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CONTINUOUS LIDAR MEASUREMENTS OF STRATOSPHERIC AEROSOLS AND OZONE AFTER THE PINATUBO ERUPTION PART II: TIME EVOLUTION OF OZONE PROFILES AND OF AEROSOL PROPERTIES

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Abstract. Two lidar systems, an aerosol lidar and an O_3 DIfferential Absorption Lidar (DIAL), have been routinely operated at the same site (L'Aquila, Italy; 42°N, 13°E) since August 1991. The multiwavelength analysis of the lidar signals allows to retrieve parameters related to equivalent aerosol size distributions and their optical properties. These are needed to correct the ozone DIAL profiles from the disturbance introduced by the stratospheric volcanic aerosols. The method and the confidence of the retrieved ozone profiles are discussed in a companion paper. Here we present the whole measurement series of ozone and backscattering ratio profiles during the period from August 1991 to December 1992. In addition, for some observations, the mode radius and the dispersion of the representative aerosol size distribution are reported. The time evolutions of aerosol surface area density and mass mixing ratio are also discussed within the uncertainties of the retrieval algorithm.

Introduction

It has been estimated that Mount Pinatubo eruptions in June 1991 injected about 20Mton of SO_2 into the stratosphere (Bluth et al., 1992). The fast gas to particle conversion following a major eruption produces an increase in stratospheric aerosol number density, and these particles have dimensions larger than those usually observed long time after or before the volcanic eruptions (Jäger and Hofmann, 1991). The increase in the aerosol loading perturbs the radiative balance of the Earth-Atmosphere system (Minnis et al., 1993) and may affect, through heterogeneous chemical reactions, the ozone photochemistry. Particle size distribution and aerosol composition are important factors for evaluating such effects.

In a companion paper (D'Altorio et al., 1993), referred as PART I thereafter, we have shown that an algorithm can be developed to retrieve reliable ozone profiles from DIAL measurements even in presence of disturbance introduced by stratospheric aerosol layers. As by product of the same procedure, aerosol parameters can also be estimated, so that aerosol lidar observations provide the time evolution of such quantities as optical thickness, surface area density, mass and aerosol number density. In the following paragraphs the method adopted to retrieve aerosols characteristics is briefly outlined, then the resulting data are discussed together with the evolution of ozone number density. The observations cover a time period starting two months after the Pinatubo eruption to December 1992.

Backscattering ratio and ozone profiles

The 589nm backscattering ratios observed at SLAQ since August 1991 are shown in Fig.1. They are calculated according to the procedure outlined in PART I. The time behavior shows that volcanic aerosols were detected at our site in early August 1991 with relatively small values for the backscattering ratios. Roughly three months after the eruption, strong and high altitude layers were observed probably as results of transport from the tropics to higher latitudes, as also observed by SAGE II (McCormick and Veiga, 1992). Then, since January 1992, after a period in which sporadic and intense aerosol layers often appeared, the backscattering ratio profiles show a decreasing trend with the maximum slowly declining to lower altitudes, possibly due to aerosol sedimentation.

Fig.2 shows the retrieved ozone profiles as a function of time after the eruption. Within the particle layer the ozone density is locally affected by an indetermination between ±15% and ±35%. The behavior of the ozone density seems to reflect the seasonal variations with the density increasing during the Winter months and the maximum moving at lower altitude. Data from previous years are not available at our site and the seasonal change may be considered only in qualitative terms. The

