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# A new method of measuring aerosol optical properties from digital twilight photographs

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## Abstract

An optimal-estimation algorithm for inferring aerosol optical properties from digital twilight photographs is proposed. The sensitivity of atmospheric components and surface characteristics to brightness and color of twilight sky is investigated, and the results

- <sup>5</sup> suggest that tropospheric and stratospheric aerosol optical thickness (AOT) are sensitive to condition of the twilight sky. The coarse–fine particle volume ratio is moderately sensitive to the sky condition near the horizon under a clean-atmosphere condition. A radiative transfer model that takes into account a spherical-shell atmosphere, refraction, and multiple scattering is used as a forward model. Error analysis shows that the
- tropospheric and stratospheric AOT can be retrieved without significant bias. Comparisons with results from other ground-based instruments exhibit reasonable agreement on AOT. A case study suggests that the AOT retrieval method can be applied to atmospheric conditions with varying aerosol vertical profiles and vertically inhomogeneous species in the troposphere.

#### 15 **1** Introduction

Twilight sky, one of the most beautiful sights in our daily life, varies from day to day. The color of the sky under clear-sky conditions gradates from orange-red and salmon red near the horizon to blue-gray and blue at zenith. This gradation results from Rayleigh scattering caused by molecules (Minnaert, 1993). Twilight sky has been studied for
<sup>20</sup> more than a half century, elucidating the effects of ozone and stratospheric aerosol on twilight sky colors. As an example, ozone has the Chappuis band, an absorption band with a peak wavelength of about 550 to 600 nm. Hulburt (1953) simulated twilight-sky brightness at zenith with a radiative transfer model (RTM) based on the Rayleigh scattering theory and taking into account ozone absorption. Hulburt's results showed that
<sup>25</sup> the contribution of ozone absorption is more important than that of Rayleigh scattering in providing the blue color at zenith. Lee et al. (2011) measured twilight sky in Antarc-





tica with a digital still camera to investigate ozone effects on twilight sky at zenith and the anti-solar horizon, with the results suggesting that scattering light by aerosols is as important as ozone absorption in explaining the color of twilight. These results are consistent with simulation results by Adams et al. (1974).

In 1963, Mt. Agung in Bali, Indonesia, erupted and injected a massive amount of volcanic ash into the stratosphere. After the eruption, stratospheric aerosol effects on twilight sky have been investigated by many researchers. Volz (1964, 1965) suggested that a persistent purple light in the twilight term seen during 1963 was caused by an increase of stratospheric aerosol as a result of the eruption, using twilight sky measurements and geometrical calculations to support this. Dave and Mateer (1968) demonstrated that stratospheric aerosol was more important than ozone as a mechanism for creating purple light, showing this by simulations with RTM taking into account

a spherical-shell atmosphere, dust, and ozone effects.

The general radiative transfer process for radiance at a large viewing zenith angle (VZA) in the twilight sky is more complicated. Because of this, the effects on twilight sky color near the horizon of atmospheric components have not been investigated. To properly do so, an RTM must incorporate a spherical-shell atmosphere, multiple scattering, and refraction effects. Shaw (1981) investigated the dependence of stratospheric aerosol vertical profiles on twilight sky at VZA of 70° by using a single-scattering analyt-

- ical RTM with a spherical-shell atmosphere. Recently, an RTM for calculating spectral radiation according to a complicated radiative transfer process has been developed (Iwabuchi and Suzuki, 2009); this model achieves realistic simulation of twilight sky near the horizon. Saito et al. (2013) showed that tropospheric aerosol optical thickness (AOT) was sensitive to twilight sky near the horizon by analyzing photographic data
- <sup>25</sup> and RTM simulations. Those results suggest that multiply scattered light is dominant in twilight sky color and brightness near the horizon.

Some researchers have tried to retrieve aerosol information from twilight sky data. Wu and Lu (1988) developed a method to infer stratospheric aerosol vertical profiles from twilight sky brightness and degree of polarization measurements at a wavelength





of 700 nm at zenith. Mateshvili et al. (2013) developed a method that uses optimal estimation to infer the upper tropospheric and stratospheric aerosol vertical profiles from spectral at zenith during twilight, providing stratospheric AOT both pre- and posteruption of the volcano Nabro. The aerosol information obtained from these twilight measurements was consistent with that from other measurements, showing that twilight sky measurement is a useful tool for gathering information on aerosols.

It is well known that tropospheric and stratospheric aerosol contributes to climate change. The optical and radiative properties of aerosol can vary strongly according to relative amount, chemical components, and particle sizes. This results in high uncer-

- tainty of climate change prediction in terms of radiation budget (IPCC, 2013). Aerosol measurement is carried out with satellite- and ground-based instruments such as the moderate-resolution imaging spectroradiometer (MODIS) devices on-board the Terra and Aqua satellites and the aerosol robotic network (AERONET) system. This provide on solar and sky measurements (Remer et al., 2005; Holben et al., 1998). How-
- ever, most of the instruments useful for aerosol measurement require passive remotesensing techniques, meaning that these instruments cannot infer aerosol properties in regions without direct sunlight. In addition, satellite-based instruments with passive sensors cannot infer aerosol properties in deserts and snow-covered regions because of high surface reflectivity. Particularly in the polar region during winter, aerosol mea-
- <sup>20</sup> surements and their data are very limited because of the length of the polar night (sunless condition in all day). In the region, a few measurements are carried out by active remote sensing techniques, in situ sampling, and difficult aerosol measurements that use direct light from the moon and stars as a substitute for direct sunlight transmittance methods (Heber et al., 2002; Berkoff et al., 2011).
- Recently, digital cameras have begun to be used as measurement instruments (e.g., Lee and Andres, 2003; Lee et al., 2011). Ehrlich et al. (2011) provided a method for constructing a bi-directional reflectance distribution function for cloud from data obtained by a digital single-lens reflex (DSLR) camera installed on an airplane. Kataoka et al. (2013) developed a method for estimating the altitude of visible aurorae from





data obtained by a pair of DSLR cameras. Hioki and Iwabuchi (2014) demonstrated a method to find the particle radius of pollen and its column number density from a solar corona image taken with a digital still camera. A two-dimensional brightness distribution can be obtained by digital camera, an ability that is likely to be useful in <sup>5</sup> many scientific fields. The aim of the present study is to infer aerosol optical properties, such as tropospheric and stratospheric AOT and particle size information, from twilight photographs taken with a DSLR camera. This paper is organized into six sections. Section 2 presents the photographic measurement and camera characteristics. Section 3 shows the forward model and the tests to determine the sensitivity of aerosol optical properties to twilight sky. In Sect. 4, the method of aerosol retrieval and the error analysis are explained. Section 5 discusses the retrieval results and presents some case studies. The results are compared with those inferred from other ground-and satellite-based instruments. Section 6 summarizes the study and discusses a few

points of interest from the retrieval results.

