Remote sensing of non-aerosol absorption in cloud free atmosphere

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[1] Knowledge of the absorption of sunlight by atmospheric gases, aerosols and clouds is key to understanding climate and its variation. Several studies suggested that clouds absorb sunlight significantly more than what models predict. Other studies suggested that the anomalous absorption is present in cloud-free conditions. Here we measure absorption in cloud free atmosphere using the Aerosol Robotic Network (AERONET). Measurements of attenuation of direct sunlight are used to derive extinction optical thickness and sky measurements to derive scattering optical thickness, Residual extinction for zero scattering, cannot be caused by aerosols or known gases and would be associated with the cloud free absorption anomaly. The anomalous absorption, if exists, is assumed not to correlate with the presence of aerosol. The measurements, taken for several years in locations around the world, show that in the atmospheric windows 0.44, 0.67, 0.87 and 1.02 μm the only significant absorbers in cloud free atmosphere is aerosol and ozone. Non-aerosol absorption, defined as spectrally independent or smoothly variable, was found to have an optical thickness smaller than 0.002 ± 0.003, thereby, absorption of sunlight smaller than 1W/m^2. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 1640 Global Change: Remote sensing; 3360 Meteorology and Atmospheric Dynamics: Remote sensing. **Citation:** Kaufman, Y. J., O. Dubovik, A. Smirnov, and B. N. Holben, Remote sensing of non-aerosol absorption in cloud free atmosphere, *Geophys. Res. Lett.*, *29*(18), 1857, doi:10.1029/2001GL014399, 2002.

1. Introduction

[2] For several decades measurements showed that cloud absorption is larger than radiative transfer models calculated based on our physical understanding. The reasons for this discrepancy were unclear [*Stephens and Tsay*, 1990]. In 1995 new measurements that indicate anomalous cloud absorption were published [*Cess et al.*, 1995; *Ramanathan et al.*, 1995; *Pilewskie and Valero*, 1995]. However other studies did not find any excess atmospheric absorption associated with clouds [*Li et al.*, 1995; *Li and Kou*, 1998] but rather associated with the known absorption of smoke and pollution aerosol in the tropics.

[3] *Kato et al.* [1997], who analyzed cloud free measurements in the ARM site in Oklahoma found that measured surface insolation is significantly smaller than modeled values. Failing to explain the differences, they proposed a possibility of an unknown absorbing gas in the atmosphere. Their finding corroborated with the findings of *Halthore and Schwartz* [2000] and *Arking* [1999] using different observational data sets. *Arking* [1999] suggested that a 0.06 fraction of the absorbed sunlight is not accounted for by models. He associated 25–55% of this discrepancy with water vapor, 15% with clouds, and 30–60% independent of either variable. *Halthore and Schwartz* [2000] showed that there is a discrepancy between direct and diffuse solar radiation that suggests an excess absorption in cloud free conditions of 0.04 ± 0.02 from unknown source. However *Rabbette and Pilewskie* [2001] showed, using spectral measurements, that variability in surface irradiance can be explained by scattering and absorption by known atmospheric constituents, e.g. clouds, water vapor and ozone. Therefore there is no consensus if in cloud free conditions there is an excess atmospheric absorption that is not associated with known trace gases or aerosol.

[4] Here we use a set of measurements designed to measure separately aerosol extinction and scattering in several locations around the world [*Holben et al.*, 1998]. The instruments have a good accuracy both for the extinction optical thickness (OT) measurements [*Eck et al.*, 1999] and for the sky measurements [*Dubovik et al.*, 2000]. The instruments have standardized calibration protocol and cloud screening [*Smirnov et al.*, 2000].

