounced differences in the ozone band ($1000 - 1080 \text{ cm}^{-1}$) between simulated and measured spectra at TOA. A more detailed analysis of the associated uncertainties can be found in [3, chapter 4.2.2].

Case 1: Thin Lofted Biomass Layer

This case study discusses the influence of an optically thin biomass burning layer on the retrieved SST. For this purpose IASI [11] observations of the upwelling radiance collected on 6 February 2008 are compared to modelled spectra. Measured and simulated vertical aerosol structures for this day are depicted in FIGURE 1 a). Owing to the abundance of small particles and the low particle temperature caused by the elevated layer altitude, this case did not produce observable aerosol emission at BOA. At TOA the situation is different, though, because the aerosol absorbs some of the TIR radiation emitted at the surface. FIGURE 2 demonstrates this effect for a pure sea surface pixel close to the measurement site. Note that the IASI measurement is very well reproduced by the simulation not including any aerosol (left panel), if an SST of 295 K is assumed in agreement with the IASI Level 2 product. Inclusion of aerosols into the simulation weakens the agreement at first glance (FIGURE 2, right), if the SST is kept at 295 K. Increasing the SST to 296 K, however, leads to an almost perfect agreement again, suggesting a cold bias of 1 K in the IASI Level 2 product. This underlines the importance of aerosols for earth observation applications, as detailed knowledge of aerosol optical properties is required in this case to retrieve the SST with an accuracy of better than 1 K. A more detailed discussion of this scenario can be found in [3, chapter 4.4.1].

Case 2: Optical Properties of Mineral Dust

This section examines a low and optically thick mineral dust layer observed on 29 January 2008. No significant impact on the upwelling radiance at TOA could be detected for this scenario, because aerosol absorption is almost entirely compensated for by thermal self-emission due to the small temperature difference between surface and aerosol. Thus, we focus our attention on the downwelling radiance at BOA measured with a ground based FTIR. The interested reader is referred to [3, chapter 2] for an in-depth treatment of the associated experimental activities.

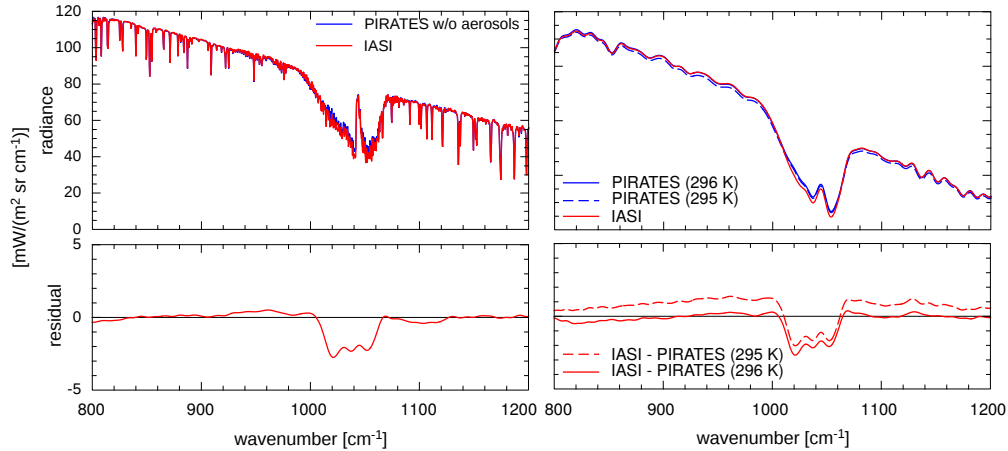


FIGURE 2. Measured (red) and simulated (blue) IASI radiances at TOA (top) and residuals obtained after subtraction of simulation from measurement (bottom) for 6 February 2008. Resolution has been coarsened to 10 cm^{-1} in all but the top left spectra. All aerosols were assumed to consist exclusively of spherical particles. Annotated temperatures indicate the SST assumed in the simulations.

FIGURE 1 b) gives an impression of the observed and modelled aerosol properties. The increased fraction of particles larger than $2.5 \mu\text{m}$ in combination with the relatively high air temperature yields a strong increase in the downwelling radiance at BOA compared to a scenario without aerosols (clear sky scenario). To quantify this downwelling aerosol radiative effect (DRE), the simulated clear sky radiance is subtracted from the measured / simulated radiance with aerosols. The resulting DRE spectra are plotted in FIGURE 3. The left hand panel shows a comparison of measured and simulated DRE spectra for several dust models. All models assume an internal mixture of spherical particles, i.e. they describe the aerosol optical properties in terms of a single effective refractive index. Apart from the frequently encountered OPAC, Shettle and Volz models, FIGURE 3 contains a custom model obtained by mixing the refractive indices of the dust constituents based on the experimentally determined mineralogical composition for this day. Obviously none of the models is capable to satisfactorily reproduce the measured DRE. Deviations are especially pronounced around the main emission peak between 1000 cm^{-1} and 1060 cm^{-1} . It is worth mentioning, that the observed deviations exceed the uncertainty originating from the ozone profile by almost an order of magnitude.

