Development of correction method for integrating nephelometer and recent trend of aerosol optical properties based on ground-based measurement at Tsukuba

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Abstract

Measurements of Integrating nephelometer has error due to the truncation of light detection area. We developed a correction method using multi-wavelength nephelometer and absorption photometer. The accuracy of the method was evaluated using the simulation data. Applying this method to the data measured at Tsukuba, the recent trend of aerosol optical properties was investigated.

***Keywords* :** Integrating Nephelometer, PSAP, Scattering Coefficient of Aerosol, Absorption coefficient of Aerosol, Single scattering albedo of aerosol, asymmetry factor of aerosol

1. Introduction

The optical property of aerosol is one of the important parameters for the earth radiation budget and the atmospheric environment. In order to investigate the optical property of aerosol and to monitor the change of atmospheric environment, scattering and absorption coefficients of aerosol are measured at ground-based observation sites. The scattering coefficient is frequently measured using an integrating nephelometer. The integrating nephelometer cannot measure the light scattered to the extreme forward direction (scattering angle is nearly 0 degree.) and the extreme backward direction (scattering angle is nearly 180 degree.). Therefore, even if the distribution of light source in the nephelometer is correct, the scattering coefficient measured by the nephelometer is underestimated. In this study, the method to correct the scattering coefficients measured using the three wavelength nephelometer and three wavelength absorption photometer is developed. The accuracy of the method is evaluated by the simulation using OPAC (Hess et al, 1998) model. The method is a statistical retrieval one and the volume size distribution and the complex refractive index are simultaneously estimated.

Applying this method to the data measured at Tsukuba during the period from 2002 to 2010, the trend of optical properties of aerosol was investigated.

2. Methods

We use statistically optimized estimation method. The optimized estimate of the parameter is considered to be a value that maximized the following likelihood function

$$
\begin{aligned}
L(\mathbf{x}^b, \mathbf{f}^o, \mathbf{x}) &= P_b(\mathbf{x}^b)P_o(\mathbf{f}^o) \\
&= \frac{1}{(2\pi)^{(n+m)/2}\left|\mathbf{B}\right|^{1/2}\left|\mathbf{R}\right|^{1/2}} \\
&\times \exp\left[-\frac{1}{2}(\mathbf{x}-\mathbf{x}^b)^T\mathbf{B}^{-1}(\mathbf{x}-\mathbf{x}^b) - \frac{1}{2}(\mathbf{f(x)}-\mathbf{f}^o)^T\mathbf{R}^{-1}(\mathbf{f(x)}-\mathbf{f}^o)\right]
\end{aligned}
$$

When the following function J is minimum, the above function is maximized.

$$
\begin{aligned}
J(\mathbf{x}) &= \frac{1}{2}(\mathbf{x}-\mathbf{x}^b)^T\mathbf{B}^{-1}(\mathbf{x}-\mathbf{x}^b) \\
&+ \frac{1}{2}(\mathbf{f}(\mathbf{x})-\mathbf{f}^o)^T\mathbf{R}^{-1}(\mathbf{f}(\mathbf{x})-\mathbf{f}^o)
\end{aligned}
$$

, where $\mathbf{x}$ is parameter to be determined, $\mathbf{x}^b$ is the initial estimate, $\mathbf{f}^0$ is measurement, $\mathbf{f}(\mathbf{x})$ is measurement estimated by forward model. $\mathbf{B}$ and $\mathbf{R}$ are covariance matrices.

In this study, scattering and backscattering coefficients measured by TSI model 3563 at the wavelength of 450, 550 700nm and absorption coefficients measured by Particle Soot Absorption Photometer (PSAP) at wavelength of 462, 526, 650nm are considered as measurement values.

Given the measurement values, we estimate the size distribution of aerosol and complex refractive index using the above method. Using the estimated parameters, we calculate the single scattering properties (scattering coefficients, single scattering albedo, and asymmetry factor) based on Mie Theory. These values are regards as the corrected scattering coefficients and single scattering albedo.

3. Simulation of measurement

The accuracy of the method was evaluated using simulation data. The simulated data is calculated based on OPAC (Hess et al, 1998) model. OPAC model consist of 10 aerosol types.

Each aerosol type is external mixture of basic models. The single scattering properties of mineral dust are calculated using the database for spheroid by Dubovik et al. (2006). The single scattering properties of the other aerosol are calculated based on Mie theory. The truncation of integrating nephelometer is taken into consideration. Each aerosol type includes hygroscopic aerosol. Therefore, scattering properties for each aerosol type depend on relative humidity.