2. Approach and Sensitivity

[5] AERONET measures both the sky brightness and the attenuation of the solar direct flux, in atmospheric windows 0.44, 0.67, 0.86 and 1.02 μm with ozone absorption OT of 0.014 at 0.67 μm. A small water vapor absorption with OT < 0.01 may be present mainly at 1.02 μm, and is addressed in the discussion section. Variation of ozone, water vapor or molecular scattering may introduce variability of OT < 0.01, and bias smaller than 0.002 in the derived absorption OT in individual channels. The sky data are measured in a horizontal plane in the solar zenith angle, the solar almucantar. The AERONET sky measurements are for narrow spectral bands and 1° field of view, thus are not complicated by sensitivity of the measurement instrument to changes spectral or angular properties of the radiation. The inversion code of *Dubovik and King* [2000] is used to derive the aerosol properties via simultaneously fitting the spectral sky radiances measured as a function of the scattering angle from the sun and the spectral transmission of direct solar flux with a proper atmospheric model. The model uses radiative transfer calculations with known molecular scattering. Aerosol particles in the model are assumed to be spherical with a size distribution and wavelength dependent complex refractive index derived in the inversion process. Numerical tests [*Dubovik et al.*, 2000] have shown that the retrieval algorithm reproduces

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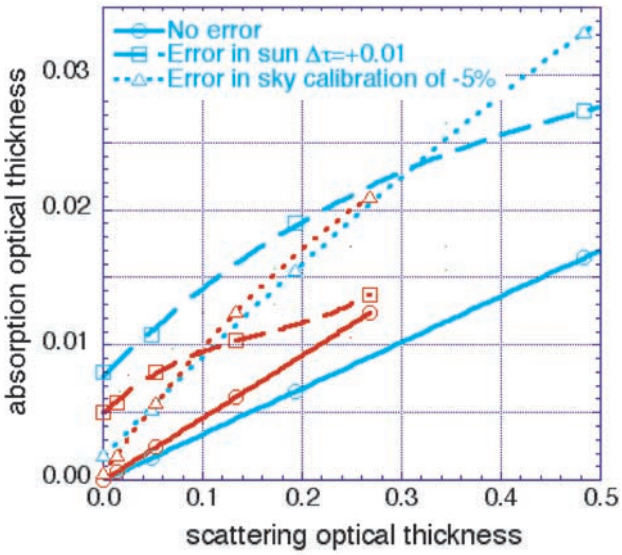

Figure 1. The effect of errors in the sun and sky measurements on the derived absorption optical thickness. The absorption OT is plotted as a function of the scattering optical thickness. Blue lines - 0.44 μm, red - 1.02 μm. Non-aerosol absorption is derived from extrapolation of the results to zero scattering. It is zero in the simulation (solid lines), negligible due to errors in sky radiance and 0.005 to 0.008 for error in the extinction OT of $\Delta\tau_{calib} = \pm 0.01$.

the measured radiance field within 10% of the radiance for specific view direction and 1% for the average radiance field. The aerosol scattering and absorption OT are derived from the screened data, within OT of ±0.01, even in the presence of experimental errors and unaccounted particle non-sphericity. However, at moderate to low aerosol OT ($\tau < 0.4$), limitations in the calibration accuracy ($\Delta\tau_{calib} = \pm 0.01$) significantly affects the retrieved aerosol properties, in particular single scattering albedo, ω_o, defined as: $\omega_o = \tau_{scat}/(\tau_{scat} + \tau_{abs})$), where τ_{scat} and τ_{abs} are the scattering and absorption OTs respectively. In this paper we use inversions of the sun and sky measurements for low optical thicknesses, however we do not derive ω_o, but rather τ_{abs} that has different sensitivity to experimental errors.

[6] To demonstrate the physics of the technique to differentiate between aerosol and non-aerosol absorption we use single scattering approximation in the following description. However sensitivity study of the method and application to data are reported using full radiative transfer. While the sky brightness is a collection of photons scattered by the aerosol and molecules, and therefore is proportional to the aerosol scattering OT, τ_{scat}, the direct sunlight is attenuated both by scattering and absorption, and therefore is directly proportional to the aerosol extinction OT, τ_{ext}. The sky radiance, L_{sky}, is therefore a combination of an aerosol term proportional to τ_{scat} and molecular scattering term:

$$L_{sky}(\Theta) = C\tau_{scat}P_{aerosol}(\Theta) + L_{sky-mol}(\Theta), \quad (1)$$