These findings support laboratory studies of Kleiber *et al.* [12] suggesting that spheroidal model particles are superior to spheres in predicting the TIR optical properties of mineral dust aerosols. To assert this statement, we simulated the DRE for oblate spheroidal model particles with an aspect ratio of 1:5. Unfortunately the T-Matrix method used to simulate these particles did not converge for the entire size range, such that particles with size parameter larger than 4 had to be modelled as spheres. However, as shown in [3, chapter 4.4.2], this approximation does not have a significant impact on the results presented here. Additionally the effective medium approach for the refractive index was dropped in favour of an external mixture of pure particles. The resulting DRE spectra are shown as blue curve in FIGURE 3 (right): Obviously they fit the measured DRE much better, although some minor deviations remain beyond 1060 cm^{-1} . These deviations can be traced to contributions by sulfates, quartz, orthoclase and illite. Unfortunately the uncertainties in the refractive indices of most of these materials is quite high [see 3, chapter 4.1.2], such that a more detailed investigation of this effect could not be performed.

Finally, our data yields interesting information regarding an approach reported by Boer [13], who suggests a method to infer the mineral composition based on a least-squares fit. Application of this method leads to the red DRE spectrum in FIGURE 3 (right), which agrees very well with its measured counterpart. A closer analysis of the resulting mineralogical composition reveals significant differences, though, when compared to the in-situ analysis of Kandler *et al.* [7] and Lieke *et al.* [8]: while the fitted mixture consists of 52% illite, 22% kaolinite and 11% montmorillonite in-situ samples always showed significant amounts of quartz, sulfates and orthoclase not identified by the fit. Furthermore, the illite fraction never exceeded 10% in sampled material. Most importantly, the fit leads to a decrease in overall particle number concentration by 15% compared to the in-situ size distributions measured by Weinzierl *et al.* [9]. Thus further research is necessary to judge the applicability of this approach.

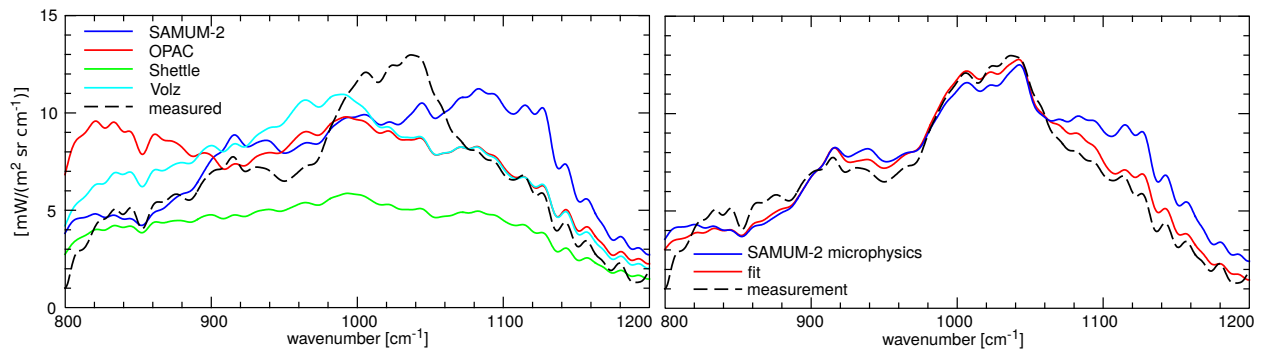


FIGURE 3. Comparison of measured (dashed black) and simulated aerosol DRE for 29 January 2008. *Left:* simulations obtained for an internal mixture of spherical particles with refractive indices of OPAC (red), Shettle (green), Volz (light-blue) and based on SAMUM-2 microphysics (blue). *Right:* DRE spectra for an external mixture of spheroids with aspect ratio 1:5. The mineralogy has been chosen based on SAMUM-2 microphysics (blue) and a least squares fit (red), respectively.

SUMMARY

Two case studies from SAMUM-2 were presented to discuss the TIR optical properties of mineral dust and biomass burning aerosol. Apart from a distinct influence on satellite measurements, we demonstrate strong evidence, that particle asphericity has to be taken into account when modelling the TIR optical properties of optically thick mineral dust layers. According to our research, oblate particles with an aspect ratio of 1:5 result in a good agreement with measured FTIR spectra. Future research on this topic would substantially benefit from more reliable laboratory measurements of TIR refractive indices for several aerosol constituents such as orthoclase and various sulfates.

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