A-3.1 Studies on deriving aerosol optical properties using solar occultation ozone sensors data

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Abstract

In the signal processing of data obtained by the infrared spectrometer of the Improved Limb Atmospheric Spectrometer (ILAS), corrections must be made for the continuous absorption spectral components originating from aerosols/PSCs (polar stratospheric clouds) from those originating from gas. Such continuous absorption spectra can provide type identification of aerosols/PSCs. The simulations showed that the proposed method for correction is capable of identifying aerosols/PSCs types and estimating particle-size distribution parameters. For actual ILAS data, however, this method could not work well because of the offset components due to yet-unknown causes. As a realistic means, the non-gaseous components were approxomated by a linear interpolation of extinctions at the window channels, which proved to provide reasonable corrections.

Key Words solar occultation, stratospheric ozone layer, aerosol, polar stratospheric clouds, ILAS, Improved Limb Atmospheric Spectrometer

1. Introduction

The Improved Limb Atmospheric Spectrometer (ILAS), which the Environment Agency installed on the Advanced Earth Observing Satellite (ADEOS) (launched in August 1996, but operations were ceased in June 1997), is a sensor for measuring atmospheric trace gas species based on the solar occultation method. The major element of ILAS is a low spectral resolution infrared spectrometer, which measures an infrared wavelength region ranging from about 6 μm to 12 μm by decomposing the wavelength by 44 pixels. Since the signals measured by this infrared spectrometer contain data of both absorption by atmospheric components of infrared radiation from the sun and contributions of absorption by aerosols/PSCs, the latter must be either corrected or estimated simultaneously when deriving the concentrations of gaseous components from those signals. This process is of great significance not merely because it is necessary for the derivation of the concentrations of gaseous components but because it provides data on aerosols/PSCs.

Since algorithms to simultaneously extract data of both gaseous components and aerosols/PSCs from the absorption spectra in the infrared region had not yet been developed when the ILAS Project was initiated, the author decided to conduct a study on such algorithms. This study proceeded as follows: 1) conceptual study on algorithms; 2) validation of algorithms

by simulations; and 3) application of the algorithms to actual data.

2. Purpose and Methodology

The purpose of this study is to develop algorithms both for extracting data on aerosols /PSCs and for isolating the contributions of absorption by aerosols/PSCs from the measured signals, which are contained in the solar occultation sensor data, to improve the precision of the derivation of gaseous component concentrations. Such algorithms will be reflected on the operational software for processing data measured by ILAS, a satellite-borne ozone layer observation sensor developed by the Environment Agency. Since data actually measured by ILAS became available during the period of this study, the author conducted a feasibility study on the algorithms using the actual data, in addition to validation of the algorithms by simulations.

3. Simulation of a Method to Use Window Channels (Sequential Estimation Method) for Discriminating the Types of Aerosols/PSCs and Estimating the Parameters of Particle-Size Distribution

This chapter describes a method to simulate the sequential estimation method, which estimates the types of aerosols/PSCs and parameters of particle-size distribution using the extinction coefficients due to aerosols/PSCs obtained by window channels selected from the 44 pixels of the infrared spectrometer, and then calculates absorption spectra by all 44 pixels.

3.1 Selection of Window Channels

First of all, pixels, of which the influence of extinction (absorption) by aerosols/PSCs is predominant, were selected as window channels from all 44 pixels of the infrared spectrometer. Absorption spectra measured by ILAS are simulated at every representative tangential altitude using the ILAS reference atmospheric models created based on the past satellite data obtained so far. The maximum and minimum values of absorption spectra were evaluated using the 10 percentile and 90 percentile values of the concentrations of gases prepared as the reference atmospheric models. As a result of selecting pixels (region of wavelength), of which the absorption is small or of which the difference between the maximum and minimum values is small, those determined to be appropriate as window channels were 7.12 μm (the 7th pixel), 8.7 μm (the 16th pixel), 10.60 μm (the 34th pixel), and 11.76 μm (the 43rd pixel). This means that contributions by non-gaseous components can be evaluated using these channels with minimum errors regardless of the concentrations of gaseous components.

3.2 Simulation of Type Discrimination of Aerosols/PSCs and Estimation of Particle-Size Parameters

The simulation was conducted as follows: aerosol/PSC models were created with reference to documents and technical papers; their optical characteristics were calculated based on the theory of Mie scattering; then a feasibility study was conducted for the type identification and discrimination of aerosols/PSCs based on the wavelength dependency of the extinction coefficients obtained from the optical characteristics.

