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Dual-wavelength aerosol vertical profile measurements by MAX-DOAS at Tsukuba, Japan

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Abstract

We present vertical profiles of the aerosol extinction coefficient retrieved from ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements at Tsukuba, Japan (36.1° N, 140.1° E), from November 2006 to March 2007. Retrievals utilizing absorption by the oxygen collision complex O_4 are first made at two wavelengths, 354 and 476 nm. A robust assessment of the MAX-DOAS aerosol data is made using coincident lidar measurements throughout the period. Agreement between aerosol extinction coefficients measured by MAX-DOAS and the lidar tends to be better at the longer wavelength and at lower altitudes. At 476 nm, the best agreement, to within 30%, is found at altitudes of 0–1 km, confirming results from a literature assessment for a two-month measurement period. These findings are supported by comparisons of the aerosol optical depth by MAX-DOAS and sky radiometer measurements and are further explained by differences in the O_4 absorption and the air mass factor profile between 354 and 476 nm. Thus, uncertainty in MAX-DOAS aerosol measurements is well quantified and characterized, providing a basis for quantitative studies using MAX-DOAS measurements.

1 Introduction

Atmospheric aerosol particles are recognized as an important contributor to climate change but the magnitude of their radiative forcing as well as its sign is highly uncertain (IPCC, 2007). To measure the aerosol vertical profile in the lower troposphere, a new remote sensing technique using Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) has been proposed recently (Wagner et al., 2004; Frieß et al., 2006; Irie et al., 2008a, b). MAX-DOAS has several technical advantages suitable for the purpose of atmospheric monitoring, including simple setup, low power consumption, and fully-automated operation without absolute radiometric calibration. Another notable advantage of MAX-DOAS is the capability for simultaneous measurements of

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aerosols and gases, including nitrogen dioxide (NO_2), formaldehyde (HCHO), glyoxal (CHOCHO), and sulphur dioxide (SO_2) (e.g. Hönninger et al., 2004; Wittrock et al., 2004; Heckel et al., 2005; Hendrick et al., 2006; Leigh et al., 2006; Sinreich et al., 2007; Inomata et al., 2008; Irie et al., 2008b), which may play direct or indirect roles in determining chemical, physical, and optical properties of aerosols. Thus, it is expected that MAX-DOAS measurements could contribute not only to the monitoring of atmospheric composition changes but also to the understanding of the Earth system. The quantitative assessment of MAX-DOAS aerosol measurements, however, has been very limited.

The present study is a follow-on to that of Irie et al. (2008a), who first reported ground-based MAX-DOAS measurements of the vertical profile of the aerosol extinction coefficient at a single wavelength 476 nm (k_{476}) at Tsukuba, Japan (36.1° N, 140.1° E, 29 m a.s.l.), from 1 November to 21 December 2006. They concluded that the overall uncertainties of MAX-DOAS k_{476} values at 0–1 and 1–2 km are better than 30% and 60%, respectively, based on comparisons with other established aerosol measurements by lidar and sky radiometer. Here we analyze measurements for a time period of more than double the duration of the initial study, from 1 November 2006 until 16 March 2007, to confirm the uncertainty estimated by Irie et al. (2008a). To characterize MAX-DOAS aerosol measurements in a more robust sense, we present retrievals of the aerosol extinction coefficient at an additional, shorter wavelength, 354 nm (k_{354}), and discuss them together with the retrievals at 476 nm.

2 MAX-DOAS measurements

We operated the MAX-DOAS system at Tsukuba, Japan, from 1 November 2006 through 16 March 2007 with the same instrumentation as described by Irie et al. (2008a). A miniaturized UV/visible spectrometer (B&W TEK Inc., BTC111) with a charge-coupled device (CCD) was used to record spectra between 280 and 560 nm. The measurements were made at six different elevation angles (ELs) of 3°, 5°, 10°, 20°, 30°, and 90°

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using a movable mirror, which repeated the same sequence of ELs every 30 min. The wavelength calibration using a high-resolution solar spectrum (Kurucz et al., 1984) indicated that the spectral resolution (Full Width at Half Maximum (FWHM)) was 0.40 and 0.55 nm at wavelengths around 354 and 476 nm, respectively, throughout the period of the measurements.