#### 15 2 Measurements from a digital camera

The instrument for photographic observations in twilight was a Nikon D7000 DSLR camera equipped with a Nikkor equisolid angle-type full-frame fisheye lens (AF DX Fisheye-Nikkor, 10.5 mm f/2.8G ED). The camera configuration simulated an International Organization for Standardization (ISO) film speed of 200 and had an aperture of

- f2.8. These values were set low enough to reduce complementary metal oxide semiconductor (CMOS) noise and to gain high exposure, even in the dark conditions of twilight. The camera was fixed on a tripod. Table 1 summarizes the measurement setup and the camera characteristics. Photographic observations of twilight sky have been carried out at the Graduate School of Science, Tohoku University (38.26° N, 140.84° E;
- 25 208 m elevation), Sendai, Japan, since January 2013. Various instruments for aerosol measurements are collocated at the site. These include a Mie scattering lidar system operated by the National Institute for Environmental Studies, a skyradiometer operated





by SKYNET and a sunphotometer (PREDE PGS-100) originally managed by Tohoku University (Shimizu et al., 2010; Takamura and Nakajima, 2004). The twilight observation criteria are (1) the absence of clouds at the zenith during the twilight, measured by the Mie scattering lidar, and (2) the absence of clouds over a 200 km area on the so-

- Iar side of the observation site, measured by infrared imagery from the Multifunctional Transport Satellite (MTSAT), a geostationary meteorological satellite. Table 2 describes the VZAs, solar zenith angles (SZAs), and relative azimuth angles (RAAs). The RAAs are the differential angle between the viewing azimuth and the solar azimuth for data used in aerosol retrievals derived from photographic observations. The measurements
- are carried out for an SZA ranging from 90 to 96° in intervals of 1°. The camera exposure time is changed according to SZA so as to be suitable for imaging. To obtain the color and brightness distribution of a twilight sky from a RAW image file, we first extract the digital numbers for the three spectral channels (RGB) in groups of 12 × 6 pixels at corresponding angles (see Table 2 for details). After that, the averaged digital counts
- <sup>15</sup> for each channel are used to eliminate pixel-level CMOS noise. The RAW format data provides a wider dynamic range than the 8 bit JPG format does, which is sufficient to provide brightness information. To read the Nikon D7000 manufacturer-specific RAW format (for the Nikon D7000 the exact format is NEF), we employed the useful open source tool DCRAW. DCRAW has been described in Ehrlich et al. (2011). Figure 1 shows twilight photographs taken under typical atmospheric conditions.

The color matching functions (CMFs) of the camera for each RGB channels were determined at the National Institute of Polar Research (NIPR) to quantify the spectral radiation from the digital numbers. The details of the method for determining the CMF for a specific camera are given in Saito and Iwabuchi (2015). This paper briefly describes

<sup>25</sup> the methods. The experiment setup is described in detail in Sigarnes et al. (2008). The camera and spectroradiometer detect narrowband (*i*) light in visible wavelengths of 380–740 nm at intervals of 5 nm, which are separated by a grating monochromator, obtaining camera RAW counts  $u'_{k,i}$  and the corresponding spectral radiance  $I_i(\lambda)$ . The camera spectral channels and wavelengths are defined as *k* and  $\lambda$ , respectively. To





enable comparison between the values, the values of  $u'_{k,i}$  are normalized by the photon flux density convert radiance-proportional RAW counts  $u_{k,i}$ , and  $u_{k,i}$ , which can be defined as

$$u_{k,i} = \int_{0}^{\infty} S_k(\lambda) I_i(\lambda) \mathrm{d}\lambda,$$

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- <sup>5</sup> where the function  $S_k(\lambda)$  is the relative CMF. The optimal estimation-based inverse calculation is applied to Eq. (1), yielding determining  $S_k(\lambda)$  with some associated uncertainty. In this experiment, the same camera configurations are used for all photographic observations, and an exposure time of 8 s is used. The CMFs for the Nikon D200, as determined by Sigarnes et al. (2008), are used as prior information. The vignetting char-
- acteristics of the fisheye lens have been investigated by Saito and Iwabuchi (2015), and we eliminate the effects of vignetting from the camera RAW counts. Figure 2 shows the relative CMFs of the Nikon D7000. The center wavelength of each channel (i.e., the most sensitive wavelength) is shifted to a shorter wavelength, by about 10 nm for R channel and 20 nm for the B channel, relative to those of the Nikon D200. Moreover, the shape of the spectral consitivity on the C shapped of the D7000 is breader in shape

the shape of the spectral sensitivity on the G channel of the D7000 is broader in shape than that of the D200.

For photographic observation in twilight, the RAW counts  $u'_k$  are converted to radiance-proportional RAW counts  $u_k$ , which can be rewritten as  $cI_k$ , where  $I_k$  is channel-averaged spectral radiance, weighted by the camera spectral response of the channel k, and c is a parameter determined by the quantum efficiency of the CMOS and the ISO film speed. Finally, the chromaticity (R/G, B/G) is obtained by dividing

- each of  $cI_R$  and  $cI_B$  by  $cI_G$ , and the normalized brightness  $G_n$  is obtained by dividing the value of  $cI_G$  by the value of  $cI_G$  measured at VZA of 70° and RAA of 0° for all VZAs, SZAs, and RAAs in the twilight sky. This process results in an 840-dimensional mea-
- <sup>25</sup> surement for each photographic observation. For prior knowledge, this method requires only the relative CMFs of the camera; absolute calibration is unnecessary because the



(1)



unknown parameter c can be excluded from the data analysis. Absolute calibration can be used to estimate the parameter c, but this is beyond the scope of the current paper and is left for future work.

