Investigation of the optical properties of stratospheric aerosols in the UV-visible range

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Abstract

In the frame of the GOMOS experiment, a model named ECSTRA (Extinction Coefficient for STRatospheric Aerosol) has been developed for describing the spectral and vertical parameters of the stratospheric aerosol extinction profile. The model is based on an analytical formulation of the extinction coefficient. The fit parameters can be related to the particle distribution (spectral dependence of the extinction) and to the parameters characterizing the vertical structures (e.g. the Junge layer). The behaviour of the fit parameters is discussed and a comparison is proposed between ECSTRA and other existing models.

1 Introduction

Stratospheric aerosols are known to have an important impact on the Earth's radiation budjet. They also play an important rule in the ozon related chemistry. Although antropogenic sources of aerosol have been identified [8], volcanism is accepted to be the most important source of aerosol injection into the stratosphere [12]. During major eruptions, the aerosol loading of the stratosphere can dramatically increase, leading to a possible increase of the optical thickness of several orders of magnitude. Therefore, volcanism appears to be an essential parameter for the characterization of the stratospheric aerosol mass loading.

In order to test algorithms developed in the context of the GOMOS experiment, we have built a climatological model called Extinction Coefficient for STRatospheric Aerosols (ECSTRA) [6] based on a simple analytical formulation of both spectral and vertical dependences of the aerosol extinction profile. The aim of the project was to describe the aerosol extinction profile on a realistic way, for a wide range of volcanic conditions and latitude coverage.

2 Basic satellite data

As basic data source, we used the aerosol extinction profiles supplied by the Stratospheric Aerosol and Gas Experiment II (SAGE II) which has been extensively described in the literature [2], [3]. A major advantage of this experiment is to supply extinction profiles on a near global scale ($80°S$ to $80°N$) and for a wide range of volcanic status. More particularly, SAGE II covers a period of quite low volcanism around 1989, several volcanic eruptions of middle importance (Nevado del Ruiz in November 1985, Kelut in Februari 1990), a part of the decay period following the major eruption of El Chichon (November 1992), and the cataclysmic eruption of the Pinatubo in June 1991. The data set we used covers the time period from October 1984 to December 1995.

In order to enhance the statistical significance of the data, a standard binning procedure [7] has been used, using intervals of one month in time and 10° in latitude. This processing gave rise, for each bin, to four mean reference extinction profiles covering the altitude range 0 to 50 km with a resolution of $1km$, and corresponding to the four SAGE II nominal channels at $\lambda = 1.020\mu m$, $0.525\mu m$, $0.453\mu m$ and $0.385\mu m$.

3 Analytical formulation

The aerosol optical properties are described by the extinction coefficient

$$\beta(z;\lambda) = \int_0^{+\infty} n(z)\ f(r)\ Q(r;\lambda)\ dr \qquad (1)$$

where $n(z)$ is the particle density, $f(r)$ is the particle size distribution and $Q(r;\lambda)$ is the particle cross section. $\beta(z;\lambda)$ is related to the optical thickness δ by

$$\delta(\lambda) = \int_{z_t+2\ km}^{+\infty} \beta(z,\lambda)dz \qquad (2)$$

The problem we are concerned with is to modelize the extinction profile $\beta(z;\lambda)$ as a function of the wavelength λ, the latitude φ, the altitude z and a parameter V representing the volcanic status of the atmosphere.

Concerning the dependence in z, the extinction profile is found to present a layered structure which can be roughly considered as parallel to the

tropopause level z_t [1]. Therefore, we chosed to use a reduced altitude z_r defined as

$$z_r = z - (z_t + 2\ km) \tag{3}$$

where the origin has been chosen $2km$ above the tropopause level in order to avoid clouds contamination.