As a next step, DOAS spectral fitting (Platt, 1994) using the nonlinear least-squares method (Irie et al., 2008a) was performed to retrieve the differential slant column density (ΔSCD), which is defined as the difference between the slant column density along the path of sunlight for an off-axis measurement (EL<90°) and that for a zenith-sky measurement (EL=90°). Two different fitting windows, 325–367 and 460–490 nm, were analyzed for aerosol retrievals at 354 and 476 nm, respectively. The wavelengths of 354 and 476 nm correspond to the O_4-cross-section-weighted mean wavelengths for the respective fitting window (Fig. 1). Fitting parameters and qualities for the ΔSCD retrievals are summarized in Table 1. Compared to 325–367 nm, the fitting window 460–490 nm yields smaller O_4ΔSCD errors (Table 1), likely due to the stronger O_4 absorption at 460–490 nm (Fig. 1). Although in the spectra recorded with our instrument the intensity is usually stronger at 460–490 nm than at 325–367 nm, smaller errors were found at the longer wavelengths even when a similar statistical comparison was made under the same intensity conditions.

Subsequently to the DOAS analysis, an aerosol retrieval algorithm employing the optimal estimation method (Rodgers, 2000; Irie et al., 2008a) was utilized to invert the O_4ΔSCD values to retrieve the vertical profiles of the aerosol extinction coefficient, k, for every 30-min interval. The mean k for each 1-km layer is analyzed below and denoted k(0–1 km) for the layer 0–1 km, for example. This inversion method refers to the differential box air mass factor (ΔA_{box}) lookup tables (LUTs) created by a radiative transfer model, the Monte Carlo Atmospheric Radiative Transfer Simulator (MCARaTS) (Iwabuchi, 2006). We prepared different LUTs for each wavelength of 354 and 476 nm. The calculations by MCARaTS have been validated through comparisons with other radiative transfer models (Wagner et al., 2007). ΔA_{box} is the difference between the

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box air mass factor (A_{box}) value for off-axis measurements and that for the reference, where the A_{box} is defined as the ratio of partial slant to vertical columns for the given layer. Thus, since the profile of ΔA_{box} (and A_{box}) contains information about the path length over which the measured sunlight traversed for each layer, it is used below as a proxy for the MAX-DOAS measurement sensitivity to aerosols at different altitudes.

It should be noted that for both inversions at 354 and 476 nm the a priori state vector and the a priori covariance matrix used are identical to those constructed by Irie et al. (2008a) based on averages and standard deviations of two months of lidar data at 476 nm. Since k values are usually greater at shorter wavelengths, this provides an opportunity to investigate a potential dependence of the retrievals on the a priori information and the sensitivity of the MAX-DOAS measurements.

3 Lidar and sky radiometer measurements

From 1 November 2006 to 16 March 2007, lidar and sky radiometer measurements were performed together with MAX-DOAS at the same site in Tsukuba, Japan. The lidar system operated was a compact Mie-scattering system utilizing the second harmonics of a flashlamp-pumped Nd:YAG laser (532 nm) as the light source (Shimizu et al., 2004). Inversion was conducted assuming a constant lidar ratio of 50 sr throughout the period. Only cloud-free lidar aerosol extinction data, judged by the method of Shimizu et al. (2004), were used. The lidar aerosol extinction data were converted to k_{354} and k_{476} values, which can be compared to MAX-DOAS data, using coincident measurements of the Ångström exponent (α) by the sky radiometer. Note that the used α is a columnar quantity, as described below, and may lead to a systematic bias in lidar k_{354} and k_{476} values, when the used α differs from α for a given altitude.

The sky radiometer measured the direct solar irradiance and the distribution of radiances in the aureole region (Aoki and Fujiyoshi, 2003). The aerosol optical depth (τ) data at 400, 500, 675, 870, and 1020 nm were derived by the inversion algorithm (SKYRAD.pack, version 4.2) developed by Nakajima et al. (1996). The α value was

estimated from the τ values at five wavelengths. Using α, τ values at 354 and 476 nm were derived from the measured τ values at 400 and 500 nm, respectively. It should be noted that τ at 354 nm was estimated using α, which may be valid between 400 and 1020 nm. Details of the lidar and sky radiometer measurements used in the present study are available elsewhere (Aoki and Fujiyoshi, 2003; Shimizu et al., 2004; Irie et al., 2008a).