Where C is a constant, $P_{aerosol}(\Theta)$ is the aerosol scattering phase function in scattering angle Θ. The phase function is determined by the aerosol size distribution and refractive index. $L_{sky\text{-}mol}(\Theta)$ is the molecular sky radiance. The measured transmission of direct sunlight is described by:

$$T_{sun} = V/V_o = \exp[-M(\tau_{ext} + \tau_{mol})] \quad (2)$$

where T_{sun} is the transmission of solar radiation obtained from the measured signal V and the calibration V_o obtained from Langley plots. T_{sun} defines the aerosol OT, τ_{ext}. The airmass M is $M = 1/\cos(\theta_o)$, θ_o is the solar zenith angle, τ_{ext} is the extinction OT and τ_{mol} is the molecular OT. The combination of AERONET sky and sun measurements is sensitive to both scattering and extinction. The absorption OT, τ_{abs} is derived from the difference between the extinction (eq. 2) and scattering (eq. 1) optical thicknesses. For individual observations we cannot distinguish between aerosol and non-aerosol absorption. However in a scatter plot between τ_{abs} and τ_{scat} for many observations, the slope is proportional to the aerosol absorption, or $(1\text{-}\omega_o)/\omega_o$ where ω_o is the true aerosol single scattering albedo and the intercept is the remaining absorption in the absence of aerosol or the non-aerosol absorption.

[7] Simulation of the method in the presence of calibration errors is shown in Figure 1. Aerosol absorption is plotted as a function of the aerosol scattering for 3–4 values of the optical thickness. The aerosol properties are kept constant in this simulation. Error in the calibration was introduced separately for sun measurements (dashed lines) and sky measurements (dotted line). Variations in the aerosol single scattering albedo, in the presence of calibration errors is discussed later with the help of Figure 3. The value of τ_{abs} for $\tau_{scat} = 0$ are calculated by a linear extrapolation of the two lowest values. For the selected sites the errors in the measurements are of $\Delta\tau_{calib} = \pm 0.01$ [*Holben et al.*, 1998; *Eck et al.*, 1999] for airmass M = 1. This calibration error in the extinction OT causes an error of $\Delta\tau_{abs}$ of 0.005 to 0.008 for the plotted spectral channels (see Figure 1). Error in the sky calibration generated a negligible error in the non-aerosol absorption, though it affects the derived aerosol absorption. Note also that the calibration error in τ_{ext} introduced a non-linearity in the dependence of τ_{abs} on τ_{scat}. This non-linearity can be used as an indicator of calibration errors. Therefore, in the presence of anomalous absorption and calibration errors the retrieved absorption OT is given by:

$$\tau_{abs}^{retr}(aeros.) \approx \tau_{abs}^{real}(aeros.) + \tau_{abs}(anomalous) \pm \Delta\tau_{calibr.} \quad (3)$$

[8] The calibration is performed by comparing every 6–12 months the instrument measurements to "master" instruments calibrated routinely in Mauna Loa, Hawaii, [*Holben et al.*, 1998]. These calibrations are assumed to be independent and therefore the error for an assembly of points across several years of measurements is smaller by the square root of the number of independent calibrations:

$$\tau_{abs}^{retr}(aeros.) \xrightarrow[\tau_{scat}^{retr}(\text{aeros.}) \to 0]{} \tau_{abs}(anom.) \pm \frac{\Delta\tau_{calibr.}}{\sqrt{N_{meas}}} \quad (4)$$