On the basis of documents and technical papers, sulfuric acid aerosols and various types of PSCs were used for creating the aerosol/PSC models. Regarding aerosols that were assumed to be sulfuric acid droplets, their optical characteristics (complex index of refraction) are subject to variations due to the percentage by weight of sulfuric acid contained. Regarding PSCs that were assumed to be nitric acid hydrates, NAT, NAD, NAM and ice (of water) were conjectured; however, the results of laboratory experiments showed that α-NAT, β-NAT and other amorphous substances (expressed with a-) may also exist as nitric acid hydrates

depending on the conditions of formation. The optical characteristics of these particles have been obtained as a complex index of refraction mainly by laboratory experiments.

Extinction coefficient spectra of these aerosols/PSCs were obtained by applying the theory of Mie scattering. To simplify the calculations, the logarithmic normal distribution was used as the particle-size distribution, and calculations were conducted for various combinations using values of the central radius and dispersion, which are generally found in the stratosphere. Table 1 shows the values used.

Table 1 Values of Particle-Size Distribution Parameters Used for the Aerosol/PSC Models

	rg (μ m)	σg
Sulfuric acid aerosols	0.075, 0.075±0.075/4 0.075±0.075/2 0.075±0.075*3/4	1.86, 1.86±0.1, 1.86±0.2
Ice	5, 10, 20, 30, 40, 50, 100	1.5, 1.5±0.1, 1.5±0.2
α-NAT, β-NAT, NAD, NAM, a-NAT, a-NAD, a-NAM	0.5, 0.5±0.5/4 0.5±0.5/2, 0.5±0.5/2	1.5, 1.5±0.1, 1.5±0.2

The extinction coefficient spectra of representative sulfuric acid aerosols and other PSCs were calculated for the four ILAS window channels. The shapes of spectra are found unique to the respective types of aerosols/PSCs. When the extinction coefficients of the four ILAS window channels are considered to be four-dimensional vectors, the vector directions can be expressed by normalizing the absolute values. This means that the influence of the total number of particles in the particle-size distribution of aerosols/PSCs is removed from the size of extinction coefficients. Here, only the vector directions are taken into account by normalizing the results of calculations based on the aerosol/PSC models. In principle, since the vector directions strongly depend upon the optical characteristics (complex index of refraction) of aerosols/PSCs, the difference in the particle-size distribution parameters results in dispersion.

Thus, types of aerosols/PSCs can roughly be discriminated by comparing the results of model calculations and the normalized vectors obtained from measured values. Specifically, those with the minimum distance between vectors can be determined to be the types of aerosols/PSCs in question. Temperature can also be added to the conditions of type discrimination, i.e., when the temperature is 215 K or over, PSCs do not exist in the stratosphere, but only sulfuric acid aerosols do.

3.3 Estimation of the Particle-Size Distribution Parameters of Aerosols/PSCs

Following the completion of type discrimination of aerosols/PSCs, an attempt was made to determine the detailed values of particle-size distribution parameters. Here, the mean radius (r_g) and the particle-size dispersion (σ_g) were considered to be unknown parameters, and values that can accurately adjust the wavelength dependency of extinction coefficients were sought.

The sizes of particles that are actually found in the stratosphere are usually submicron except for those of ice particles, and their scattering phenomenon can be described by Rayleigh's principle when the infrared wavelength measured by ILAS is considered. Since the extinction coefficient spectra of infrared wavelength are almost determined by the optical

characteristics (wavelength dependency of complex index of refraction) of particles, it is difficult to estimate two particle-size distribution parameters accurately and simultaneously. Since the extinction coefficients in the infrared wavelength region do not depend much on the average particle size, it is easier to obtain only the particle-size dispersion (σ_g) and the total number of particles (N_0) using an invariable value for the mean diameter (r_g).

Having determined the types of aerosols/PSCs and the particle-size distribution parameters, spectral interpolation calculations for all 44 pixels of the infrared spectrometer based on the theoretical calculations were performed.

4. Analysis of Measured Data Using Window Channels (Sequential Estimation Method)
4.1 Evaluation of the Distribution of Extinction Coefficients Using Four Window Channels

The precondition for using window channel data is that extinction coefficients can be obtained properly. First of all, the contributions of gaseous components contained in the window channel signals are estimated to remove from the total absorption, as mentioned in Chapter 3.1. Here, climatic values obtained from the reference model atmosphere created based on the satellite observational data acquired so far were used, i.e., mean average values per month and per latitude (at intervals of 5 degrees) and other statistics prepared based chiefly on the data obtained in 1992 and 1993 by the sensors installed on UARS.