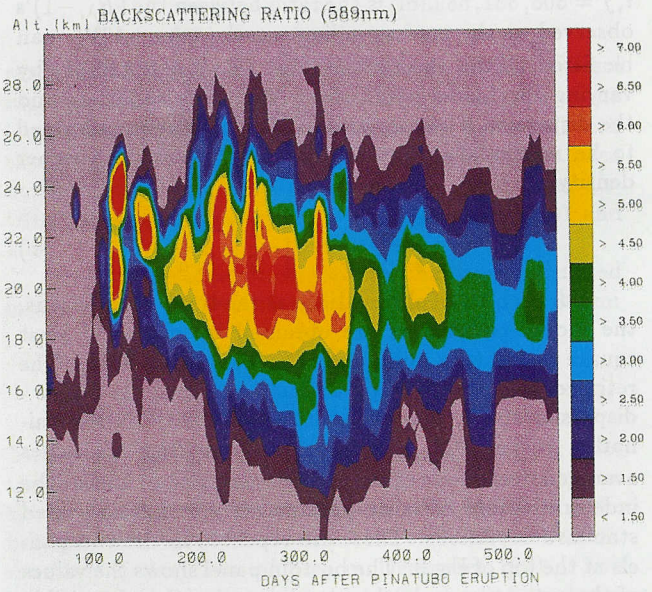

Fig.1. Time evolution of the backscattering ratio profiles at 589nm wavelength. The time scale starts 50 days after the Pinatubo eruption (early August 1991) and stops on December 1992; on the ordinate the altitudes range from 10 to 30km. The grid has been obtained with a resolution of 5 days and 0.5km respectively for the time after the eruption and the altitudes.

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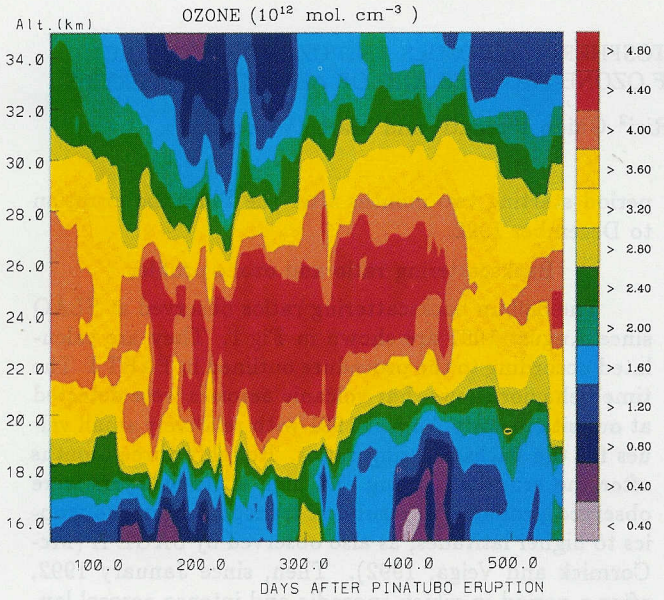

Fig.2. Time evolution of the ozone profiles in 15-35km altitude range from August 1991 to December 1992. The grid has the same resolutions of Fig.1.

indetermination in retrieved ozone and the limited length of the record prevent from any attempt to assess the local effects of heterogeneous chemistry. It is likely that the column density may be a better indicator of the change related to injection of sulfate aerosols (see PART I).

Aerosol size distributions and related quantities

Referring to PART I we recall that $A^{obs}_{\lambda_i,\lambda_j}$ ($i \neq j$ and $i,j = 308, 351, 589$nm) is the ratio between the $(R_{\lambda_i} - 1)$'s observed at the corresponding wavelengths. $A^{obs}_{\lambda_i,\lambda_j}$ can be fitted to the theoretical value (Eq.2 of PART I) by varying the model parameters: the mode radius r_o and the dispersion σ of the aerosol size distribution, assumed to be unimodal lognormal for unitary particle number density,

$$n(r) = \frac{1}{\sqrt{2\pi} r \ln\sigma} exp\left[-\frac{1}{2}\left(\frac{ln\frac{r}{r_o}}{ln\,\sigma}\right)^2\right] \qquad (1)$$

For each set of observations at the three wavelengths, the model parameters are obtained by means of least square fitting (the minimum of χ, Eq.3 in PART I). The retrieved r_o and σ occupy a region in the mode radius - dispersion space, whose amplitude reflects the indetermination on the $A^{obs}_{\lambda_i,\lambda_j}$ coefficients. We assume the values averaged over such region as representative r_o and σ; the indetermination of these parameters are the associated standard deviations. These are reported in the two panels at the top of Fig.3. The bottom panel shows the values of the extinction to backscattering ratio (C_{λ_i}) for visible (589nm) and DIAL wavelengths (308, 351nm). Although the precision of r_o and σ is very low, the C_{λ_i} coefficients are retrieved with a reasonable indetermination for the purposes of ozone DIAL retrieving.