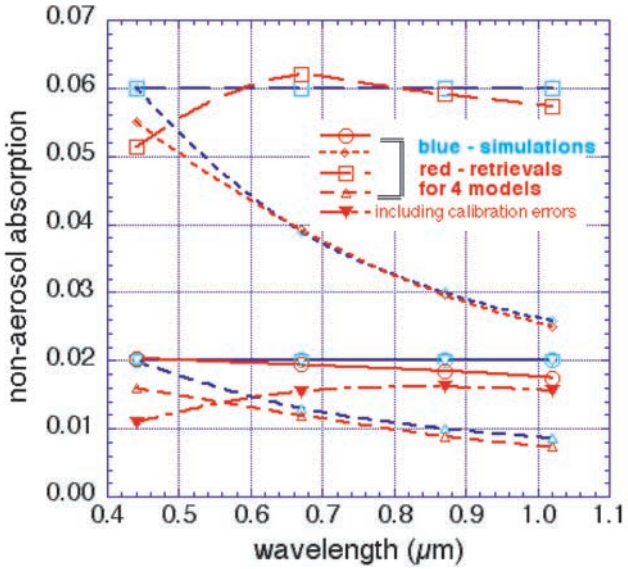

Figure 2. Simulation of the inversion process in the presence of non-aerosol absorption. The simulated non-aerosol absorption is given for 5 cases by the blue symbols and lines, and the retrieved values by the red symbols and lines. Fixed absorption OT τ_{abs} of 0.02 (open circles) and 0.06 (open squares) is simulated as well as spectrally decreasing τ_{abs} with spectral dependence of $1/\lambda$ (small symbols). For comparison the effect of calibration error of $\Delta\tau_{calib} = 0.01$ is also shown.

The results presented in section 3 are for airmass of M = 2 decreasing the calibration error by factor 2. Five oceanic sites and 2 land sites are used with 1–4 years of measurements, decreasing the error further by factor of 1.5–2.5. Therefore the predicted error in the maritime and land non-aerosol absorption is $\Delta\tau_{abs} = 0.002–0.003$ for individual sites and $\Delta\tau_{abs} = 0.001–0.002$ for the combined land and ocean data set presented in this paper. Bias in water vapor and ozone absorption for all the sites and period of time, introduces a bias of $\Delta\tau_{abs} \leq 0.002$, for a total error of $\Delta\tau_{abs} = \pm 0.003$. Uncertainties in the aerosol nonsphericity and surface albedo were shown to have a negligible effect on the aerosol absorption derivation *Dubovik et al.* [2000]. The derived absorption is effectively the missing energy between attenuation of direct sunlight and the integrated sky radiance, not sensitive to the shape of the scattering phase function, and thus to nonsphericity. The error in the sky calibration is ±5% and affects the accuracy of the scattering OT but not the retrieved non-aerosol absorption.

[9] The inversion code associates any unaccountable absorption, e.g. anomalous or water vapor absorption, in individual measurements to aerosol. Only the scatter plots between absorption and scattering OT derive the anomalous absorption that is not attributed to aerosol. In Figure 2 we test if the present procedure can detect anomalous cloud free absorption in case it does exists. To a given aerosol model we add both large and small absorption, spectrally constant or gradually changing with wavelength, but independent of the aerosol optical thickness. The results, in Figure 2, show that in the blue channel, with the strong molecular scattering, the method detects the non-aerosol absorption within an error of 0.01. In the other wavelengths the errors are smaller, down to 0.004 in the longer wavelengths. The detection is good both for spectrally neutral or linearly varying absorption. We also tested the response of the model to the presence of the water vapor absorption of $\tau_{abs} = 0.01$ in only one channel–1.02 μm (not shown in the figure). Since aerosol cannot produce such spectrally strong gradient in absorption, the inversion distributed the absorption among the channels. We conclude that the AERONET data and the presently suggested analysis should be able to detect anomalous absorption of $\tau_{abs} \geq 0.01$.

3. Results

[10] The first application of the method is to a data set of 3 years of measurements in the Cart site in Oklahoma.

