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Supplement of

Long-term MAX-DOAS network observations of NO_2 in Russia and Asia (MADRAS) during the period 2007–2012: instrumentation, elucidation of climatology, and comparisons with OMI satellite observations and global model simulations

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Overview and Features of AOD

The methodology used to derive AOD values at 476 nm is described in the main text, and the results are briefly summarized in this supplementary material. A color index (defined as the ratio of the intensities at 500 and 380 nm) was used to screen out cloudy cases. The threshold values used for the color index are listed in Table S1. The threshold value for Cape Hedo was changed from 1.50 (Takashima et al., 2009) to 2.40 (this study) with equivalence, because the offsets at the two wavelengths were newly taken into account for the revised calculation of the color index. For other locations, we tentatively determined the equivalent color index threshold values based on the assumption that the color ratio under conditions where the sky was the most whitish was similar to that at Cape Hedo; this provided calibration information for the relative responses of the individual instruments at 380 and 500 nm.

Figure S1 shows the time series of monthly means of the AODs at six sites, with and without cloud screening, based on the MAX-DOAS color index. The site-to-site differences were not very large in comparison with those in the case of NO_2; in particular, the AOD levels for Yokosuka (0.24 ± 0.05 (1σ) as averages of monthly mean values after cloud screening, see Table S2), an urban site, were similar to those at Cape Hedo (0.33 ± 0.13 (1σ)), a remote island. Hefei had the highest average value (0.59 ± 0.13 (1σ)) among the studied locations. The levels were roughly comparable to the climatological AOD values derived from satellite sensors, i.e., Moderate Resolution Imaging Spectroradiometer (MODIS)/Terra, MODIS/Aqua, and Multi-angle Imaging Spectroradiometer (MISR)/Terra (Fig. S1 and Table S2). The used monthly average satellite data are from MODIS/Terra (Collection 5) and MODIS/Aqua (Collection 5.1) at a 1° × 1° grid resolution, and MISR/Terra (ver. 31) at a 0.5° × 0.5° grid resolution, available from the NASA Goddard Earth Sciences Data and Information Services Center (http://daac.gsfc.nasa.gov/giovanni/). For the MODIS sensors, the AOD values at 550 nm were converted to those at 476 nm using the reported Ångström parameters. For MISR, the Ångström parameters were estimated from the reported AOD values at multiple wavelengths (443, 555, 670, and 865 nm), and then the AOD values at 476 nm were estimated. Although the MODIS/Terra and MISR/Terra observations were made in the morning, and the MODIS/Aqua observations were made in the afternoon, they were all compared with the daytime averages of the MAX-DOAS observations. The AODs derived from MAX-DOAS did not show significant diurnal variations (data not shown).

Similar seasonal variation patterns at remote islands (Cape Hedo and Fukue) were found for the MAX-DOAS and satellite observations, with higher values in winter–spring as a result of long-range transport from the Asian continent along the westerlies. The agreement was excellent for Zvenigorod. Common increases in August 2010 were attributable to intense forest/peat fires. At Yokosuka, the average MAX-DOAS AOD level with cloud screening was more consistent with MISR than with MODIS (especially in summer), partly because of better spatial resolution. Similar tendencies were found for Gwangju and Hefei. For Hefei, the month-to-month variation patterns were qualitatively similar for MAX-DOAS and satellite data. Comparisons with satellite data with finer spatial resolutions will be studied in the future.

Figure S2 shows hourly-averaged AODs derived from MAX-DOAS and sky radiometer (Aoki and Fujiyoshi, 2003) data at Fukue for 2009. The AOD value at 476 nm for sky radiometer was calculated from the reported AOD value at 500 nm and the Ångstöm exponent. A strong positive correlation was found around the 1:1 line. Similar comparisons with sky radiometers and Mie lidar observations were successful at Tsukuba (Irie et al., 2008) and at Cape Hedo (Takashima et al., 2009). Based on these features, we conclude that our AOD products, with an estimated 30% uncertainty, are basically consistent with available observations. As mentioned in the main text, 30% uncertainty in the AOD introduced only 10% or less uncertainty in the TropoNO2VCD. Therefore the aerosol information can be satisfactorily used to estimate NO_2 optimally and to study the general trends in the OMI(NASA)/MAX-DOAS ratio of TropoNO2VCD against the AOD.

Table S1. Recommended threshold values for color index used for cloud screening.

Instrument	Threshold value
Cape Hedo #1	2.40
Yokosuka #1	1.67
Fukue #1	0.81
Fukue #2	2.40
Fukue #3	1.55
Gwangju #1	2.20
Gwangju #2	2.10
Gwangju #3	2.02
Hefei #1	1.74
Hefei #2	2.01
Zvenigorod #1	1.57

Table S2. Averages and 1σ ranges of monthly mean AOD values derived from MAX-DOAS observations and satellite observations.