### 3 The forward model

## **5 3.1 Model setup for twilight simulations**

For twilight sky simulations, we use JACOSPAR, a spherical-shell atmosphere RTM that accounts for refraction and multiple scattering (Iwabuchi and Suzuki, 2009). The RTM calculates singly scattered light by a semi-analytical approach and multiply scattered light by a backward Monte Carlo (BMC) method. The RTM also considers 300 wavelength bands in the range from 370 to 740 nm, and models gaseous absorption by 10 the correlated k-distribution method used in the RSTAR RTM (Nakajima and Tanaka, 1986; Sekiguchi and Nakajima, 2008). The earth's surface is assumed to be a Lambertian surface with an albedo of 0.1. In the model atmosphere, we assume the presence of 70 atmospheric layers, from ground to an altitude of 120 km, with geometrically thin layers, particularly in the lower part of the atmosphere. For simulating their scattering 15 properties, aerosols are assumed to be spherical particles and the effects are calculated according to the Lorenz–Mie theory (Bohren and Huffman, 2008). The normalized vertical profile of stratospheric aerosol extinction coefficient  $b_{st}(z)$  at a wavelength of 550 nm is assumed to follow a Gamma distribution b(z), defined as

$$_{20} \quad b(z)=z^{\xi-1}\frac{e^{z/\psi}}{\Gamma(\xi)\psi^{\xi}},$$

where  $\Gamma(\xi)$  denotes the Gamma function, and  $\xi$  and  $\psi$  are parameters that depend on the shape and scale, respectively. The normalized vertical profiles of the tropospheric aerosol extinction coefficient  $b_{tr}(z)$  are assumed to be the sum of the vertical profiles of background aerosol  $b_{bg}(z)$ , aerosol in boundary layer  $b_{bl}(z)$ , and upper tropospheric



(2)



aerosol  $b_{up}(z)$ , each of which follows a Gamma distribution. The shape and scale parameters for each aerosol vertical profile are summarized in Table 3. By substitution, the tropospheric aerosol vertical profile  $b_{tr}(z)$  can be written as

 $b_{\rm tr}(z) = a_{\rm bg}b_{\rm bg}(z) + a_{\rm bl}b_{\rm bl}(z) + a_{\rm up}b_{\rm up}(z),$ 

<sup>5</sup> where  $a_{bg}$ ,  $a_{bl}$ , and  $a_{up}$  are, respectively, the AOT fraction of background aerosol, boundary layer aerosol, and upper tropospheric aerosol relative to total tropospheric AOT at a wavelength of 550 nm. In this study, the AOT fractions are assumed to be in the proportions  $a_{bg}: a_{bl}: a_{up} = 1: 1.2: 0.4$ . In general, the extinction coefficient in the middle-upper troposphere shrinks exponentially with increasing altitude, and aerosol is abundant in the boundary layer. The extinction coefficients  $\beta(z)$  can be calculated by multiplying the AOTs by b(z).

The distribution of aerosol particle volumes dV/dr is assumed to follow a lognormal distribution:

$$\frac{\mathrm{d}V}{\mathrm{d}r} = \frac{V_{0,\mathrm{F}}}{r\sqrt{2\pi}\ln s_{\mathrm{F}}} \exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_{\mathrm{mod},\mathrm{F}}}{\ln s_{\mathrm{F}}}\right)^{2}\right] + \frac{V_{0,\mathrm{C}}}{r\sqrt{2\pi}\ln s_{\mathrm{C}}} \exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_{\mathrm{mod},\mathrm{C}}}{\ln s_{\mathrm{C}}}\right)^{2}\right].$$

<sup>15</sup> Here, *r* is particle radius, and  $r_{mod}$ , *s*, and  $V_0$  are the mode radius, geometrical SD, and particle volume, respectively, with subscript F indicating fine particle mode and subscript C indicating coarse particle mode. The parameters  $r_{mod}$  and *s* for this aerosol model are taken from the rural aerosol model proposed by Hänel (1976). To simplify for aerosol properties representation, this paper introduces the coarse–fine particle <sup>20</sup> volume ratio  $\varsigma$ , defined as

$$\varsigma = \frac{V_{0,C}}{V_{0,F}}.$$

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(4)

(5)

(3)



From the above model setting, the spectral radiance-integrated RGB signals can be calculated by multiplying the simulated spectral radiance of twilight sky with the camera CMFs. The normalized brightness  $G_n$  and the chromaticity (R/G, B/G) can then be obtained in the way described in Sect. 2.

#### 5 3.2 Sensitivity tests

The sensitivities of the chromaticity and normalized brightness in twilight sky to tropospheric AOT  $\tau_{tr}$ , stratospheric AOT  $\tau_{st}$ , coarse–fine particle volume ratio  $\varsigma$ , and other atmosphere–surface characteristics were tested. For the sensitivity tests, the complex refractive indices of aerosol were assumed to be 1.5 for the real part and 0.01 for the imaginary part. The results are described in Figs. 3, 4, and 5. In the twilight sky, and particularly near the horizon, as  $\tau_{tr}$  increases,  $G_n$  and R/G decrease and B/G increases (Fig. 3a and b). These relations suggest that the twilight sky becomes dark and blue near the horizon, which is consistent with the results of Saito et al. (2013). Figure 3c and d shows the sensitivities of the twilight sky to  $\tau_{st}$ . With large  $\tau_{st}$ , the value of

- $G_n$  at VZAs of 75–85° increases and all R/G (resp., B/G) values increase (decrease), so that the reddened twilight glow is represented. These results are consistent with several reports on the very intense twilight glow (the so-called twilight phenomenon) that results from volcanic aerosol injection into the stratosphere by volcanic eruptions (Volz, 1964, 1965). The sensitivity tests show that R/G increases under high  $\varsigma$  con-
- <sup>20</sup> ditions only near the horizon (Fig. 3e and f). Figure 4 shows the sensitivities of the brightness and the chromaticity in twilight glow with VZA of 85°, SZA of 93°, and RAA of 0°. In the R/G–G<sub>n</sub> cross-section (Fig. 4b), G<sub>n</sub> depends strongly and inversely on  $\tau_{tr}$ , decreasing under typical aerosol property condition ( $\varsigma = 1$ ) as  $\tau_{tr}$  increases. In contrast, for R/G, sensitivity to  $\tau_{st}$  predominates, and R/G increases with large  $\tau_{st}$  (Fig. 4b).
- <sup>25</sup> The sensitivities of twilight glow are sufficient for modeling with  $\tau_{tr}$  in 0.01–0.5 and  $\tau_{st}$  in 0.002–0.1, while the chromaticity and normalized brightness in this sector are less sensitive to  $\varsigma$  (Fig. 4c and d). Figure 5 is the same as Fig. 4 except near the horizon with VZA of 88°, SZA of 91°, and RAA of 0°. The sensitivity of the twilight sky to  $\tau_{st}$  and



 $\varsigma$  is weak for  $\tau_{tr} = 0.13$  (Fig. 5a and b). Figure 5c and d suggests that the sky is more sensitive to  $\varsigma$  when  $\tau_{tr}$  is smaller. In such cases, R/G and G<sub>n</sub> increase with increasing  $\varsigma$ . As above, twilight sky is sensitive to  $\varsigma$  in the range of 0.2 to 5 under the clean atmospheric condition ( $\tau_{tr} \lesssim 0.1$ ). To summarize this section, the sensitivity tests showed that the normalized brightness and the chromaticity in the twilight sky have sufficient

sensitivity to tropospheric and stratospheric AOT and have moderate sensitivity to the coarse–fine particle volume ratio under small AOT conditions.