4. Parameter setting and initial value

The statistical optimized method needs the error covariance for each data of measurements and the initial estimates of parameters to be retrieved. We assume that the off-diagonal elements of covariance matrices are zero and diagonal elements are non zero. The following standard deviations (σ) are used in our method.

Scattering and back scattering coefficients:
σ=0.5E-06 (1/m)

Absorption coefficient: σ=0.5E-06 (1/m)

Index of refraction (real part):
σ=(log(1.6)-log(1.4))/2～0.029

Index of refraction （imaginary part） :
σ=(log(0.05)-log(0.005))/2～0.5

Volume size distribution : σ=Δlog(dV/dlogR)～2

Smoothing constrain is also treated as a measurement. It is assumed that the second derivative is zero.

Wavelength dependence of index of refraction:
difference between next points is 5%.

Volume size distribution spectrum:
difference between next points is factor 2.

The range of particle radius is R=0.00631 to 25.1 μm and divided into 19 bins with ΔlogR＝0.2 width. The range of wavelength is λ=0.316 to 1.0 μm and divided into 6 wavelength with Δlogλ＝0.1.

The power low distribution is assumed as the initial estimate of number size distribution.

$$dN/dr = C \qquad r < 0.05\mu\text{m}$$
$$= Cr^{-(\alpha+3)} \qquad 0.05\mu\text{m} \le r < 8\mu\text{m}$$
$$= Cr^{-6} \qquad 8.0\mu\text{m} \le r$$

, where α is Ångström exponent.

It is assumed that the initial real part of index of refraction is 1.5. The imaginary part is determined as the estimated single scattering albedo (SSA) at each wavelength is equal to measurement SSA. When we minimize $J(\mathbf{x})$, Gauss-Newton method is used and the initial estimate is replaced by the solution of the previous step.

5. Test results

In Fig. 1, the comparison among truth, measurement and corrected SSA at the wavelength of 550nm is shown. In every case, corrected values are closer to the truth one than measurements. In Desert model (5), the truth is calculated by non-spherical particle model. Even in this case, the correction by spherical model works well. The RMS difference between truth and measurement values is 0.009 to 0.015. The RMS difference between truth and corrected values is 0.002 to 0.004; relative error is 0.3 to 0.5%.

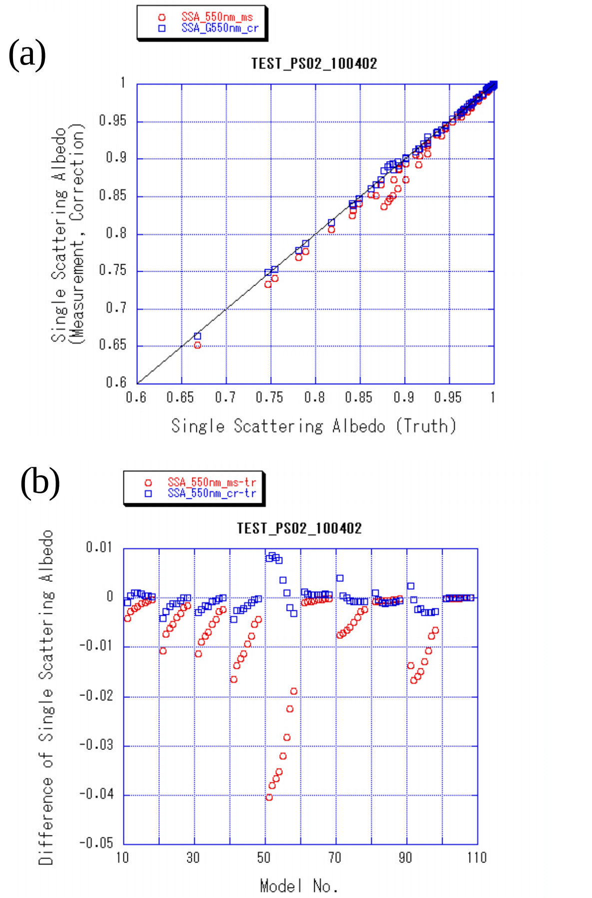

Fig. 1. (a) Comparison among truth, measurement (open circles) and corrected (open squares) single scattering albedo (SSA) at 550nm. (b) difference of SSA between truth and measurement (open circles) and difference of SSA between truth and corrected one (open squares). Model no. = OPAC model no. x10 + humidity ID(1-8). Relative humidity values are 0, 50, 70, 80, 90, 95, 98, 99%.