Since the optical thickness δ shows a near exponential decay after a volcanic eruption, we found that the logarithm of $\delta(1.020\mu m))$ is a good measure of the volcanic status of the atmosphere. Therefore, we defined the volcanism parameter V as :

$$V = \ln(1 + \frac{\bar{\delta}}{\delta_0}) \tag{4}$$

with

$$\delta_0 = 10^{-4} \tag{5}$$

The mean optical thickness $\bar{\delta}$ was preferred to δ in order to smooth out local variations of the optical thickness, and to dispose on a value representative for the whole atmosphere at a given time. $\bar{\delta}$ was obtained by averaging δ over the whole latitudinal and longitudinal range, using a two-month averaging window.

An examination of the extinction profiles shows that the spectral dependence varies slowly with respect to the altitude, compared to the vertical dependence of the vertical structures of the profile. Therefore, we used a formal separation between the spectral and vertical dependences $\beta_a(\lambda)$ and $\beta_b(z)$ of the extinction coefficient:

$$\beta(z;\lambda) = \beta_a(\lambda) \cdot \beta_b(z) \tag{6}$$

The vertical part $\beta_b(z)$ is reffered to as the extinction coefficient at $\lambda = 1.020\mu m$.

3.1 Spectral dependence

In order to modelize the spectral dependence $\beta_a(\lambda)$, we used the formulation proposed by Yue [13]

$$\beta_a(\lambda) = \exp\left(a_1\Delta\lambda + a_2\Delta\lambda^2\right) \tag{7}$$

where

$$\Delta\lambda = \lambda - 1.020\mu m \tag{8}$$

This formulation allows to describe typical 'background' situations characterized by a monotonic decreasing spectral dependence of $\beta_a(\lambda)$, and volcanic situations for which the spectral dependence presents an extremum, as well.

The a_i dependence in the basic parameters was modelized as

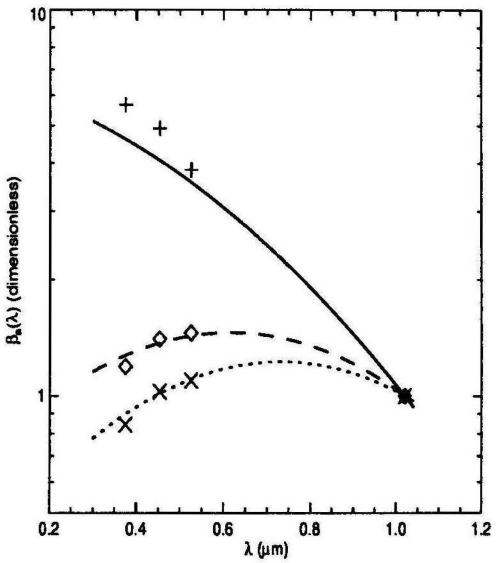

Figure 1: Spectral dependence of $\beta_a(\lambda)$ at $18km$ and $\varphi = 45°N$. The lines represent the ECSTRA simulation, and the symbols correspond to representated SAGE II events. (a) June 1991; $V = 2.7$ (representative for a situation before the Pinatubo volcanic cloud contamination); solid, +; (b) June 1992; $V = 6.7$; dotted, ×; (c) May 1998; $V = 5.4$; dashed, ◇.

$$a_i = \sum_{j=0}^{2}\sum_{k=0}^{2}\sum_{l=0}^{3} \alpha_{ijkl} \cdot P_{2j}(\theta) \cdot V^k \cdot {z_r}^l \tag{9}$$

where the colatitude θ is related to φ as $\theta = \frac{\pi}{2} - \varphi$, and P_{2j} are the first symetric Legendre polynomials. The a_i coefficients were determined from the SAGE II binned data by a linear least-square fit procedure with 9.

As an example, figure 1 illustrates the spectral dependence of $\beta_a(\lambda)$ at $18km$ absolute altitude for various volcanic situations.