The sensitivity to other atmospheric and surface characteristics was investigated, with the results showing that the sensitivity to ozone column of twilight sky color is moderate, as previously shown by Hulburt (1953). However, the sensitivities of sky brightness and color to surface albedo, refractive indices, and water vapor column were found to be very weak (not shown).

## 3.3 Model setup for aerosol retrievals

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To apply the JACOSPAR RTM to the forward model for aerosol retrieval, we refined the
 RTM to allow faster computation by semi-analytically calculating singly scattered light and then linearly interpolating multiply scattered light from a lookup table (LUT), which was pre-calculated by the BMC method. Ozone column amount data were taken with the ozone monitoring instruments (OMI) onboard the Aura satellite and considered in the atmospheric model and in the LUT of the RTM. We assumed that other surface atmospheric conditions (such as: surface albedo, water vapor, and refractive indices) agreed with climatic values.

#### 4 Retrieval method and error analysis

#### 4.1 The retrieval method

The retrieval method is a maximum a posteriori (MAP) estimation method based on optimal estimation (Rodgers, 2000). This method derives an optimal solution from mea-





surement data under the constraints of prior information, and is presently in use for aerosol and cloud optical property remote sensing (e.g., Watts et al., 2011; Mateshvili et al., 2013; Iwabuchi et al., 2014). A measurement vector y derived from twilight photographs, a state vector x that includes aerosol optical properties as elements, and a set of model parameters p are defined as

$$\boldsymbol{x} = \begin{pmatrix} \ln \tau_{\rm tr} \\ \ln \tau_{\rm st} \\ \ln \zeta \end{pmatrix},$$
$$\boldsymbol{y} = \begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_n \end{pmatrix},$$
$$\boldsymbol{p} = \begin{pmatrix} \boldsymbol{\omega}_{\rm H_2O} \\ \boldsymbol{\omega}_{\rm O_3} \\ \boldsymbol{\alpha} \\ m_{\rm r} \\ m_{\rm i} \end{pmatrix},$$

where  $\omega_{H_2O}$  is column water vapor amount,  $\omega_{O_3}$  is column ozone amount,  $\alpha$  is surface albedo, and  $m_r$  and  $m_i$  are the real and imaginary parts, respectively, of the refractive indices of aerosol. The number of measurement vector elements *n* is set at 840 because the number is defined as the number of measurement elements multiplied by the number of measurement directions (3 of measurement elements and 280 of measurement directions, which result from 10 VZAs, 7 SZAs, and 4 RAAs). We can formally describe the physics as the forward model function  $f(\mathbf{x})$ :

 $\boldsymbol{y} = f(\boldsymbol{x},\boldsymbol{p}) + \boldsymbol{e},$ 

(6)

(7)

(8)

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where e is the measurement-model error vector. The cost function J(x) can be described as

$$J(\boldsymbol{x}) = (\boldsymbol{x} - \boldsymbol{x}_{a})^{\mathsf{T}} \mathbf{S}_{a}^{-1} (\boldsymbol{x} - \boldsymbol{x}_{a}) + [\boldsymbol{y} - f(\boldsymbol{x}, \boldsymbol{\rho})]^{\mathsf{T}} \mathbf{S}_{\boldsymbol{y}}^{-1} [\boldsymbol{y} - f(\boldsymbol{x}, \boldsymbol{\rho})],$$
(10)

where  $x_a$  is an a priori state vector,  $S_a$  is an a priori state error covariance matrix, s and  $S_v$  is a measurement-model error covariance matrix. The derivation of  $S_v$  will be described in Sect. 4.2. We summarize the a priori state and its error variance in Table 4. The assumed  $\mathbf{S}_{a}$  is a diagonal matrix, with the main diagonal taken as a vector of error variances from prior information. The prior state and its geometrical SD for  $\tau_{\rm tr}$  are assumed to be 0.13 and 1.19, respectively, which is taken from the values in Dubovik et al. (2002) for AOTs in urban, continental, and desert regions, which range 10 from 0.04 to 0.43, as found from ground-based observations. The a priori value of  $\tau_{st}$  is set to be 0.005 with a geometrical SD of 1.15 (Hess et al., 1998). The a priori value for c is assumed to be that of a rural aerosol model with a bimodal distribution of particle volume, as proposed by Hänel (1976). The geometrical SDs for the prior information are set to be large value to avoid overfitting of the state vector x, which 15 has components  $\ln \tau_{tr}$ ,  $\ln \tau_{st}$ , and  $\ln \varsigma$ . When the state vector becomes optimal, the cost function should be below the degrees of freedom of the measurement vector (Rodgers, 2000). Therefore, an optimal state  $x_{opt}$  gives

 $J(\mathbf{x}_{opt}) \approx n.$ 

<sup>20</sup> Because the measurement vector calculated according to the forward model is nonlinearly dependent on the state vector, we apply the Levenberg–Marquardt method (LMM) to solve the inverse problem. The LMM can provide a stable solution for moderately nonlinear problems (Rodgers, 2000). The method is iteratively applied to find an optimal solution for *x*. The state vector at the *i* + 1th iteration,  $x_{i+1}$ , is given by

<sup>25</sup> 
$$\mathbf{x}_{i+1} = \mathbf{x}_i + [(1+\gamma)\mathbf{S}_a^{-1} + \mathbf{K}_i^{\mathsf{T}}\mathbf{S}_y^{-1}\mathbf{K}_i]^{-1} \left\{ \mathbf{K}_i^{\mathsf{T}}\mathbf{S}_y^{-1}[\mathbf{y} - f(\mathbf{x}, \mathbf{p})] - \mathbf{S}_a^{-1}[\mathbf{x} - \mathbf{x}_a] \right\},$$
 (12)



(11)

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where  $\mathbf{K}_i$  is the Jacobian of the *i*th iterative calculation, and  $\gamma$  represents an adjustment adaptively chosen at each step so as to minimize the cost function. This construction lets the LMM avoid convergence at only local minima more readily than the Gauss– Newton method can. When the cost function is not sufficiently minimized to *m*, the forward model is unable to represent the measured twilight sky with the available data under the given assumptions and a priori constraint in the retrieval method. This can occur under two conditions: when clouds, fog and/or haze is present; and when the values of the state vector and model parameters are out-of-range for the retrieval method, which results in underestimation of the measurement–model error covariance. Simultaneously with obtaining an optimal solution, the uncertainty can be calculated by the following equation:

$$\mathbf{S}_{\boldsymbol{X}} = \left[\mathbf{S}_{a}^{-1} + \mathbf{K}^{\mathsf{T}}\mathbf{S}_{\boldsymbol{y}}^{-1}\mathbf{K}\right]^{-1},$$

where  $\mathbf{S}_{x}$  is an error covariance matrix whose diagonal elements represent the error variances of the solutions.