We also examined accuracy of corrected (estimated) values in the cases that scattering and absorption coefficients have error. In Fig. 2, the comparison among truth, measurement and corrected SSA at 550nm is shown in the case of absorption coefficient with +3% systematic errors. When SSA is small, the error becomes large. But, in most cases, the influence of error was small. We also examined accuracy in the case of absorption coefficient with +5% errors (Figures are not shown here). Even in this case, the influence of error was small in most cases, because the scattering coefficients are larger than absorption coefficients by several times or one order.

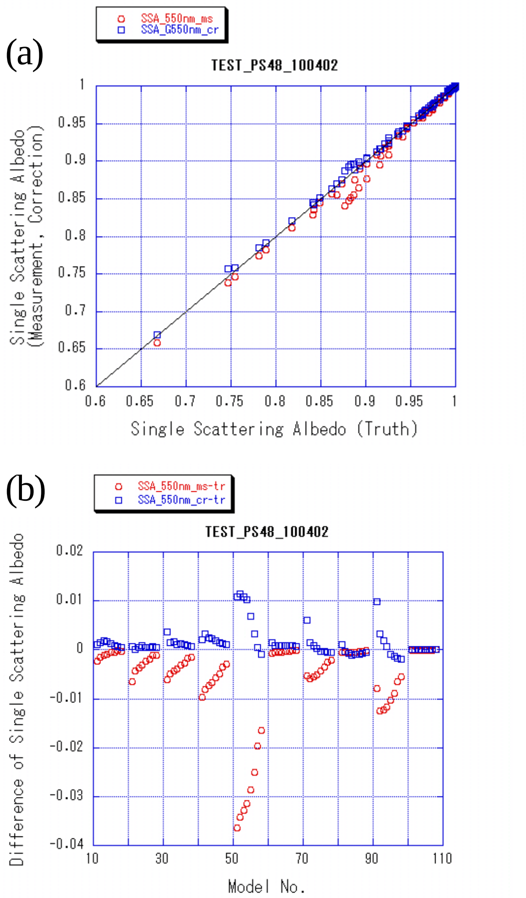

Fig. 2 Same as Fig. 1 except results in the case of absorption coefficient with 3% error.

Though we do not show figures here, the following additional results were obtained.

(1) The measured scattering and absorption coefficients are reconstructed within RMS 3E-07(1/m) and 8E-08(1/m), respectively.

(2) The scattering coefficients are estimated within RMS 1.2E-03 (1/m). When particles are large (for example, relative humidity is high), error is large.

(3) The extinction coefficient shows the same tendency as the scattering coefficients.

(4) The 1% and 3% error of scattering coefficient do not affect to the SSA estimation. In the case of 5% error, the error of the estimated SSA is the same order as the SSA calculated using measurement values.

(5) The RMS error of estimated asymmetry factor is 0.03.

(6) The small structure of size distribution cannot be estimated. In many cases, the mono-modal size distribution is estimated, even if the original size distribution is bi-modal one.

(7) When smoothing constrain of size distribution is strict, there is a tendency that the size distribution in the region of larger particle size becomes large as the particle size increased.

(8) The correlation between the original and estimated effective radiuses is 0.85. But, the estimated effective radius is 1.6 times as large as the original one.

(9) The volume weighted index of refraction is not close to the estimated one. The wavelength dependence is similar to the volume weighted one.

6. Aerosol optical properties at Tsukuba

Applying the above method to the data measured at Tsukuba during the period from 2002 to 2010, the trend of optical properties of aerosol was investigated.

The scattering coefficients are measured using TSI model 3563 at 3 wavelengths 450,550,700nm during the period from January 2002 to present. The absorption coefficient is measured using PSAP at a wavelength 565nm during the same period as TSI nephelometer. The absorption coefficients are measured using PSAP at 3 wavelengths 462, 526, 650nm during the period from March 2006 to present. We only analyzed the data in the dry condition; the relative humidity in nephelometer inlet is less than 50%. When the data of one wavelength PSAP is used, it is assumed that the wavelength dependence of absorption coefficient is $\lambda^{-1.1}$. 1.1 is the mean value from 2006 to 2010.

In Fig. 3, scattering and absorption coefficients are shown. After 2006, both scattering and absorption coefficients are decreasing. The change of absorption coefficients (Cabs) shows seasonal variation. In the winter season, Cabs is large and in the summer season, Cabs is small.

In Fig. 4, SSA at the wavelength of 550nm is shown. The large part of SSA (550nm) is between 0.8 and 0.9 and almost constant. After 2007, SSA(550nm) is slightly increasing. The change of SSA shows seasonal variation. In the winter season, SSA is small, and in the summer season, SSA is large. This seasonal variation is consistent with the change of absorption coefficient.