Before the eruption of the PINATUBO, May 1991 is characterized by a low volcanic situation, close to the 'background' situation. In June 1992, the aerosol mass load is important due to the Pinatubo eruption, one year before. Different studies from ***in situ*** experiments [4], [5], [9] and satellite observations as well [2], [10], [11] confirm the presence of important quantities of large particles in the stratosphere. As a consequence, the spectral depedendence is found to be very weak, as seen in the figure. In May 1993, due to progressive unloading of the stratosphere by sedimentation mechanisms, the quantity of large particles decreases in the stratosphere, and the spectral behaviour of β_a tends to a background condition again. Figure 1 is found to be in good agreement with figure 2 of [2], related to similar temporal peri-

Table 1: Mean value of $\beta(z,\varphi)$ at $\lambda = 1.020\mu m$ for the time period October 1984 up to December 1990, every $2km$ from $z = 8km$ to $34km$ altitude.

$80°S$	$70°S$	$60°S$	$50°S$	$40°S$	$30°S$	$20°S$	$10°S$	$0°$	$10°N$	$20°N$	$30°N$	$40°N$	$50°N$	$60°N$	$70°N$
.03	.03	.02	.02	.03	.05	.07	.07	.08	.07	.06	.04	.03	.02	.02	.03
.04	.04	.03	.03	.05	.09	.12	.13	.13	.12	.10	.06	.03	.03	.03	.03
.05	.05	.04	.05	.09	.19	.26	.28	.28	.26	.21	.10	.05	.04	.04	.04
.08	.08	.07	.09	.21	.48	.63	.66	.65	.62	.50	.23	.10	.08	.08	.07
.14	.16	.15	.22	.51	1.10	1.39	1.46	1.42	1.37	1.15	.55	.24	.17	.16	.13
.29	.34	.35	.53	1.17	2.22	2.66	2.77	2.72	2.63	2.29	1.22	.56	.42	.36	.28
.60	.73	.79	1.17	2.29	3.72	4.24	4.42	4.37	4.25	3.83	2.34	1.24	.95	.81	.60
1.19	1.46	1.62	2.27	3.75	5.11	5.56	5.79	5.81	5.67	5.28	3.79	2.35	1.90	1.64	1.23
2.17	2.59	2.88	3.69	5.04	5.72	5.98	6.27	6.39	6.24	5.95	5.07	3.74	3.24	2.89	2.26
3.51	4.00	4.34	5.00	5.57	5.58	7.84	12.39	14.72	10.65	6.37	5.61	4.97	4.64	4.34	3.65
4.99	5.34	5.49	5.62	5.55	9.47	29.06	71.44	97.18	66.18	21.44	6.06	5.52	5.50	5.49	5.08
9.31	13.32	7.14	6.15	7.00	18.47	47.22	95.60	126.07	99.36	48.60	10.99	5.78	5.96	7.40	6.38
74.60	27.38	11.30	7.46	8.72	13.88	23.17	36.15	44.61	39.16	29.65	17.64	7.33	7.16	11.36	26.95
50.84	19.50	10.19	6.80	6.06	4.64	5.72	7.86	9.38	8.49	7.83	12.59	8.54	7.34	9.99	24.01

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3.2 Vertical dependence

The extinction profile at $1.020\mu m$ can be reasonably modelized by

$$\begin{aligned} \beta_b(z_r) &= \beta_{aero}(z_r) + \beta_{cloud}(z_r) \qquad z_r < 0 \\ &= \beta_{aero}(z_r) \qquad z_r \geq 0 \end{aligned} \tag{10}$$

In this expression, $\beta_{aero}(z_r)$ and β_{cloud} represents respectively the aerosol and the clouds contributions to the vertical extinction. The latter is expected to vanish above the tropopause level.