#### 15 4.2 Measurement–model error

Measurement–model error originates from the measurement error, assumed errors in the forward model, and uncertainties about model parameters. The matrix  $\mathbf{S}_{y}$  combining these three error components can be found as

$$\mathbf{S}_{\boldsymbol{y}} = \mathbf{S}_{\boldsymbol{y},m} + \mathbf{S}_{\boldsymbol{y},\boldsymbol{p}} + \mathbf{S}_{\boldsymbol{y},\mathsf{fwd}},$$

<sup>20</sup> where  $S_{y,m}$ ,  $S_{y,fwd}$  and  $S_{y,p}$  are the covariance matrices of the measurement error, the assumed errors in the forward model, and the uncertainties about the model parameters, respectively. These errors and the assumptions of the forward model are listed in Table 5. The real ( $m_r$ ) and imaginary ( $m_i$ ) parts of the refractive indices are assumed to be 1.5 and 0.01, respectively, which is taken from Hayasaka et al. (1992) by simplifying from the annual variation of refractive indices for aerosol optical properties in

(13)

(14)



Sendai, Japan, as obtained by ground-based measurements (1.44–1.57 real part and 0.007–0.057 imaginary part). The surface albedo  $\alpha$  is assumed to be 0.1 because of the annual variation of  $\alpha$  from 0.05 to 0.3, which was found by Zhou et al. (2003).

- The forward model error originates from uncertainties about the particle size distri-<sup>5</sup> bution, from the vertical profile shape assumed in the forward model, and also includes a smoothing error resulting from linear interpolation in the LUT. The forward model error is evaluated by adding Gaussian random noise to the parameters for aerosol particle size distribution and vertical profile shape (specifically, to the mode radii for the volume particle size distributions, and to the scale and shape parameters of the Gamma distributions for the vertical profile) and muscing the forward model and LACOCEAR DTM
- tributions for the vertical profile) and running the forward model and JACOSPAR RTM for perturbed sets of model-assumed parameters and finally calculating the difference between each model's output. The results from JACOSPAR RTM are taken as ground truth data. The dependence of assumption errors in the forward model on the measurement angles is shown in Fig. 6. The root-mean-square (RMS) errors of the chromaticity and the permetized brightness are capacitated by larger with larger SZAs and are amplied.
- and the normalized brightness are consistently larger with larger SZAs and are smaller with smaller VZAs and RAAs. The RMS errors are in the range 1-3% for R/G, 3-5% for B/G, and 5-10% for G<sub>n</sub>.

The model parameter errors are evaluated by adding Gaussian random noise to the model parameters shown in Table 5 and running the forward model for perturbed sets

- of model parameters. We show the dependence of the model parameter errors on the measurement angles in Fig. 7. The chromaticity errors depend weakly on the value of RAA and SZA and are moderately larger with larger values of VZA. In contrast, the error in G<sub>n</sub> rapidly increases with increasing SZA and is slightly larger with smaller values of RAA and larger values of VZA. The RMS errors for R/G and B/G are in the range 3–
- 5 %. The RMS error for G<sub>n</sub> is below 5 % for SZA of 90–94°, but in the range 20–80 % for SZA of 95–96°. The model parameter errors are the largest errors and are dominant in the model–measurement error. The covariance matrices for errors in the forward model assumptions and the model parameters are taken as constant matrices for the aerosol retrievals in this study. The applied algorithm could be improved by accounting for the





effects of the state vector and model parameter values on these matrices. This is left for future work.

The measurement errors include CMOS noise and biases, uncertainties in the inferred camera response functions, camera-orientation errors, and time-lag error during

- taking the photographs. CMOS biases can arise from exposure time, aperture, and temperature. CMOS noise errors for each angle are determined by the variance of RAW counts among pixels taken from the corresponding angles. The time-lag error is assumed to be normally distributed with SD 5 s, which gives an uncertainty of ~ 0.015° for SZA, and the camera-orientation error is assumed to be normally distributed with SD
- 0.5° for VZA and 0.3° for RAA. Further, the errors are estimated by running the forward model with Gaussian random noise perturbation of the camera angles. As a result, the CMOS noise is on the order of 3 to 5%, and other errors are very small at all angles, on the order of 0.5 to 1% for the chromaticity and normalized brightness.

## 4.3 Retrieval-error analysis

- <sup>15</sup> The errors in retrieved aerosol properties originate from the measurement–model error and the sensitivity of the state vector of the sky to atmospheric conditions. For the error sources in Table 3, the retrieval error is evaluated by retrieval simulations. First, after assuming a state *x* and model parameters *p*<sub>noise</sub> and adding Gaussian random noise, the forward-model-simulated measurements *y*<sub>noise</sub> are calculated, and Gaus-
- sian random noise is added to simulate measurement error and the forward model error. Second, we try to retrieve the state vector  $x_{noise}$  from  $y_{noise}$  with this algorithm. Finally, the truth-retrieval difference  $x_{noise} - x$  is obtained. Each state was simulated 100 times and the retrieval error was taken as the average error. The retrieval results with filtering (J(x) < 1000) are shown in Fig. 8. The value of  $\tau_{tr}$  is almost correctly re-
- <sup>25</sup> trieved, with RMS error of 0.14% and a slightly negative bias (~ 5%) in the range of 0.01–0.5 (Fig. 8a). The retrieval uncertainty is very small under the clean and moderately turbid atmospheric conditions for  $\tau_{\rm tr}$  from 0.01 to 0.3, suggesting that this method can retrieve tropospheric AOTs with high precision. In the more turbid atmospheric