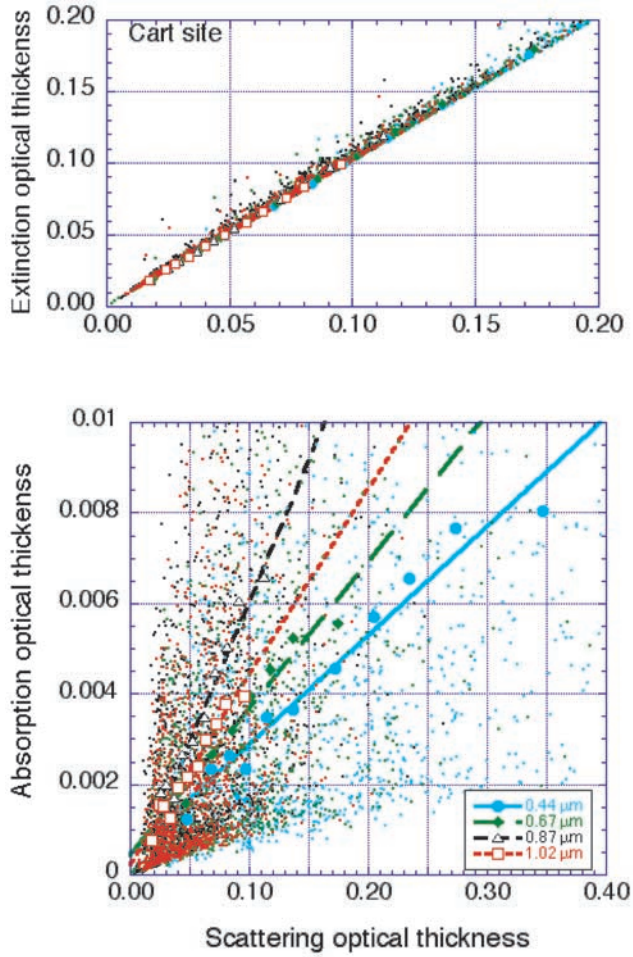

Figure 3. Scatter plot of the extinction (top) and absorption (bottom) OT as a function of the scattering OT for 3 years of measurements in the Cart site in Oklahoma. Individual measurements are shown by dots and averages for a given range of τ_{scat} by circles for the 4 wavelengths indicated in the figure. The averages show a close to linear dependence with very small intercepts, or non-aerosol absorption of $\tau_{abs} = (-1 \text{ to } 5) \pm 10^{-4}$ well under the measurements error of 0.002.

Table 1. Summary of the Offset in the Scatter Plots of τ_{abs} vs. τ_{scat}

Location	0.44 μm	0.67 μm	0.87 μm	1.02 μm
Lanai	0.0000	−0.002	−0.0001	−0.0002
Tahiti	0.0039	0.0029	0.0025	0.0025
Nauru	0.0008	0.0005	0.0003	0.0004
Ascension Island	0.0023	0.0037	0.0047	0.0054
Bermuda	0.00006	−00010	0.0004	−0.0001
All maritime	0.0017 ± 0.0017	0.0017 ± 0.0019	0.0018 ± 0.0022	0.0020 ± 0.0025
Cart Site	0.0005	0.0004	−0.0001	0.0002
Turkey	−0.0002	0.0011	0.0011	0.0014

For each case the data were first averaged for several ranges of τ_{scat} and then fitted with a linear fit. The uncertainty in the method is estimated to be $\Delta\tau_{abs} = \pm 0.002$.

Figure 3 shows a scatter plot of the extinction and absorption OT derived from the data as a function of the scattering optical thickness. Both individual measurements and averages for a progressing range of τ_{scat} are shown. The individual points show large variations in the aerosol absorption for the same τ_{scat} due to variability in the aerosol properties and random error in $\tau_{abs.}$ Due to small noise in the extinction OT. However, as evident by the smoothly varying averages τ_{abs} for give range of τ_{scat}, there is no significant effect on the intercept of non-aerosol absorption. The individual values of τ_{ext} and averages of τ_{ext} and τ_{abs} show a close to linear dependence on τ_{scat} with very small intercepts, corresponding to non-aerosol absorption of $\tau_{abs} = (-1 \text{ to } 5) \times 10^{-4}$ well under the measurements error. The linearity of the average data shows that the aerosol properties do not change systematically with the aerosol loading, and that the calibration is proper. The systematic low intercept shows that in the Cart site there is no absorption independent of the presence of aerosol in the four atmospheric windows. This values of τ_{abs}, for the average air mass of 2 corresponds to <0.001 fraction of sunlight being absorbed, in comparison to 0.04 [*Halthore et al.*, 1998] and broadband 0.06 [*Arking*, 1999] for cloud free conditions. The method was also applied to 5 oceanic sites in the Atlantic and Pacific Oceans and one additional land site. The results are summarized in Table 1. The results are similar for the oceanic and land sites with offset, or non-aerosol absorption $\tau_{scat} \leq 0.002$, which is the theoretical estimate for accuracy of this method.