The contributions of gaseous components contained in the pseudo-transmittance measured by ILAS were corrected by giving climatic values to the altitudinal distribution as the gaseous component concentrations. The remainder, which is due to non-gaseous components, was assumed to be due to aerosols/PSCs. The altitudinal distribution of extinction coefficients can be obtained by resolving the pseudo-transmittance (expressed by a function of tangential altitude obtained in this manner) by the onion peeling method. The errors of the extinction coefficients are determined by the difference between the climatic values and the actual distribution of atmospheric components, random errors of signals, analytical errors, and so forth. The main cause of errors is considered to be the difference between the climatic values and actual atmosphere in the distribution of gaseous components. In fact, when calculations are conducted by giving the 10 percentile and 90 percentile values to the distribution of gaseous component concentrations, resulting solutions are attributable to the difference between the climatic values and actual atmosphere.

Figure 1 shows an example of extinction coefficients obtained in this manner; three values at a tangential altitude of 17 km are presented for the respective window channels, which were obtained by giving values of 10 percentile, mean average, and 90 percentile to the distribution of gaseous component concentrations, respectively. According to this figure, the 16th and 43rd pixels are particularly prone to the influence of the distribution of gaseous component concentrations, while the 7th and 34th pixels are not dependent upon the concentrations of gaseous components. The three curves represent the extinction coefficient spectra in the case when background sulfuric acid aerosols of small particle size are assumed (results of theoretical calculations using three different complex indexes of refraction).

4.2 Estimation of Types of Aerosols and Particle-Size Distribution Parameters

Two cases were established for analyzing the data actually measured by ILAS to clarify the problem, i.e., 1) only sulfuric acid aerosols are taken into account as aerosols/PSCs, and 2) other PSCs must also be taken into account. The former is when the temperature is around 215 K or higher in principle, while the latter must rely on empirical knowledge when selecting the types of aerosols based on the status of signals because they cannot indiscriminately be determined by temperature.
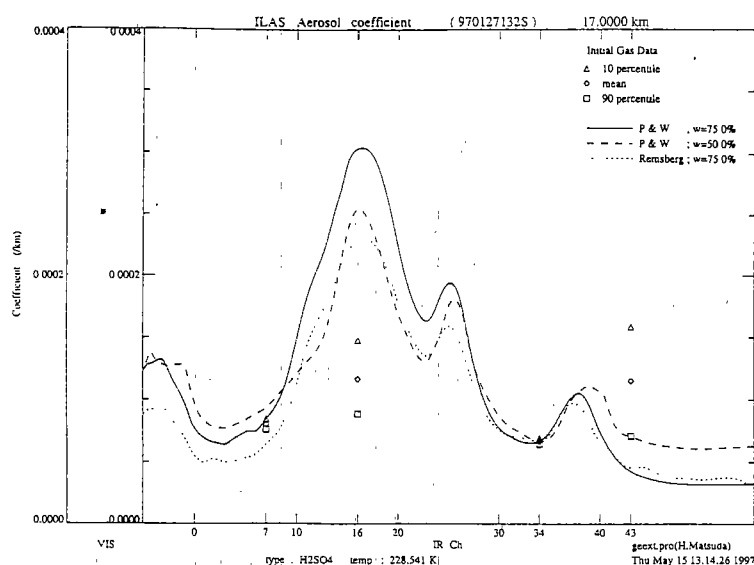
Figure 1 Values of the coefficients of extinction due to non-gaseous components (in the case of sulfuric acid aerosols) in the window channels obtained from the data measured by ILAS and the values obtained by theoretical calculations assuming background sulfuric acid aerosols

(1) In the Case of Sulfuric Acid Aerosols

Looking at the curve of extinction coefficient spectra of sulfuric acid aerosols obtained by model calculations shown in Figure 1, the curve hits the first peak of absorption near the 16th pixel and the second peak near the 26th pixel, but it resumes a comparatively flat shape for the rest of the wavelength regions. However, Figure 1 also shows that the values of extinction coefficient of both the 16th and 43rd pixels, which were obtained using measured data, are greater than those of other pixels. Although this phenomenon was observed in almost all the cases at an altitude of 20 km or lower when sulfuric acid aerosols were suspected, it can hardly be explained theoretically. This subject will not be addressed further in this paper.