conditions ( $\tau_{\rm tr} > 0.3$ ), the uncertainty in the retrieved  $\tau_{\rm tr}$  is around ±0.1. The value of  $\tau_{\rm st}$  is also correctly retrieved with a RMS error of 2.4 % and no notable bias (Fig. 8b). The retrieved value of  $\tau_{st}$  is overestimated under small tropospheric AOT conditions as a result of the particular characteristics of this retrieval method (not shown). The s uncertainties for  $\tau_{st}$  retrieval are small for large values of  $\tau_{st}$  and are slightly larger for small values of  $\tau_{st}$ , suggesting that the value of  $\tau_{st}$  can be retrieved accurately under the dense  $\tau_{st}$  conditions caused by volcanic eruption, such as the Mt. Pinatubo eruption in 1991. Figure 8c shows that the value of c can be retrieved when it is in the range of 1 to 5 and there is a clean atmospheric conditions ( $\tau_{tr} \sim 0.1$ ). In this case (blue marks in the figure), the RMS error is somewhat large (18.2%). Additionally, the retrieved val-10 ues of c are scattered, centered mostly around the a priori assumption under moderate and dense aerosol conditions ( $\tau_{tr} > 0.13$ ). This occurs because the sensitivity of  $\zeta$  on twilight sky is weak with large tropospheric AOTs, as shown in Sect. 3.2. When small particles dominate ( $\zeta < 1$ ), the retrieved values of  $\zeta$  are almost equal to the a priori assumption. Figure 8d shows the retrieved c with the retrieval method for fixed and given 15 truth  $\tau_{st}$  values. The RMS error in the  $\zeta$  retrievals is smaller (at 13.3%) than that for retrievals under non-fixed  $\tau_{st}$  values regardless, and this is independent of  $\tau_{tr}$  in the range 0.01–0.5. This suggests that the accuracy of the  $\zeta$  retrievals can be improved if the  $\tau_{st}$  is accurately known from other instruments. Furthermore, no dependence of

retrieved  $\zeta$  on stratospheric AOTs is found, suggesting that the slightly large RMS error in the  $\zeta$  retrievals might result from error in retrieving  $\tau_{st}$ . In addition, the degrees of freedom for the signals obtained from the measurements are around 3 in most cases, showing that the measurement data are sufficient for retrievals by this method.





#### 5 Results

#### 5.1 Validation

To validate this retrieval method, the retrievals from photographic observations were compared with those from the skyradiometer measurements located at the same site and with satellite-based measurements (Fig. 9). The comparison is limited to selected results with J(x) < 3000. The skyradiometer and an the software tool Skyrad.pack developed by Nakajima et al. (1996) were used as the measurement system and tool to infer aerosol optical properties, such as AOTs and distributions of particle volumes, from direct and diffused sunlight. For twilight, skyradiometer measurements are not available because of the absence of direct sunlight. Typically, aerosol optical properties do not vary much within a few hours except when a strong emission source (e.g., forest fire) is present near the site. For this reason, we used the skyradiometer data obtained an hour before sunset and an hour after sunrise for comparison in this study. Figure 9a shows the comparison of AOTs ( $\tau_{tr} + \tau_{st}$ ) retrieved from the photographic ob-

- <sup>15</sup> servations and the skyradiometer measurements. Mostly, the retrieved AOTs from the digital camera were consistent with those from the skyradiometer, with little uncertainty and a coefficient of determination of 0.86. The values of  $\tau_{st}$  retrieved from the camera data were compared with those from the Canadian optical stratospheric and infrared imaging system (OSIRIS) onboard the Swedish satellite Odin (Murtagh et al., 2002;
- <sup>20</sup> Llewellyn et al., 2004). OSIRIS measures atmospheric limb radiance of scattered sunlight across visible wavelengths, and optical thickness, and the vertical profiles of the extinction coefficient in stratospheric aerosol were inferred by the use of the algorithm given by Bourassa et al. (2007). The photographic observations are carried out under the background stratospheric aerosol conditions, in other words, without any effects of
- <sup>25</sup> volcanic eruptions in the stratosphere. Figure 9b shows the scatterplot of  $\tau_{st}$  retrieved from the camera and from OSIRIS. A weak correlation exists for this measurement period, and this weakness results from the large variability of  $\tau_{st}$  according to the camera providing the data, with little variability in  $\tau_{st}$  for data from OSIRIS. The values of





 $\varsigma$  retrieved from the camera are compared with those calculated from particle volume distributions retrieved from the skyradiometer (Fig. 9c). The retrieved value of  $\varsigma$  tends to be close to the a priori assumption for moderate and large AOTs ( $\tau_{tr} > 0.13$ ) and vary widely for small AOTs ( $\tau_{tr} < 0.13$ ). This disparity results from the weak correlation between the camera and the skyradiometer.

#### 5.2 Case studies

To test the flexibility under various aerosol conditions (e.g., vertical profiles and aerosol species) of this retrieval method, we chose three characteristic cases and analyzed the simulation results in detail with respect to the vertical profiles of the aerosol extinction coefficients. Figure 10 shows photographs of the twilight sky taken on (a) 31 January 2013, (b) 23 May 2013, and (c) 3 June 2013. These photographs were taken at an SZA of 94°. In Fig. 10a, a bright red-orange twilight glow is seen near the horizon, and the twilight sky color gradates from orange to blue. The brightness rapidly decreases as

- the VZA decreases. In contrast, the color in Fig. 10b seems to be dull across the sky in comparison with Fig. 10a. An orange-to-gray twilight glow is present above a dark-blue segment that appears near the horizon (Saito et al., 2013). In Fig. 10c, the range of the twilight sky color is the smallest among the three cases, and a dark-blue segment can be seen near the horizon, similar to that in Fig. 10b. Figure 11 shows the vertical profiles of aerosol extinction coefficients in the troposphere from the ground to an al-
- titude of 6 km; these are derived from the lidar measurements for each case (Shimizu et al., 2010). In the lidar products, the lidar ratio of 50 sr is assumed, and the inferred extinction coefficients are separated into spherical and non-spherical particles.

Table 6 summarizes the results from the ground-based observations (the sunphotometer and the skyradiometer, collocated at the photographic observation site) and

from the daily means of aerosol optical properties, as calculated by the aerosol transport model SPRINTARS (Takemura et al., 2000, 2002, 2005). The sunphotometer measures direct solar light in the visible to near infrared wavelengths to infer the AOTs. For 31 January 2013 (case a), very clean atmosphere with an AOT of 0.02–0.03 is





simulated by the SPRINTARS and weak backscatter signals are obtained from lidar measurement. Moreover, spherical particles are dominant in this case. In contrast, the SPRINTARS products suggest that sulphate and dust particles were transported from the eastern part of continental China to Japan from 20 to 26 May 2013. Consistently,

- the lidar measurements show the presence of nonspherical particles at altitudes of 3–6 km and the presence of spherical/nonspherical mixed particles in the boundary layer for 23 May 2013 (case b). For 3 June 2013 (case c), dust transported from the north inland area of China to Japan from 2 to 5 June 2013 is shown by the model. A spherical/nonspherical mixture of particles is present in the boundary layer (altitudes of 0–2 km), and nonspherical particles are dominant above 6 km, according to the lidar
- of 0–2 km), and nonspherical particles are dominant above 6 km, accord measurements.