4 Results and discussion

In Fig. 2, the MAX-DOAS k_{476} and k_{354} values are plotted against those derived from lidar measurements for layers of 0–1 and 1–2 km over the whole period of more than 4 months. As in Fig. 2a, the MAX-DOAS and lidar k_{476}(0–1 km) values agree to within 30% for most cases (Fig. 2a), confirming the results of Irie et al. (2008a) obtained from an only-two-month comparison. Similarly, the MAX-DOAS k_{476}(1–2 km) values generally agree with the lidar to within 60% (Fig. 2b).

Mean MAX-DOAS values for each bin of the lidar data are also plotted with red symbols in Fig. 2. It can be seen from the mean MAX-DOAS data that the correlations of k_{476}(1–2 km) values tend to deviate from linearity at higher lidar values (Fig. 2b). On the other hand, the individual comparisons for k_{354}(0–1 km) values show more scatter than for k_{476}(0–1 km), but the differences are usually less than 50% (Fig. 2c). Although MAX-DOAS and lidar k_{354}(0–1 km) values are correlated linearly, a linear regression analysis suggests that MAX-DOAS values are systematically smaller (slope = 0.78). The same tendency is also seen in the k_{476}(0–1 km) comparisons (Fig. 2a), suggesting that the differences can be attributed to the use of a constant lidar ratio, which is a common systematic component of the uncertainty in both lidar k values. Further systematic differences can occur only in the k_{354}(0–1 km) comparisons, due to the Ångström exponent α, which was used to derive lidar k_{354}(0–1 km) values, although it may be valid only at wavelengths longer than 400 nm, as mentioned above. Indeed, the α value derived from the sky radiometer is slightly different from that derived directly

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from the two wavelengths of MAX-DOAS k(0–1 km) values, as discussed later.

As seen in Fig. 2d, the correlations of k_{354}(1–2 km) show that MAX-DOAS k_{354}(1–2 km) at higher lidar values tends to be underestimated, similar to the result for the same layer at 476 nm (Fig. 2b). In addition, more significant underestimation of MAX-DOAS values occurs at 2–3 km, especially as seen in the individual MAX-DOAS/lidar comparisons shown with gray symbols in Fig. 3. These tendencies are further supported by the fact that a similar underestimation is seen also in the MAX-DOAS/sky radiometer comparisons of the aerosol optical depth (τ) (Fig. 4), which contains information on high-altitude aerosols.

Thus, MAX-DOAS k data at higher lidar values tend to be underestimated more at higher altitudes. In addition, uncertainty in MAX-DOAS k data is likely larger at shorter wavelengths, especially considering the comparison results for k_{476}(0–1 km) and k_{354}(0–1 km) shown in Figs. 2a and 2b, respectively.

To investigate the cause of these tendencies, we plot the median k_{354} and k_{476} profiles retrieved from MAX-DOAS measurements for the whole period of the measurements analyzed (Fig. 5a). As we expect, k_{354} values are generally greater than k_{476} values, especially at altitudes below 1 km, although we have used the same a priori information for both wavelengths. This indicates that the aerosol retrieval method employed here is less dependent on the a priori, especially below 1 km. Indeed, a sensitivity test by Irie et al. (2008a) revealed that the error in the retrieved k_{476}(0–1 km) due to the choice of a priori conditions was estimated to be less than 5%.

For both 354 and 476 nm, the median profiles of the maximum ΔA_{box} at different ELs for each retrieval are shown in Fig. 5b. The maximum ΔA_{box} values weighted by the partial vertical column of O_4 for a given 1-km layer, [O_4], are also shown in Fig. 5c. Both quantities giving a measure of the measurement sensitivity decrease with altitude (Figs. 5b and 5c). Moreover, they are smaller at 354 nm than at 476 nm. Thus, altitude and wavelength dependencies in the measurement sensitivity can explain the observed tendencies of the differences between MAX-DOAS and lidar data. The observed tendencies have been explained also with a simulation by Frieß et al. (2006). They in-

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vestigated four absorption bands of O_4 peaked around 360, 477, 577, and 630 nm and argued that the highest sensitivity to aerosols should occur at 577 and 630 nm, followed by 477 and 360 nm. This should be a result of the combination of two effects: (1) higher transparency of the atmosphere leads to better geometric enhancements (i.e. greater ΔA_{box}) at longer wavelengths and (2) the largest cross section occurs at 577 nm, followed by 630, 477, and 360 nm. In addition, the sensitivity for aerosols near the surface is expected to be greatest, owing to the longer light paths through the lowermost atmospheric layers, consistent with the ΔA_{box} profiles shown in Fig. 5b.

