# Quantification of the Aerosol Direct Radiative Effect from Smoke over Clouds using Passive Space-borne Spectrometry

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Abstract. The solar radiative absorption by smoke layers above clouds is quantified, using the unique broad spectral range of the space-borne spectrometer Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) from the ultraviolet (UV) to the shortwave infrared (SWIR). Aerosol radiative effects in the UV are separated from cloud radiative effects in the shortwave infrared (SWIR). In the UV, aerosol absorption from smoke is strong, creating a strong signal in the measured reflectance. In the SWIR, absorbing and scattering effects from smoke are negligible, allowing the retrieval of cloud parameters from the measured spectrum using existing retrieval techniques. The spectral signature of the cloud can be modelled using a radiative transfer model (RTM) and the cloud parameters retrieved in the SWIR. In this way, the aerosol effects can be determined from the measured aerosol-polluted cloud shortwave spectrum and the modelled aerosol-unpolluted cloud shortwave spectrum. This can be used to derive the aerosol direct radiative effect (DRE) over marine clouds, independent of aerosol parameter retrievals, significantly improving the current accuracy of aerosol DRE estimates. Only cloud parameters are needed to model the aerosol-unpolluted cloud reflectance, while the effects of the aerosol absorption are in the aerosol-polluted cloud reflectance measurements.

In this paper we present a case study of the above method using SCIAMACHY data over the South Atlantic Ocean west of Africa on 13 August 2006, when a huge plume of smoke was present over persistent cloud fields. The aerosol DRE over clouds was as high as  $128 \pm 8 \text{ Wm}^{-2}$  for this case, while the aerosol DRE over clouds averaged through August 2006 was found to be  $23 \pm 8 \text{ Wm}^{-2}$  with a mean variation over the region in this month of  $22 \text{ Wm}^{-2}$ .

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## **INTRODUCTION**

Aerosols play an important role in the global energy balance, by absorbing and scattering solar radiation, and by modifying cloud properties and atmospheric column stability. The direct and indirect effects of aerosols are still largely unidentified, while simultaneous measurements of cloud and aerosol properties are difficult, especially from satellite platforms.

The direct effect of smoke aerosols over clouds can be measured using the spectral dependence of smoke absorption in the shortwave spectrum. Smoke absorbs strongly in the UV, but the extinction optical thickness decreases rapidly with increasing wavelength. Therefore, the properties of the clouds below the smoke layer can be retrieved in the SWIR, where smoke extinction optical thickness is sufficiently small. Then, using radiative transfer computations, the contribution of the clouds to the reflected radiation can be modeled for the entire solar spectrum. In this way, cloud and aerosol effects can be separated for a scene with aerosols above clouds. Aerosol microphysical assumptions and retrievals are avoided by modeling only the pure (aerosol-free) cloud spectra. The difference between the spectra are directly linked to the aerosol direct radiative effect (DRE).

## **AEROSOL DIRECT RADIATIVE EFFECT OVER CLOUDS**

A radiative forcing or radiative effect of an atmospheric constituent x can be defined<sup>1</sup> as the difference in the net irradiance  $\Delta E_x$  at a certain level with and without the forcing constituent  $\Delta E_x = E_{\text{with }x}^{\text{net}} - E_{\text{without }x}^{\text{net}}$ , where the net irradiance is defined as the difference between the downwelling and upwelling irradiances  $E^{\text{net}} = E^{\downarrow} - E^{\uparrow}$ . Therefore, at the top of the atmosphere (TOA), where the downwelling irradiance is the incoming solar irradiance  $E_0$ for all scenes, the radiative effect of aerosols overlying a cloud is given by

$$\Delta E_{\rm aer}^{\rm TOA} = E_{\rm cld}^{\uparrow \, \rm TOA} - E_{\rm cld + aer}^{\uparrow \, \rm TOA},\tag{1}$$

where  $E_{\text{cld}}^{\uparrow \text{TOA}}$  is the upwelling irradiance at the TOA for an aerosol-free cloud scene and  $E_{\text{cld+aer}}^{\uparrow \text{TOA}}$  is the upwelling irradiance for an aerosol-polluted cloud scene. By equation (1), if energy is absorbed in the atmosphere by the aerosols, the radiative forcing is positive.