Another issue requiring investigation is that the values of ratio of extinction coefficients between the 16th and 7th pixels obtained by measured data were far smaller than those obtained by model calculations. This cannot be explained unless the existence of particles that are far larger than those usually known as sulfuric acid aerosols is assumed. For reference, the extinction coefficient spectra (dotted lines) calculated assuming the existence of large particles are shown in Figure 2. In general, values obtained by assuming large particles are closer to the values obtained by measured data, except for the discrepancy of the 43rd pixel (see Figure 1).

Although the cause of this is not known, it is necessary to consider the possibility that some unknown offset component is mixed into the entire wavelength area. Possible causes of the offset of extinction coefficients may be : 1) 100 % value variations due to tracking mistakes; 2) influence of solar limb darkening; 3) influence of drift corrections; 4) influence of deconvolution; 5) influence of crosstalk; 6) influence of stray light; etc. The causes of this offset had not yet been identified at the time of writing this paper.

Irrespective of the causes, it was concluded that the method discussed in Chapter 3 was inapplicable without modification, since the contributions of non-gaseous components were assumed to be appearing in the form of offset in the data actually measured by ILAS. The chief mistake was to use the ratio of extinction coefficients between the 7th and 16th pixels, which corresponded favorably with the weight concentrations contained in aerosols when sulfuric acid was assumed, for estimating the weight concentrations.

According to conventional knowledge, the predominant particles of sulfuric acid aerosols

ILAS Aerosol Extinction Coefficient   970210347R   15.0000 km

NO,Offset
Large Rg

VIS   IR Ch   ncl_lrg.pro(H.Matsuda)
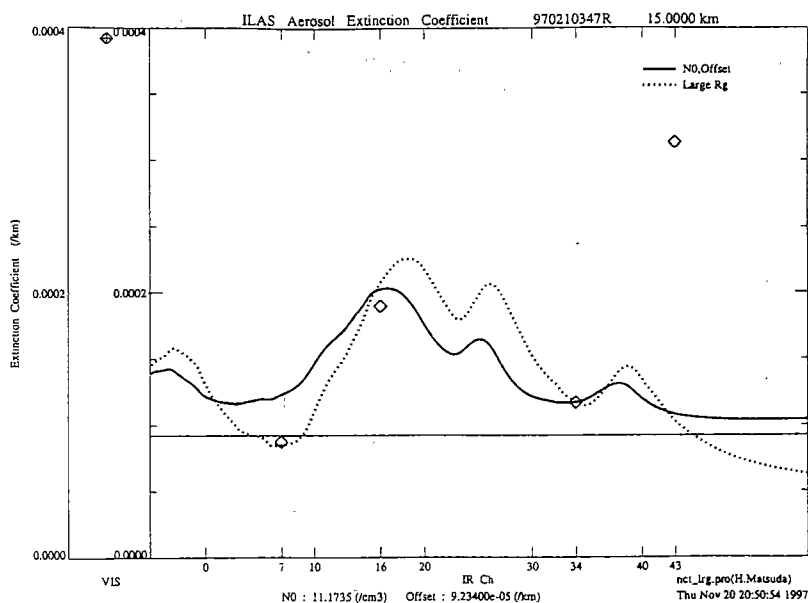NO : 11.1735 (/cm3)   Offset : 9.23400e-05 (/km)   Thu Nov 20 20:50:54 1997

Figure 2 Extinction spectra of sulfuric acid aerosols (the dotted line denotes large
particles, while the solid line denotes small particles + offset.)

have a mean radius of 0.1 μ m or smaller. Thus, an attempt was made to explain the measured values by the sum of offset that is independent of the wavelength and extinction coefficients that were obtained by assuming small-sized sulfuric acid aerosols with the average particle size of r_g = 0.075 μ m, particle size spread of σ_g = 1.86, and weight concentrations of sulfuric acid of 75 %, considering the particle-size distribution function to be a logarithmic normal distribution. Using also the values of extinction coefficients at the 780 nm data obtained by the visible spectrometer, the number of particles of small-sized aerosols and the contributions of offset were calculated for two combinations, i.e., visible data + 16th pixel, or visible data + 34th pixel. Since the extinction coefficient of the 34th pixel is hardly prone to the influence of gaseous components, values that appear to be comparatively reliable can be obtained (the thick solid line and the thin solid line in Figure 2).