It is therefore reasonable to expect that better aerosol retrieval performance can be achieved using absorption bands around 577 or 630 nm compared to those around 477 and 360 nm, which have been used in the present work. Note that this is only the case when analyzing a single absorption band, while Frieß et al. (2006) have proposed to analyze multi-wavelength absorptions together for better performance, compared to the use of a single wavelength.

In the present study we have analyzed O_4 absorption bands at 354 and 476 nm individually, not combined as a single inversion problem. However, it is worth investigating the internal consistency between MAX-DOAS k_{354}(0–1 km) and k_{476}(0–1 km) values, both of which have shown better agreement with lidar data than at other altitudes (Fig. 2). Figure 6 shows correlations between the MAX-DOAS k_{354}(0–1 km) and k_{476}(0–1 km) values. Also shown are the correlations between aerosol optical depths at 354 and 476 nm derived from sky radiometer measurements. As expected, values at 354 nm are systematically greater than at 476 nm for both MAX-DOAS and sky radiometer data. The slopes of the linear regression are 1.28±0.03 and 1.40±0.01 for MAX-DOAS and sky radiometer measurements, respectively. The corresponding α values were estimated to be 0.83±0.08 and 1.13±0.03. Although more works are needed to interpret the difference, it is interesting to note that the MAX-DOAS α value has been derived from k(0–1 km) values, supporting that MAX-DOAS is capable of deriving α vertical profile information.

We found that the correlation coefficient (R^2) for MAX-DOAS k(0–1 km) data is as

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high as 0.83 (Fig. 6), although retrievals have been done individually for 354 and 476 nm. In particular, the scatter of the correlations is much smaller than that anticipated from the 50%-range of k_{354}(0–1 km) and the 30%-range of k_{476}(0–1 km), which are shown with error bars in Fig. 6. These results confirm that the uncertainty in the MAX-DOAS aerosol data has been well quantified using comparisons with lidar data in this study and can be regarded as upper-limit estimates. While MAX-DOAS measurements are thus expected to provide aerosol datasets useful for quantitative studies, an improvement of this technique, for example, by utilizing the O_4 absorption at longer wavelengths, is highly desirable.

5 Conclusions

To establish a quantitative basis for MAX-DOAS aerosol measurements, aerosol extinction coefficient vertical profiles were retrieved at two wavelengths, 354 nm (k_{354}) and 476 nm (k_{476}), from spectra measured by MAX-DOAS at Tsukuba, Japan. A robust comparison of MAX-DOAS aerosol extinction profiles with those measured by lidar has been done for more than 4 months between 1 November 2006 and 16 March 2007. The retrieved k_{476} values for the layer at 0–1 km (1–2 km) generally agreed with the lidar data to within 30% (60%), confirming the results of Irie et al. (2008a) that the uncertainty is less than 30% (60%) based on comparisons using only two months of data. On the other hand, agreement for k_{354} values at 0–1 km was within 50%. Thus, we found that uncertainty in MAX-DOAS aerosol extinction data is smaller at a longer wavelength and at lower altitudes (in the case that the instrument is located near the surface). MAX-DOAS k_{354} and k_{476} values tend to be underestimated at upper layers. This is supported by the fact that a similar underestimation was seen in the comparison between aerosol optical depths obtained from MAX-DOAS and sky radiometer measurements. These dependencies of the systematic differences on wavelength and altitude were, however, well characterized by the stronger O_4 absorption and the greater differential box air mass factor (ΔA_{box}) at the longer wavelength. ΔA_{box} also

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showed greater values at lower altitudes. Considering that ΔA_{box} gives a measure of the measurement sensitivity, the observed underestimation likely occurred at altitudes to which the analyzed O_4 absorption had insufficient sensitivity. Thus, uncertainty in MAX-DOAS aerosol data was quantified, and the cause leading to systematic errors was identified, showing that MAX-DOAS measurements are suitable for quantitative studies.

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Table 1. DOAS fitting parameters and quality.