$\beta_{aero}(z_r)$ is characterized by a peak situated a few kilometers above the tropopause and corresponding to the Junge layer, a slope break around $30km$ and an exponential decay at higher altitude. Those structures can be described by the analytical formula

$$\beta_{aero}(z_r) = \exp(x_1 + x_2\ z_r) + \exp\left(x_3 - \left(\frac{z_r - x_4}{x_5}\right)^2\right) \tag{11}$$

where the 2 terms represent respectively the asymptotic decay at high altitude and the aerosol peak of amplitude $\exp(x_3)$ (km^{-1}), width x_5 (km), centered at x_4 (km). The first term of (11) is found to be independent of the volcanism. Therefore, it corresponds to some minimal estimation of the volcanism value which is estimated from (2), (4) to

$$V_{min} = \ln\left(1 - \frac{e^{x_1}}{\delta_0 \cdot x_2}\right) \tag{12}$$

assuming that δ does not differ too much from $\bar{\delta}$.

The clouds contribution β_{cloud} is modelized as

$$\beta_{cloud}(z_r) = \exp\left(x_6 + x_7\ z_r + x_8\ {z_r}^2 + x_9\ {z_r}^3\right) \tag{13}$$

It must be pointed out that β_{cloud} is only intended to described a mean cloud coverage representative for the considered bin, which can significantly differ from the clouds coverage related to an isolated event. Nevertheless, it is important to take this term into account, in order to allow the slope at high altitude to be freely determined.

In order to fit the x_i coefficients by means of the binned data, the following transformation was used:

$$V_{\min} = u_1 \tag{14}$$

$$-\frac{1}{x_2} = u_2 \tag{15}$$

$$\frac{x_4}{x_5} = u_{45} + v_{45}\Delta V^2 + w_{45}\Delta V^3 \tag{16}$$

$$\frac{1}{x_5} = u_5 + v_5\Delta V^2 \tag{17}$$

$$x_6 = u_6 \tag{18}$$

$$x_7 = u_7 \tag{19}$$

$$x_8 = u_8 \tag{20}$$

$$x_9 = u_9 \tag{21}$$

A Levenberg-Marcquardt procedure was used for the fit of $V_{\min}$, $-1/x_2$, x_4/x_5, $1/x_5$, $x_6 \cdots x_9$.

Figure 2 shows the behaviour of the peak parameters x_3, x_4, x_5 as a function of the volcanism. It can be pointed out that the position of the Junge layer decreases with the volcanism and reaches a minimum value around $V = 4.5$. The peak width is found to be little dependent on volcanism.

In order to check the validity of ECSTRA, a mean value of $\beta(\lambda = 1.020\mu m)$ was derived for the binned SAGE II data and for the ECSTRA model from October 1984 up to December 1990. The result is reported in table 1. This was compared to the decadal mean values obtained by Hitchman *et al.* [7]. Notice that in the latest case, the authors use data from Februari 1979 to November 1981 and from October 1984 to December 1990. Since the first of those time periods is known as showing a very low volcanic level,