Theoretically speaking, it is possible to uniquely determine the inclination of the offset components against wavelength by the combination of visible data + 16th pixel + 34th pixel, if the offset components are regarded as a linear function of wavelength. At any rate, the estimation precision of these methods depend on the reliability of the extinction coefficients of the respective pixels.

(2) In the Case of PSCs

Although values of weight concentrations of sulfuric acid aerosols can, in principle, be estimated by the ratio of extinction coefficients between the 7th and 16th pixels, theoretical values based on models have shown a difference in the values for other PSCs. The ratio of extinction coefficients between the 7th and 16th pixels of the measured data of ILAS also shows some characteristics of PSCs that are different from those of sulfuric acid. Nevertheless, it is still difficult to separate the PSC components assuming the existence of offset in the same manner as in the previous section, attributable to the difficulty of estimating the representative types of PSCs in advance due to the numerous possible types of PSCs.

(3) Estimation of Spectra by the Simple Interpolation Method and Correction of Contributions of Non-Gaseous Components

Here, the sum total of contributions of aerosols/PSCs and the offset components are

called "contributions of non-gaseous components." When the contributions (spectra) of non-gaseous components which correspond to all the pixels of the infrared spectrometer are estimated, those of small-sized sulfuric acid aerosols and offset can respectively be obtained if the components can apparently be determined to be sulfuric acid aerosols, as discussed in the previous section. Although temperature is the criterion for the determination in general, problems arise such as difficulty in conducting determination when the temperature is near the criterion, and deciding the determination method when the temperature is below the criterion. Thus, an alternative method that can handle the routine processing of a huge volume of data was adopted in the Version 3.10 data processing and operation software as follows:

The values of extinction coefficients of the four window channels (the 7th, 16th, 34th and 43rd pixels) obtained in Chapter 4.1 were linearly interpolated with respect to wavelength, then they were regarded as the extinction coefficients of the pixels between the 7th and the 43rd pixels, respectively. The extinction coefficients of the pixels in the area of wavelength that is shorter than the 7th pixel (with smaller pixel numbers) were considered to be equivalent to that of the 7th pixel, while those of the pixels immediately outside the 0th and 43rd pixels, which are necessary for calculating the contributions of crosstalk, were considered to be equivalent to those of the 0th and 43rd pixels, respectively. These extinction coefficients were calculated for the respective altitudes to obtain their altitudinal distribution of all the pixels. Figure 3 shows the spectra estimated assuming the components to be small-sized sulfuric acid aerosols and offset, as well as the linearly interpolated spectra proposed in this section. The difference between the estimated and linearly interpolated spectra is comparatively small in the case of sulfuric acid aerosols except for the pixels near the 43rd pixel, since the window channels draw near the peaks of absorption spectra by coincidence.
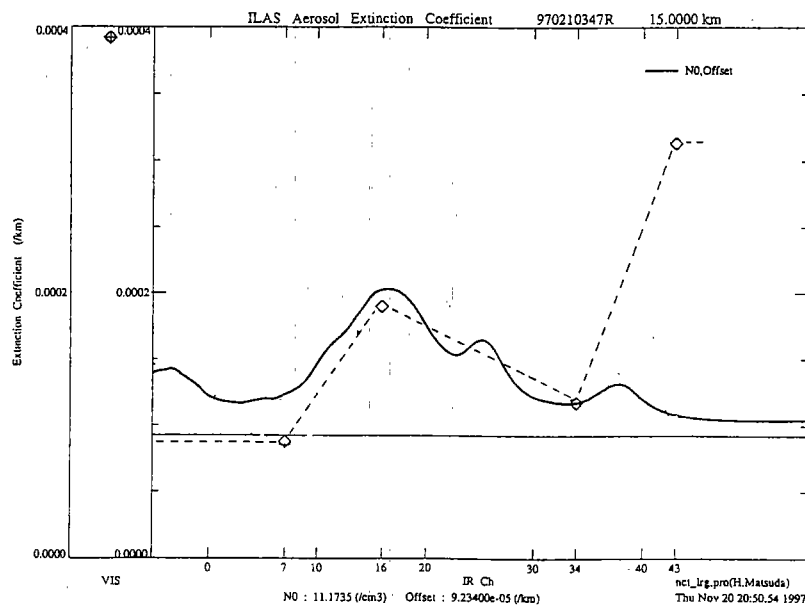