4. Discussion and Conclusions

[11] Several years of spectral measurements from several AERONET locations of sky angular distribution and attenuation of direct solar flux were used to study non aerosol absorption. Non aerosol absorption is defined as atmospheric absorption not associated with a known gas, e.g., ozone or water vapor and not correlated with the presence of aerosol. If organic gases emitted in the process of biomass burning or gases emitted from pollution sources together with the aerosol have an unknown absorption, it would not be reflected in the non-aerosol absorption described here but rather in a reduced aerosol single scattering albedo. The non-aerosol absorption is determined as the extrapolation of the measured absorption optical thickness as a function of the aerosol scattering optical thickness, τ_{scat}, to $\tau_{scat} = 0$. Non-aerosol absorption in the atmospheric windows 0.44, 0.67, 0.87 and 1.02 μm was found to have an optical thickness with an upper bound of 0.002 and uncertainty of ±0.003.

[12] It is therefore concluded that there is no unknown absorption of sunlight in the atmospheric windows covered by AERONET.

[13] **Acknowledgments.** We would like to acknowledge the use of the data of the AERONET and SIMBIOS sites. Data from the Cart site were collected by R. N. Halthore and Mark Miller of Brookhaven National Laboratory as part of the ARM program.

References

Arking, A., The influence of clouds and water vapor on atmospheric absorption, *Geoph. Res. Lett.*, *26*, 2729–2732, 1999.

Cess, R. D., and M. H. Zhang et al., Absorption of Solar-Radiation By Clouds - Observations Versus Models, *Science*, *267*, 496–499, 1995.

Dubovik, O., and M. D. King, A flexible inversion algorithm for retrieval of aerosol optical properties from sun and sky radiance measurements, *J. Geophys. Res.*, *105*, 20,673–20,696, 2000.

Dubovik, O., A. Smirnov, and B. N. Holben et al., Accuracy assessments of aerosol optical properties retrieved from AERONET sun and sky radiance measurements, *J. Geophys. Res.*, *105*, 9791–9806, 2000.

Dubovik, O., B. N. Holben, and T. F. Eck et al., Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atmos. Sci.*, *59*, 590–608, 2002.

Eck, T. F., B. N. Holben, and J. S. Reid et al., Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, *J. Geophys. Res.*, *104*, 31,333–31,349, 1999.

Halthore, R. N., and S. E. Schwartz, Comparison of model estimated and measured diffuse downward irradiance at surface in cloud free sky, *J. Geophys. Res.*, *105*, 20,165–20,177, 2000.

Holben, B. N., T. F. Eck, and I. Slutsker et al., AERONET – A federated instrument network and data archive for aerosol characterization, *Rem. Sens. Environ.*, *66*, 1–16, 1998.

Kato, S., T. P. Ackerman, and E. E. Clothiaux et al., Uncertainties in modeled and measured clear–sky surface shortwave irradiances, *J. Geophys. Res.*, *102*, 25,881–25,898, 1997.

Li, Z. Q., H. W. Baker, and L. Moreau, The variable effect of clouds on atmospheric absorption of solar–radiation, *Nature*, *376*, 486–490, 1995.

Li, Z., and L. Kou, Atmospheric direct radiative forcing by smoke aerosols determined from satellite and surface measurements, *Tellus (B)*, *50*, 543–554, 1998.

Pilewskie, P., and F. P. J. Valero, Direct observations of excess solar absorption by clouds, *Science*, *267*, 1626–1629, 1995.

Rabbette, M., and P. Pilewskie, Multivariate analysis of solar spectral irradiance measurements, *J. Geoph. Res.*, *106*, 9685–9696, 2001.

Ramanathan, V., and X. Subasilar et al., Warm pool heat–budget and shortwave cloud forcing – a missing physics, *Science*, *267*, 499–503, 1995.

Smirnov, A., B. N. Holben, and T. F. Eck et al., Cloud screening and quality control algorithms for the AERONET data base, *Rem. Sens. Env.*, *73*, 337–349, 2000.

Stephens, G. L., and S. C. Tsay, On the cloud absorption anomaly, *Quart J. ROYAL Meteor. Soc.*, *116*, 671–704, 1990.

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