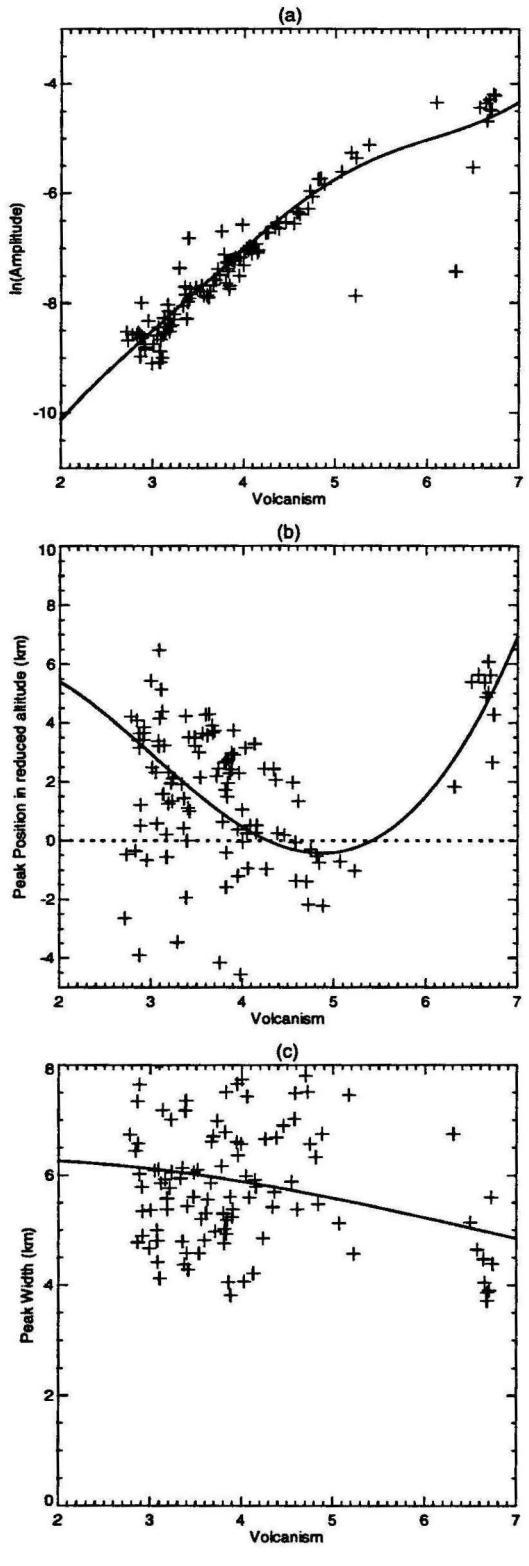

Figure 2: Dependence on volcanism V of the peak parameters (a) x_3, (b) x_4, (c) x_5 derived by the Levenberg-Marcquardt procedure applied to the binned SAGE II profiles (+), and by ECSTRA (solid line). $\varphi = 40°N$.

it is expected that the mean value found by ECSTRA is higher than Hitchman's one.

3.3 Total extinction

As an example of ECSTRA simulation, figure 3 illustrates the evolution of the total aerosol extinction profile $\beta(z;\lambda)$ at $\lambda = 0.453\mu m$. The binned SAGE II profile is compared to the ECSTRA model by means of an error parameter defined as

$$\varepsilon = \frac{\log(\beta_{SAGE}) - \log(\beta_{ECSTRA})}{\log(\beta_{SAGE})} \qquad (22)$$

It can be seen that the ECSTRA modelization is in agreement with the basic data set, excepted during transient periods following volcanic eruptions. Seasonal effects are also found to be not correctly described. This is due to the choice of characterizing the volcanism by means of a global parameter, which cannot reflect local details of the volcanic satus of the stratosphere. Also seasonal effects are found not to be correctly described, due to the fact that transport mechanisms have not been taken into account.

4 Conclusions and perspectives

The ECSTRA model has been found to be a simple tool allowing to describe the aerosol extinction coefficient as a function of the wavelength, latitude, altitude, and volcanic status of the stratosphere. Comparisons of the ECSTRA simulated extinction profiles with the basic data set SAGE II on the one hand, and with other models available in the litterature on the other hand, show a good agreement between the considered profiles, except during transient periods. Due to the basic conception of ECSTRA, this model is also unable to reproduce seasonal effects on a reliable way. Nevertheless, it allows to simulate the behaviour of the characteristic vertical structures of the aerosol profile, and to study the influence of volcanism on those parameters. On the other hand, the modelization of the spectral content of the extinction profile offers perspectives in order to modelize the particle distribution characteristics (mean particle size, dispersion of the particle distribution) on a global scale.