Figure 3 Contributions of the representative stratospheric aerosols (sulfuric acid) and the offset components evaluated by assuming a composite of those two component, and an example of spectra obtained by linear interpolation (dotted curve)

Using the altitudinal distribution of the contributions (extinction coefficients) of non-gaseous components obtained in this manner, the altitudinal distribution of gaseous component concentrations was obtained using pseudo-transmittance only of gaseous components, which was created by correcting the pseudo-transmittance of infrared 44 pixels in advance. Figure 4 (left) shows the altitudinal distribution of methane components obtained by correcting the

contributions of non-gaseous components (Version 3.10), in which the components were apparently determined to be sulfuric acid aerosols, as well as the altitudinal distribution of methane components without any corrections of non-gaseous components (Version 3.00). Gaseous concentrations are overestimated unless the contributions of non-gaseous components are corrected, since the entire contributions are deemed to be attributable to gaseous components.

A comparison between the spectra of PSCs and the spectra obtained by interpolating the window channels reveals that there are considerable discrepancies between some of these spectra. Despite this limitation, however, linear interpolation is a practical method, since it is not easy to specify the type of PSCs to evaluate the contributions of PSCs and offset, when the components are assumed to be PSCs. Figure 4 (right) shows an example of altitudinal distribution of corrected methane components, when the PSCs were assumed.
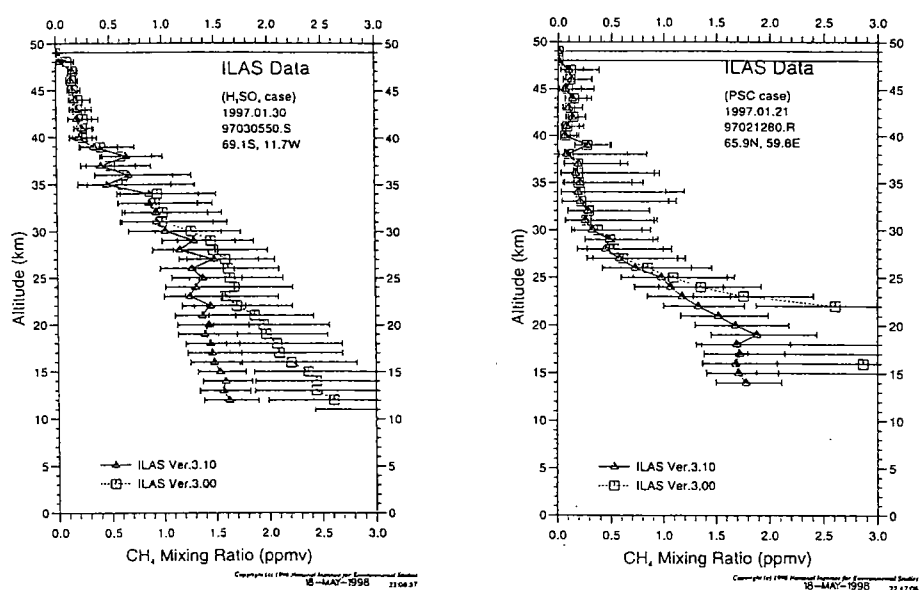

Figure 4 Distribution of methane concentrations corrected by aerosols/PSCs (Version 3.10) in the case of sulfuric acid aerosols (left) and in the case of PSCs (right). The methane distribution without corrections (Version 3.00) is also shown in the figure.

5. Simultaneous Estimation of Gaseous and Aerosol Components

A method to simultaneously estimate the gaseous components, aerosol/PSC components and the contributions of offset, using the data of all the channels of the ILAS infrared spectrometer, is proposed in this chapter. This method does not require climatic values.

5.1 Spectral Linear Combination Method

When the concentrations of gaseous components are calculated from the data obtained by the infrared spectrometer of ILAS, several parameters (concentrations of gaseous species) are estimated at the same time using the non-linear least squares method. Here, calculations are conducted by considering that the spectra of absorption cross section, which are unique to the respective gases, are known as a function of temperature and pressure, and that gaseous concentrations are unknown. Spectra of extinction (absorption) due to aerosols/PSCs can also be calculated in advance, if their types and optical characteristics are assumed to be known. Therefore, it is possible to conduct simultaneous estimation in the same manner as the calculations of gaseous components, provided that the number densities of the respective

aerosols/PSCs are assumed to be unknown. The same applies to the offset contributions (spectra are assumed to be flat). When this method is used, models of aerosols/PSCs must be created to obtain representative types of aerosols and representative shapes of their respective particle-size distributions. In this study, models of aerosols/PSCs were created using the parameters shown in Table 2.