	MAX-DOAS (with cloud screening)	MAX-DOAS (without cloud screening)	MODIS/Terra	MODIS/Aqua	MISR/Terra
Cape Hedo	0.33 ± 0.13	0.40 ± 0.14	0.26 ± 0.13	0.25 ± 0.14	0.26 ± 0.12
Yokosuka	0.24 ± 0.05	0.30 ± 0.06	0.37 ± 0.16	0.42 ± 0.18	0.27 ± 0.17
Fukue	0.29 ± 0.08	0.42 ± 0.12	0.38 ± 0.14	0.34 ± 0.12	0.37 ± 0.17
Gwangju	0.40 ± 0.10	0.50 ± 0.11	0.47 ± 0.23	0.53 ± 0.23	0.33 ± 0.18
Hefei	0.59 ± 0.13	0.73 ± 0.15	0.80 ± 0.28	0.86 ± 0.28	0.60 ± 0.23
Zvenigorod	0.21 ± 0.05	0.29 ± 0.10	0.19 ± 0.19	0.21 ± 0.22	0.18 ± 0.11

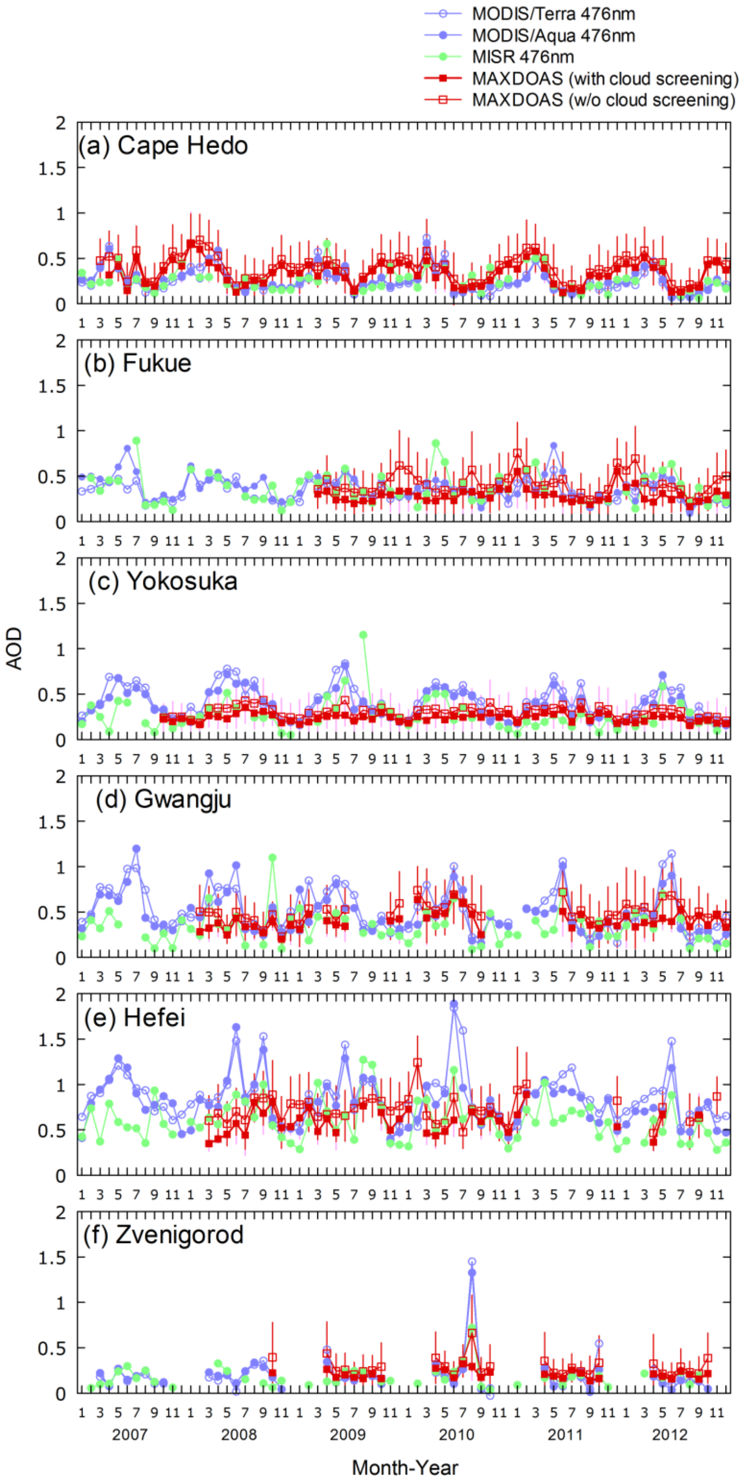

Fig. S1. Time series of monthly averages of AOD derived from MAX-DOAS and satellite observations. The satellite observations are derived using MODIS and MISR sensors on board Terra and Aqua. MAX-DOAS data, with and without cloud screening, are provided with error bars representing the 1σ range of the included data.

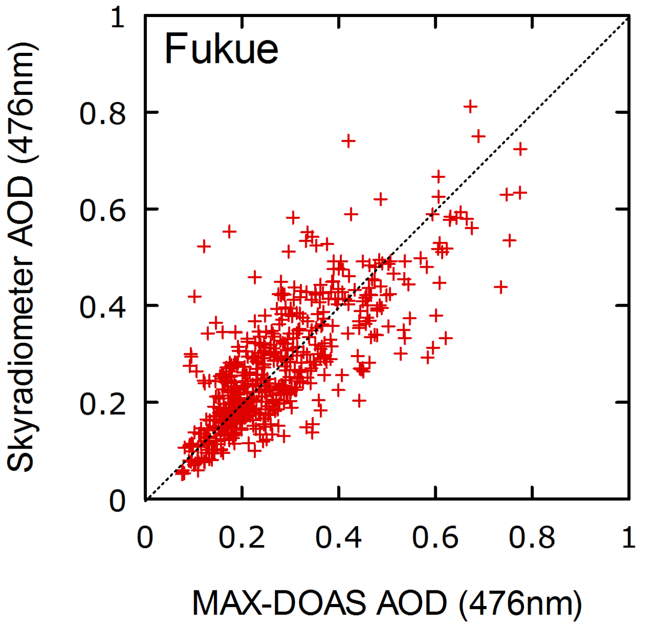

Fig. S2. Scatterplot between AODs observed by MAX-DOAS and a sky radiometer at Fukue site in 2009.

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