5 acknowledgments

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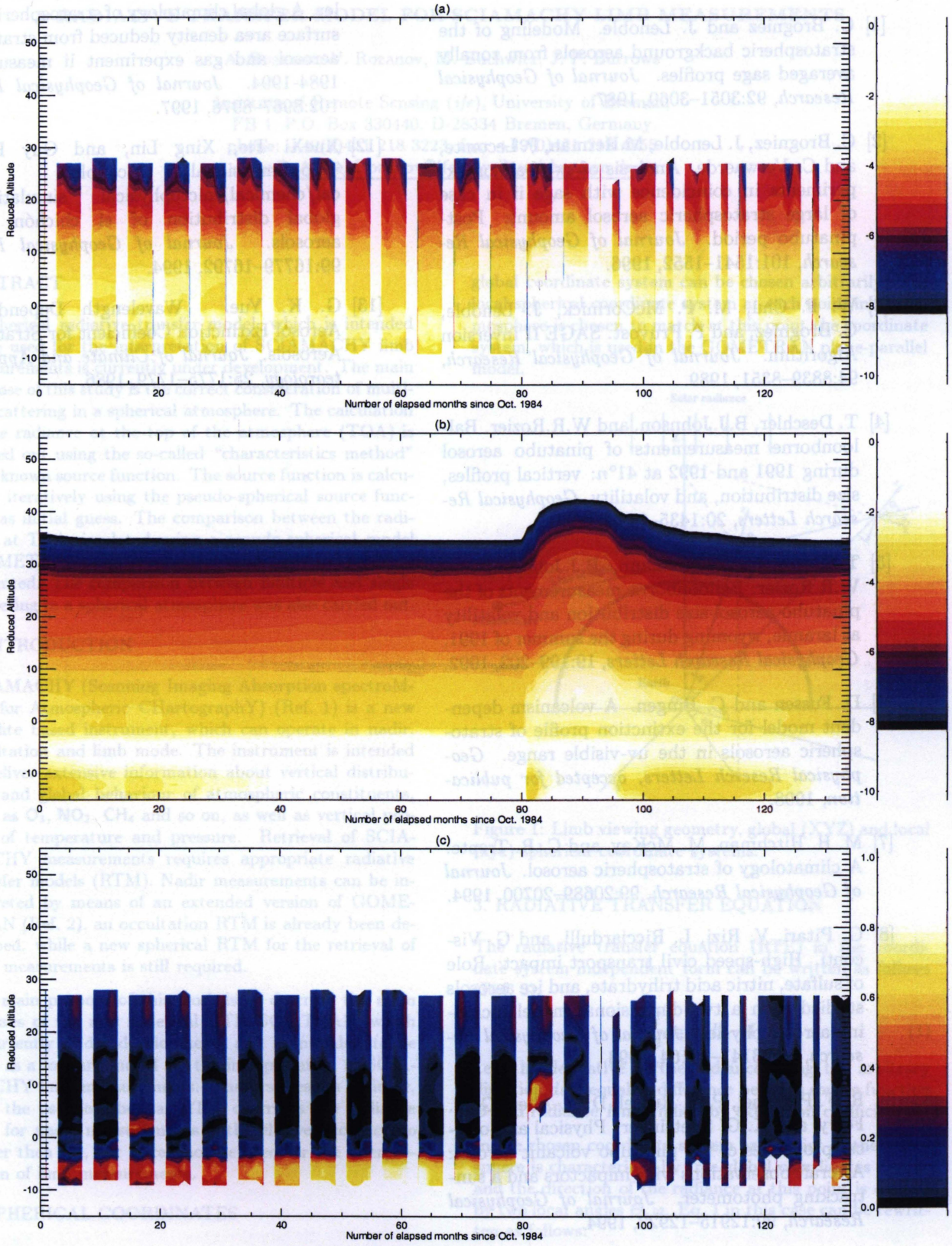

Figure 3: Aerosol extinction profile as a function of time and altitude at $\varphi = 0°$ and $\lambda = 0.453\mu m$; (a) binned SAGE II data; (b) ECSTRA simulation; (c) error parameter ε.

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