Table 2 Models of Aerosols/PSCs

	Composition	rg	σg
Sulfuric acid aerosols	75% H2SO4	0.075	1.86
Ice	H2O	10.0	1.5
Nitric acid	22.3% HNO3	0.5	1.5
β-NAT		0.5	1.5
Offset	-	-	-

An attempt was made to simultaneously estimate the contributions of gaseous components and aerosols/PSCs, using data actually measured by ILAS. Figure 5 shows the altitudinal distribution of the concentrations of methane components, which were obtained by applying the method to the case where only sulfuric acid aerosols were assumed to facilitate the interpretations.
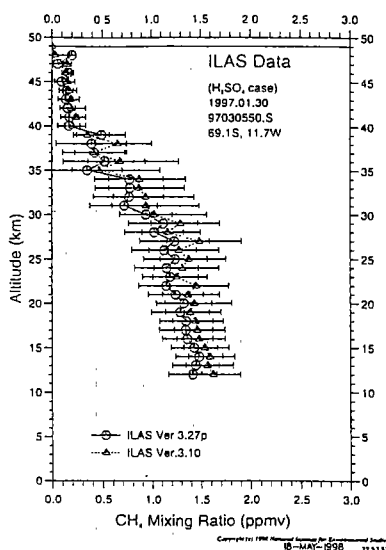

Figure 5 Distribution of methane concentrations simultaneously estimated by the linear combination method of the representative spectra of aerosols/PSCs (In the case of sulfuric acid aerosols: Version 3.27p) The methane distribution obtained by Version 3.10 is also shown in the figure.

This method was also applied to the case where PSCs were assumed. Although it is theoretically possible to consider all PSCs to be unknown parameters, this is not practical in terms of the stability and reliability of solutions. The selection of the target PSCs to be resolved is arbitrary. Considering also that the PSC models themselves involve uncertainty and the cause of offset is still unidentified, this method requires further improvements.

6. Other Methods
    Dubovik proposed two methods. One is to approximate the spectra of extinction due to

aerosols/PSCs by polygonal lines, and to conduct estimation by assuming the parameters that express the polygonal lines to be unknown values. Another is to approximate the particle-size distribution of aerosols/PSCs also by polygonal lines, and to conduct estimation assuming the parameters that express the polygonal lines to be unknown values. In the case of the latter, the types of aerosols/PSCs must be identified in advance.

In the case of the former, however, it is not necessary to know the types of aerosols /PSCs in advance. In addition, this method can be regarded as a modified version of the method proposed in Chapter 5. Specifically, triangular spectra at appropriate pixel intervals are prepared instead of the extinction coefficient spectra of the respective aerosols/PSCs modeled in Chapter 5; it is considered that unknown extinction coefficient spectra of aerosols/PSCs are expressed as a linear combination of these triangular spectra; then, simultaneous estimation is conducted by the same method as in the previous chapter, since the coefficient of the linear combination can be assumed to be an unknown value. The results of simple simulation analysis showed that both of these methods were applicable theoretically, but more detailed analysis and further improvements are required.

7. Conclusion

The first method proposed in this study was to conduct sequential analysis by using the window channel data of the ILAS infrared spectrometer, i.e., recognition of the types of aerosols, estimation of particle-size distribution parameters, and spectral interpolation by theoretical calculations. Although the results of simulation showed that this method was sufficiently workable, the existence of offset, which was estimated from the data actually measured by ILAS, required modification of this method for more realistic calculations (estimation of spectra by the linear interpolation method). Since the ILAS data processing and operational software (Version 3.10) employs this spectral estimation method based on the linear interpolation method, the distribution of gaseous component concentrations obtained using this version was reasonably adequate except for some discrepancies due to some other problems.

The second method was to simultaneously estimate the contributions of gaseous components and aerosols/PSCs using the data of all channels of the infrared spectrometer. Since offset must also be taken into account, calculations were conducted by assuming the offset components to be unknown values. Since it is extremely difficult to obtain direct validation data in the case of aerosols/PSCs, particularly the extinction coefficients in the infrared wavelength region, types of PSCs, and concentrations, it is hard to estimate the validity of the derived results. Further research to establish an effective validation method is required.

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