In Fig. 5, asymmetry factor (g) at the wavelength of 550nm is shown. g(550nm) is between 0.5 and 0.7. After 2007, g(550nm) is slightly decreasing. The change of asymmetry factor also shows seasonal variation. In the winter season, g(550nm) is small, and in the summer season, g(550nm) is large.

Though figures are not shown here, the changes of

Ångström exponent and effective radius (Reff) show seasonal variation. In the winter season, Ångström exponent is high and in the summer season, Ångström exponent is low. In winter season, Reff is small, and in the summer season, Reff is large. After 2008, Ångström exponent is slightly increasing and Reff is slightly decreasing. The trend of g(550nm) is consistent with those of Ångström exponent and Reff.

7. Summary

We examined accuracy of the method developed using that simulation data based on OPAC model. The measured scattering and absorption coefficients can be reconstructed within RMS =2.4 to 3.1E-07(1/m), 0.4 to 0.8E-07(1/m). SSA can be estimated with RMS =0.002～0.004 (relative error 0.3～0.5%). In all cases, the estimated SSA is closer to the truth than the non-corrected measurement one. The RMS error of corrected scattering coefficients was 1.2E-05(1/m), and when the aerosol consists of the larger size particles (for example, relative humidity is high case), the accuracy of estimated scattering coefficient is not good. The RMS error of asymmetry factor is 0.03.

In our method, volume size distribution and index of refraction are retrievel. The small structure of size distribution cannot be estimated. The correlation between the original and estimated effective radiuses is 0.85. But, the estimated effective radius is 1.6 times as large as the original one. We compared between the estimated index of refraction and the volume weighted one, but it is not found the quantitative coincidence between both indexes of refraction. The wavelength dependence was qualitatively coincident.

Applying this method to the data measured during 2002 to 2010 at Tsukuba, the trend of aerosol optical properties is investigated. After 2006, the scattering and absorption coefficients are gradually decreasing. SSA is almost constant, but after 2007, SSA is slightly increasing. The asymmetry factor and Ångström exponent of extinction coefficient also has similar trend. After 2007, the asymmetry factor and Ångström exponent are slightly decreasing and increasing, respectively.

References

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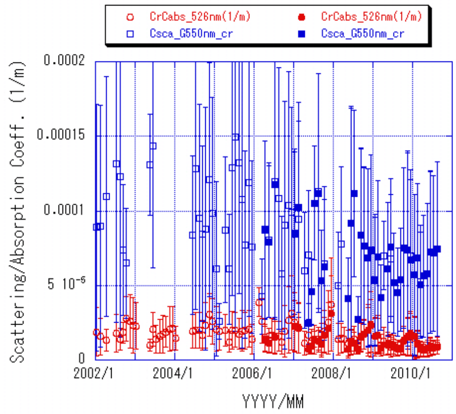

Fig. 3 monthly mean values and standard deviation of scattering coefficients at 550nm and absorption coefficient at 526nm. Open symbols mean that the correction was made using 3wavelngth (3λ) nephelometer and 1wavelength (1λ) PSAP. Closed symbols mean that the correction was made using 3λ nephelometer and 3λ PSAP.

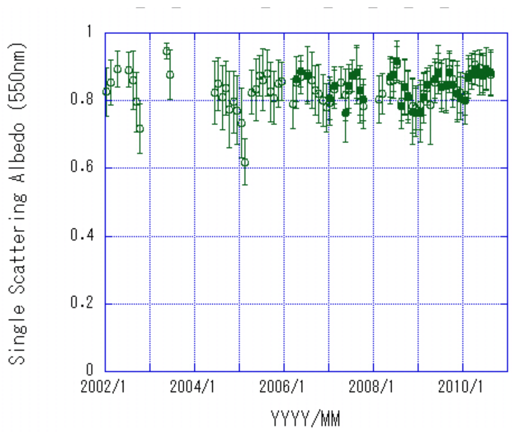

Fig. 4 Same as Fig. 3 except SSA.

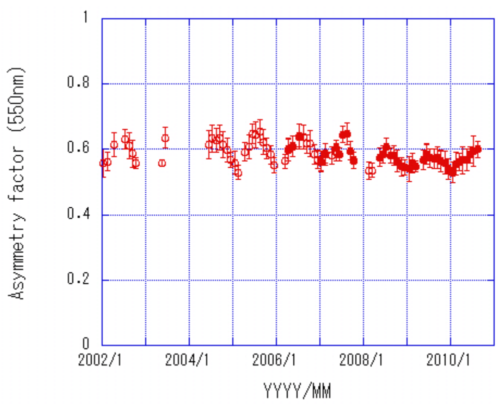

Fig.5 Same as Fig. 3 except asymmetry factor.