Equation (1) can be evaluated using an RTM, with cloud and aerosol parameters estimated from models or observations. However, an estimation of the flux in the cloud+aerosol atmosphere (second term in equation (1)) can also be made directly from spectral reflectance measurements. In this case equation (1) can be written as<sup>2</sup>:

$$\Delta E_{\text{aer}} \cong \int_{0}^{\infty} \frac{\mu_0 E_0(R_{\text{cld}}(\lambda) - R_{\text{cld+aer}}(\lambda))}{B_{\text{cld}}} d\lambda.$$
<sup>(2)</sup>

 $\mu_0 E_0$  is the TOA solar irradiance incident on a horizontal surface unit,  $B_{cld}$  the anisotropy factor of the scene, relating the reflectance in one direction to the flux in one hemisphere. It is assumed equal for both the aerosol-free cloud scene and the aerosol-polluted cloud scene.  $R(\lambda)$  is the spectral reflectance, which is modelled in the aerosol-free case, and measured for the aerosol-polluted case. See ref. 2 for additional details. Equation (2) is used to derive the aerosol DRE over clouds.

## SMOKE ABSORPTION OVER THE SOUTH ATLANTIC OCEAN

The algorithm is illustrated in Figure 1. The SCIAMACHY reflectance spectrum observed on 13 August 2006, 09:19:43 UTC is shown in red. This is a typical measurement of a scene with smoke from the African continent that was advected over a marine low level cloud layer. The geometry for this scene was  $[\theta, \theta_0, \varphi - \varphi_0] = [18.5^\circ, 43.2^\circ, 131.0^\circ]$ . The TOSOMI total O<sub>3</sub> column  $\Omega$  was 248 DU. The FRESCO cloud pressure and cloud fraction were 902 hPa and 0.6, respectively. The cloud droplet effective radius and cloud optical thickness, retrieved from the SWIR, were 11.2 µm and 14.4, respectively. With these parameters an aerosol-unpolluted cloud reflectance spectrum was modelled, which is indicated by the blue curve. The measured and modelled spectra are close for wavelengths longer than 1246 nm, due to the assumption that the aerosol absorption optical thickness is negligible at these wavelengths. At wavelengths shorter than about 1100 nm, the reflectance spectra start to deviate, which is indicated by the yellow



**FIGURE 1.** SCIAMACHY reflectance spectrum of the scene indicated by the arrow in Figure 4b (red), measured on 13 August 2006, 09:19:43 UTC; and the modelled equivalent unpolluted cloud reflectance spectrum (blue) for this scene. The difference between these two spectra (yellow, labelled 'Aerosol absorption') indicates the irradiance absorbed by the aerosols. The parameters to model the cloud scene were retrieved at various parts of the spectrum (ozone (O3) between 325–335 nm, cloud fraction (CF) and cloud pressure (CP) around 760 nm, cloud phase index (CPI) around 1700 nm, cloud optical thickness (τcld) and effective radius (reff) at 1246 and 1640 nm). The AAI is retrieved from the reflectances at 340 and 380 nm.

area. The difference increases with decreasing wavelength. This is caused by the aerosol absorption optical thickness, which increases with decreasing wavelength. At wavelengths below about 300 nm the differences disappear, because at these wavelengths ozone absorps all radiation. The aerosol absorption in the scene is confirmed by the high value of the AAI of 4.2.

A typical horizontal distribution of the cloud and aerosol fields off the west coast of Namibia is shown in Figure 2a, where SCIAMACHY AAI measurements are overlaid on a MERIS RGB image. Clearly, the horizontal distributions of aerosols and clouds are very variable. Furthermore, they change rapidly from day to day. The corresponding aerosol DRE field over marine clouds is also shown in Figure 2b, for all scenes over the ocean containing water clouds with effective cloud fractions greater than 0.3. It shows the unprecedented details of measured absorbed energy by aerosols over clouds.