Fitting window (nm)	460–490	325–367
Degree of polynomial	3	3
Degree of offset polynomial	2	2
Absorber	O_4, NO_2, O_3 H_2O, Ring	O_4, NO_2, O_3 HCHO, BrO, Ring
Typical residual[a]	0.0007	0.0011
Typical error in O_4ΔSCD[a] (molecules^2 cm^{-5})	10×10^{40}	19×10^{40}

[a]Medians for SZA=50°±5°.

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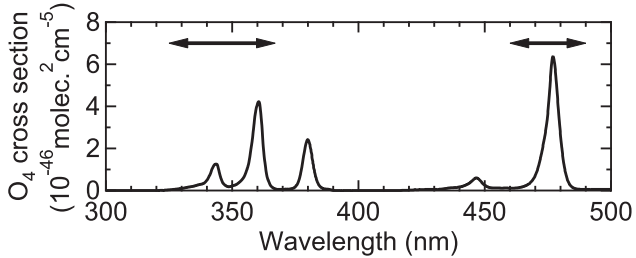

Fig. 1. Absorption cross section of O_4. Two windows, at 325–367 and 460–490 nm, indicated by arrows, are analyzed for aerosol retrieval at 354 and 476 nm, respectively.

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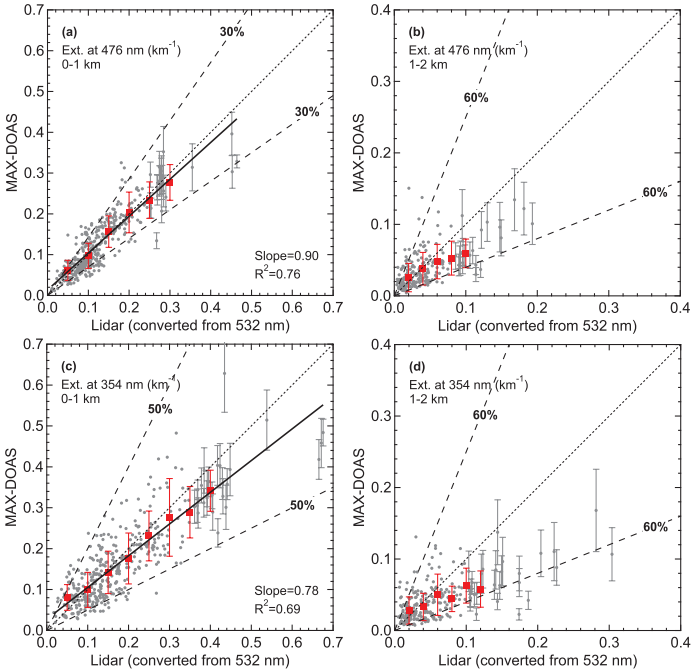

Fig. 2. Correlations between MAX-DOAS and lidar aerosol extinction coefficients at 476 nm for layers of **(a)** 0–1 and **(b)** 1–2 km. The same correlations but for 354 nm are shown in **(c)** and **(d)**. Red symbols indicate the averages of the original MAX-DOAS values (gray) for each bin of the lidar data. Error bars represent 1σ standard deviations. For the original MAX-DOAS data, uncertainty estimated from the retrieval covariance matrix is shown only for large lidar values for clarity. Solid lines in (a) and (c) represent the linear least-squares fit to the original data before binning.

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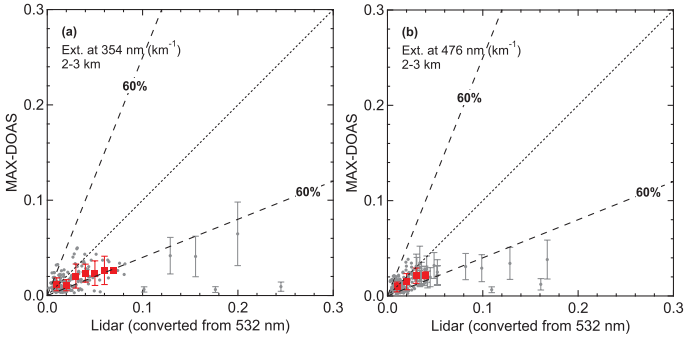

Fig. 3. Same as Fig. 2, but for the aerosol extinction coefficient at **(a)** 354 and **(b)** 476 nm for the layer at 2–3 km.