From the above results, the atmospheric characteristics are clean atmosphere in case (a), dust-pollutant mixed particles transported from eastern China in case (b), and yellow sand transported from inland China in case (c). Additionally, the vertical profiles

- and aerosol species are vertically inhomogeneous in cases (b) and (c). A comparison of the results retrieved from the photographic observations with the corresponding results from other products shows that the retrieved AOTs are reasonable for cases (a) and (b), with AOTs of  $0.074 \pm 0.012$  (a) and  $0.101 \pm 0.006$  (b) relative to estimates from other ground-based passive instruments. The retrieved values of  $\zeta$  are 2.86 ± 0.34 (a)
- <sup>20</sup> and  $0.98 \pm 0.07$  (b). For case (c), an AOT of  $0.251 \pm 0.006$  is inferred from the camera, which is slightly lower than the AOT obtained from other products. The main reason for this is that the inferred  $\tau_{tr}$  is negatively biased at larger AOTs due to the characteristics of the present method. Additionally, it might be that the temporal–spatial variability of aerosol optical properties under dust events affects the bias. Specifically, during
- <sup>25</sup> dust events, aerosol optical properties showed high variability, and so they varied more widely during the time-lag between camera observations and other measurements than they did in the other cases, resulting in the difference in AOT. The retrieved value of gis eliminated in the case (c) because of low confidence in the g retrieval, which was predicted by the retrieval error analysis (Sect. 4.3) for large AOT conditions. The AOTs





for those three cases are consistent with the total AOTs calculated by SPRINTARS. Thus, the case studies demonstrate that the proposed method can infer AOTs without much dependence on knowing the vertical inhomogeneity for aerosol species and the extinction coefficients.

## **6** Conclusions and discussions

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An optimal estimation-based algorithm was developed to infer aerosol optical properties from twilight photographs taken with a digital camera at SZAs of 90–96°. The following advantages are offered by this method and measurement techniques: (1) tropospheric and stratospheric AOTs can be retrieved by a passive remote sensing technique, even in the absence of direct sunlight conditions, (2) the retrievals and their uncertainties are simultaneously calculated in the optimal estimation framework; and (3) a low cost instrument (a digital camera) can be used.

The measurement vector was set to the normalized brightness (G<sub>n</sub>) and the chromaticity (R/G, B/G) around the solar direction in the angle range of about 0–30°, as <sup>15</sup> derived from camera RAW data in RGB channels. Ozone column data were taken from OMI/Aura. The forward model includes a spherical-shell atmosphere, refraction, and multiple scattering. Additionally, in the model, singly scattered radiances are calculated semi-analytically, and multiply scattered radiances are calculated by linear interpolation from a pre-calculated LUT. The values of diagonal elements in the a priori state

- <sup>20</sup> covariance matrix were assumed to be large in order to avoid overfitting. The model parameters and their uncertainties were determined by prior measurement and error analysis. The error sources during retrieval were chiefly the model parameter uncertainties, with each of CMOS noise, the assumed distribution of particle volume, and vertical profile shape contributing some error. The retrieval-error analysis showed that
- the tropospheric and stratospheric AOTs were accurately inferred for  $\tau_{tr}$  in the range 0.01 to 0.5 and  $\tau_{st}$  in the range 0.001 to 0.1, obtaining a very small RMS error (below 1 % for  $\tau_{tr}$  and 3 % for  $\tau_{st}$ ). The retrieved AOTs from the digital camera were compared



with those from the skyradiometer measurements, and this comparison showed consistency between the results, with a coefficient of determination as large as 0.86. A case study for three different atmospheric conditions was presented to demonstrate the usefulness of the method. Comparison with products provided by other instruments and results from other model was used to investigate the effects of vertical inhomogeneity

for aerosol species and profiles in detail. This investigate the enects of vertical innomogeneity retrieved with reasonable accuracy without knowing the details of such properties.

The difficulty in retrieving the coarse–fine particle volume ratio  $\varsigma$  can be explained by the following. In cases with a large AOT, the following are true: the sensitivity of  $\varsigma$  in

- <sup>10</sup> twilight sky is weak, and retrieval uncertainty for  $\tau_{st}$  may lead to errors in c retrievals. In cases with a small AOT, one possible explanation is that polarization effects cause non-negligible differences between the twilight sky brightness and color as found by the forward model simulations and that found by measurement. The forward model uses a scalar approximation to model the radiative transfer. Saito et al. (2013) investi-
- <sup>15</sup> gated the contribution of multiply scattered light to twilight sky near the horizon with the JACOSPAR RTM, from which the present forward model is descended. The fraction of multiply scattered light is in the range of 20 to 50 % near the horizon for a VZA of 90–85° under small AOT conditions (0.05 <  $\tau_{\rm tr}$  < 0.13). Mishchenko et al. (2006) demonstrated that scalar approximation leads to biases for calculated radiance, with the magnitude
- of the bias in the range of  $\pm 3-5$  % depending on the RAA, relative to the vector RTM outputs for AOTs of 0.1–0.3. This may cause an error in the retrieved brightness and color in twilight near the horizon. Another possible reason is the seeming insensitivity of  $\zeta$  in twilight sky when looking near the horizon (with SZA of ~ 90°), and this sensitivity may be too low to overcome the uncertainties resulting from model errors and camera noise. An investigation of this possibility and improvements to the algorithm to
- compensate are left for future work.

The retrieval method proposed by this paper can be applied to aerosol measurements in desert, snow-covered, and polar regions, areas where it is difficult to apply the conventional solar reflection-based methods. Particularly in the polar region in winter,





aerosol measurements are very limited, and the present twilight photometry method should be useful to increase the availability of aerosol measurements. Furthermore, it is easy to extend the aerosol observations at many points with this method because of the use of a digital camera, which is both inexpensive and easily movable. Digital

cameras are widely used in daily life, and many amateur photographers have the opportunity to take pictures of beautiful sights, including twilight sky. In addition to their beauty, twilight photographs have the potential to be used as measurement data for aerosol retrievals. Therefore, the present aerosol retrieval method has unique characteristics, and it may complementarily be used in combination with other measurement techniques.

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measuring aerosol

optical properties

from digital twilight

photographs

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 Table 1. Camera equipment characteristics.

Equipment	Period of observation	Coordinates	ISO	f-stop
Nikon D7000 equipped with a fish-eye lens	Jan 2013 to Aug 2014	Sendai, Japan (38°15′ N, 140°50′ E)	200	f2.8

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Table 2. Viewing and solar directions used for aerosol retrievals.

Direction	Angles [°]
Viewing zenith angles (VZA)	60, 70, 75, 80, 82, 84, 85, 86, 87, 88
Relative azimuth angles (RAA)	0, 10, 20, 30
Solar zenith angles (SZA)	90, 91, 92, 93, 94, 95, 96





Table 3. Parameters for the aerosol vertical profile assumed in the forward model.