Clearly, the aerosol DRE is highly variable with location. The maximum aerosol DRE on this day is 128  $\text{Wm}^{-2}$ , indicated by the arrow. The measured reflectance spectrum for this scene was given in Figure 1. The DRE drops off to zero at the edges of the smoke field, corresponding with the AAI gradient. The minimum DRE was less than 0, which is not an indication of a negative aerosol DRE, but caused by the uncertainty of the measurements. The scale is cut at  $-1 \text{ Wm}^{-2}$ , indicating that the aerosol DRE has vanished and aerosol-unpolluted clouds remain.



**FIGURE 2.** MERIS RGB composite showing the horizontal cloud distribution over the west coast of Africa on 13 August 2006, from 09:13:27 - 09:22:48 UTC, overlaid with a) SCIAMACHY Absorbing Aerosol Index; b) SCIAMACHY Aerosol Direct Radiative Effect [Wm<sup>-2</sup>], retrieved over marine clouds only. This shows the horizontal distribution of smoke over clouds over the Atlantic and the subsequent positive DRE due to the absorption of radiation by the aerosols over the clouds. The aerosol absorption in the selected pixel (shown by the arrow) is analysed in Figure 1.

#### **DISCUSSION AND CONCLUSIONS**

A new method was presented to retrieve the aerosol DRE over clouds using space-based spectrometer measurements and RTM results of cloud scene TOA reflectances. In order to avoid the difficulties in retrieving aerosol parameters from satellite instruments in general, and in cloudy scenes in particular, only cloud parameters are retrieved from the measurements, along with scene parameters that are needed to characterise the cloud reflectance spectrum. The latter include generally available parameters like scattering geometry, total ozone column and surface albedo. Cloud fraction and cloud height can be retrieved using the FRESCO algorithm, relatively unaffected by aerosol contamination. Cloud droplet effective radius  $r_{eff}$  and cloud optical thickness  $\tau_{cld}$  can be retrieved in the SWIR with also well-established algorithms used for MODIS and SEVIRI, among others. However, special care must be taken when retrieving these parameters in aerosol contaminated cloud scenes. Absorption by aerosols in the cloud retrieval bands can bias the retrieved cloud parameters, which is relevant in the current context. Therefore, the cloud parameters are retrieved as far in the SWIR as possible, where the aerosol extinction optical thickness  $\tau_{aer}$  becomes negligible.

The aerosol DRE over clouds from SCIAMACHY was analysed for cloudy scenes over the South-Atlantic Ocean on 13 August 2006. During this day an extensive biomass burning aerosol plume was advected from the African mainland over a permanent marine boundary layer cloud deck. The maximum aerosol DRE found in a single pixel was  $128 \pm 8 \text{ Wm}^{-2}$ . The regionally and monthly averaged aerosol DRE over clouds in August 2006 was  $23 \pm 8 \text{ Wm}^{-2}$  with a variation over the region in this month of 22 Wm<sup>-2</sup> (ref. 2).

SCIAMACHY's 0.25 s pixels have a spatial resolution of approximately  $60 \times 30 \text{ km}^2$ , and global coverage is reached once every six days. Although with this design SCIAMACHY does not have an optimal spatial resolution, the averaged retrieved aerosol DRE over clouds shows unprecedented details. The aerosol DRE over clouds is dependent on cloud cover and aerosol presence, which are both highly variable. SCIAMACHY has been measuring successfully from mid-2002 to 8 April 2012 which can be used to retrieve time series of the aerosol DRE over clouds and relate this to changes in cloud cover and aerosol presence. To retrieve the aerosol DRE at an even higher spatial resolution, the method presented here for SCIAMACHY may be used for other instruments as well. For example, MODIS and OMI, flying in the A-Train constellation, may be used to retrieve cloud parameters in the SWIR (from MODIS), while spectral UV reflectance measurements from OMI can be used to determine the aerosol absorption. The spectral range of OMI covers only the wavelength region up to 500 nm, but the current study shows that this will suffice to capture the bulk of the absorbed energy in the solar spectrum. The reflectance spectrum in the visible and SWIR may be estimated using the few reflectance measurements from MODIS at longer wavelengths. With a precomputed cloud reflectance LUT at OMI/MODIS wavelengths the aerosol DRE in cloud scenes may then be estimated at a superior spatial resolution.

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