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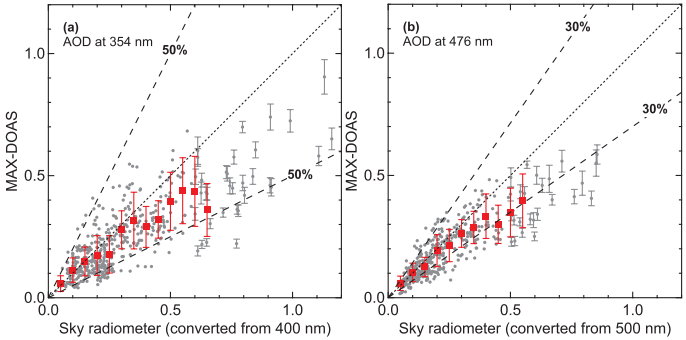

Fig. 4. Same as Fig. 2, but for the aerosol optical depth at **(a)** 354 and **(b)** 476 nm.

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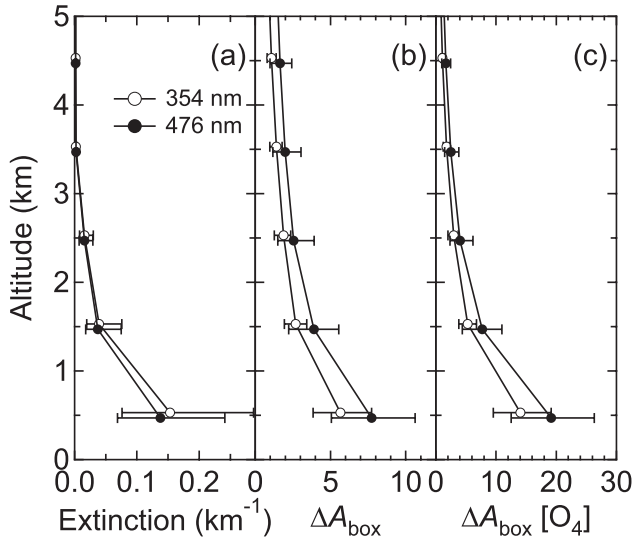

Fig. 5. Median profiles of **(a)** the aerosol extinction coefficient, **(b)** the differential box air mass factor (ΔA_{box}), and **(c)** $\Delta A_{box} \times [O_4]$ (10^{42} molecules^2 cm^{-5}) for 354 nm (open symbols) and 476 nm (solid symbols) from 1 November 2006 to 16 March 2007 at Tsukuba, Japan. [O_4] is the partial vertical column of O_4 for a given 1-km layer. Error bars represent the central 67% of the ranges.

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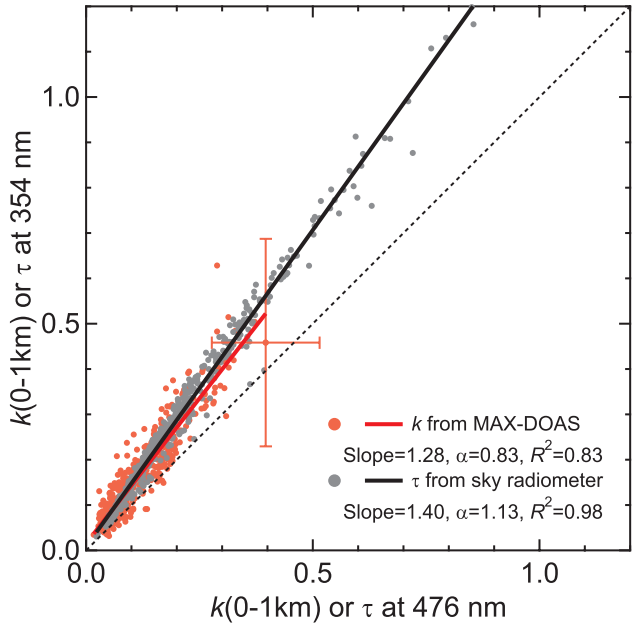

Fig. 6. Correlations between MAX-DOAS aerosol extinction coefficients (0–1 km) at 354 and 476 nm (red symbols). Error bars represent 50%- and 30%-uncertainties estimated from comparisons with the lidar data for 354 and 476 nm, respectively. Correlations between sky radiometer aerosol optical depths at 354 and 476 nm (gray symbols) are superimposed. The linear least-square fits to MAX-DOAS and sky radiometer data are shown by black and red lines, respectively. The slope of the line and the Ångström exponent (α) calculated from the slope are given.

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