Vertical profile	ξ	Ψ
Background bbg	1	1
Boundary layer b <sub>bl</sub>	2	1
Upper layer b <sub>up</sub>	5	0.8
Stratosphere $\dot{b}_{st}$	12	1.5





**Table 4.** Prior information and its error range for the state vector elements.

Variable	x	<b>x</b> a	Geometric SD	Min	Max
Tropospheric AOT Stratospheric AOT Coarse–fine particle volume ratio	In $ au_{ m tr}$ In $ au_{ m st}$ In $ au$	ln 0.13 ln 0.1 ln 1.08	1.19 1.15 2.3 (= ln 10)	ln0.01 ln0.002 ln0.2	ln0.5 ln0.1 ln5



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Table 5. Reference values and variabilities assum	ned for error analysis.
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Error source	Variable	Assumed value	Assumed variability ( $\sigma$ )
Measurement error	у	_	Defined by <b>S</b> <sub>v.m</sub>
Forward model approximation	As above	-	Defined by $\mathbf{S}_{y,\text{fwd}}$
Model parameters	In <i>w</i> <sub>H₂O</sub>	8.62 kg m <sup>-2</sup>	$2.58 < \omega_{H_2O} < 25.8 \mathrm{kg m}^{-2}$
	Ø <sub>O3</sub>	321 DU	280 < $\hat{\omega}_{O_3}$ < 360 DU
	m <sub>r</sub>	1.5	1.44 < m <sub>r</sub> < 1.57
	In m <sub>i</sub>	0.01	0.001 < <i>m</i> <sub>i</sub> < 0.1
	ln a	0.1	$0.05 < \alpha < 0.3$





<b>Table 6.</b> Retrieval result	s and SPRINT	ARS models	simulati	on resu	ilts for cas	se studies.	
Date		Retrie	vals			SPRINTA	AS AOT
Date	Cam	Retrie	vals Sky	/rad	Sunpho	SPRINIA	ARS AOT
Date	Cam	Retrie era ς	vals Sky AOT	rad ς	Sunpho AOT	SPRIN IA Sulfate	Dust

 $0.98 \pm 0.07$ 

NA

1.193

1.366

0.122

0.311

0.03-0.05

0.03-0.05

0.1-0.2

0.1-0.2

0.112

0.296

 $0.101 \pm 0.006$ 

 $0.251 \pm 0.006$ 

NA: not available

(b) 23 May 2013, morning

(c) 3 Jun 2013, morning







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**Figure 2.** Inferred color-matching functions of Nikon D7000 in this study (solid lines) and of D200 (dotted lines) from Sigarnes et al. (2008).







**Figure 3.** Sensitivity test results for normalized brightness ( $G_n$ ) and chromaticity (B/G, R/G) of twilight sky: (**a**, **b**) sensitivities to tropospheric aerosol optical thickness; (**c**, **d**) sensitivity to stratospheric aerosol optical thickness; (**e**, **f**) sensitivity to coarse–fine particle volume ratio. Panels on the right side (**b**, **d**, **f**) show the dependence of normalized brightness variations along with the viewing zenith angles. Panels on the left side (**a**, **c**, **e**) show the dependence of chromaticity variations along with the viewing zenith angles on R/G–B/G cross sections. SZA and RAA are 93 and 0°, respectively.







**Figure 4.** Sensitivity test results for chromaticity values of the twilight glow (VZA of 85° and SZA of 93° at the solar azimuth angle): **(a, b)** show the dependence of chromaticity variations on the R/G-B/G cross section; **(c, d)** show the dependence of normalized brightness variations on the  $R/G-G_n$  cross section. Dashed curves in the panels on the right **(b, d)** are isolines of coarse–fine particle volume ratio, and solid curves are isolines of tropospheric aerosol optical thickness. Dashed curves in the panels on the left **(a, c)** are isolines of stratospheric aerosol optical thickness, and solid curves are isolines of tropospheric aerosol optical thickness.







**Figure 5.** Sensitivity test results for chromaticity values of the twilight sky near the horizon (VZA of 88° and SZA of 91° at the solar azimuth angle): **(a, b)** show the dependence of chromaticity variations on the R/G-B/G cross section; **(c, d)** show the dependence of normalized brightness variations on the  $R/G-G_n$  cross section. Dashed curves in the panels on the right **(b, d)** are isolines of tropospheric aerosol optical thickness, and solid curves are isolines of coarse–fine particle volume ratio. Dashed curves in the panels on the left **(a, c)** are isolines of coarse–fine particle volume ratio, and solid curves are isolines of stratospheric aerosol optical thickness.







**Figure 6.** Forward model errors in (**a**, **b**) R/G, (**c**, **d**) B/G, and (**e**, **f**) G<sub>n</sub> caused by the model assumptions as functions of SZA and VZA (see text for details). Panels on the left (**a**, **c**, **e**) have RAA = 0°, and those on the right (**b**, **d**, **f**) have RAA = 30°.







**Figure 7.** Forward model errors in (**a**, **b**) R/G, (**c**, **d**) B/G, and (**e**, **f**) G<sub>n</sub> caused by uncertainties in the model parameters as functions of SZA and VZA. Panels on the left (**a**, **c**, **e**) have RAA =  $0^{\circ}$ , and those on the right have RAA =  $30^{\circ}$ .







**Figure 8.** Initial and retrieved (a) tropospheric aerosol optical thickness, (b) stratospheric aerosol optical thickness, and (c) coarse–fine particle volume ratio  $\zeta$  in the retrieval-error analysis. (d) Same as (c) but retrievals under fixed stratospheric aerosol optical thickness. Blue and green marks show  $\zeta$  retrievals under very and moderately clean conditions, respectively.







**Figure 9.** Comparison of aerosol property retrievals from camera measurements and skyradiometer measurements. Error bars show uncertainty of retrievals. Color scales show total retrieval cost J(x). The vertical axis and horizontal axis show retrievals from a camera and those from other instruments, respectively: **(a)** aerosol optical thickness, from camera and skyradiometer, with coefficient of determination  $r^2$  at 0.86 and 18 trials; **(b)** stratospheric aerosol optical thickness, from camera and OSIRIS onboard the Odin satellite; **(c)** coarse–fine particle volume ratio, from camera and skyradiometer, with black X marks indicating retrievals in dense aerosol conditions ( $\tau_{tr} > 0.13$ ).







Figure 10. Photographs of twilight sky on the solar side, for the case studies of (a) 31 January 2013, (b) 23 May 2013, and (c) 3 June 2013. In all cases, SZA is uniformly 94°, and VZA is 90.45° at the horizon, 91.5° at the bottoms of the images, and 30° at the tops of the photographs.



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