In any case it has to be considered that r_o and σ have the meaning of representative parameters of an equivalent particle size distribution (see PART I). On the other hand, backscattering measurements can only give informations about the particle radius in a size range where the ratio between the Mie backscattering cross sections at different wavelengths is not a constant (Larsen, 1992). For our observations at 308, 351 and 589nm no size informations are obtained for particles smaller than 0.05μm and larger than 0.6μm.

Our results appear to be consistent with similar data obtained in other mid-latitude observations. As an example, Ferrare et al., 1992 show that the extinction to backscattering ratios at 351nm have changed within 18 to 28sr during November and December 1991; which are similar to those we observe. Also the extinction to backscattering ratios evaluated by Jäger (personal communication, 1992) from in situ balloon soundings at Laramie, Wyoming, compare well with ours. He suggests that, in 15-25km altitude layer, C_{589nm} is in the range 20-40sr during June-October 1992 and 28-48sr during November-December 1992; our retrieved data lie in 30-50sr interval during the first period and in 45-50sr during the last months of 1992. Lognormal aerosol size distributions fitted to the measurements following the major volcanic eruptions (i.e. El Chichòn and Pinatubo) had mode radii of 0.1 to 0.3μm (Hofmann, 1987 and Pueshel

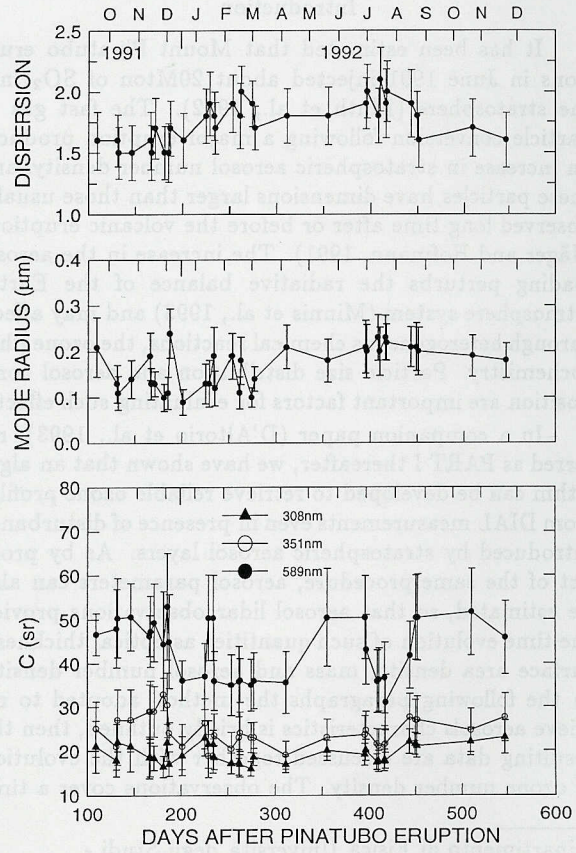

Fig.3. The dispersion σ and the mode radius r_o of the aerosol size distributions are shown in the top panels as function of time after the Pinatubo eruption. In the lower part of the figure the extinction to backscattering ratios at the wavelength available at SLAQ are also shown. There are reported only the cases for which the fitting procedure is statistically significative (see text). The error bars represent 1σ standard deviations.

et al., 1992), which again agrees with our retrieved mode radii (0.1 to 0.25μm).

Using the backscattering ratios reported in Fig.1, when it is possible to obtain a statistically significative fitting of the $A^{obs}_{\lambda_i,\lambda_j}$'s, the altitude dependent aerosol number density can be evaluated from:

$$N = \beta^R_{\lambda_i} \left[R_{\lambda_i} - 1 \right] \frac{1}{\beta^a_{\lambda_i}} \qquad (2)$$

(where $\beta^a_{\lambda_i}$, determined from the fitting procedure, has been normalized to unitary particle number density). The precision of the backscattering ratios, the indetermination of the fitted $\beta^a_{\lambda_i}$ and the error affecting the atmospheric density soundings (necessary to evaluate the molecular backscattering coefficient $\beta^R_{\lambda_i}$) determine the 1σ standard deviation on N's, which typically range between $\pm 15\%$ and $\pm 30\%$.

Knowing the mode radius and the dispersion, it is possible to obtain the particle surface area density and the mass density, respectively by:

$$S = 4\pi r_o^2 \; N \; exp\left(2 \, ln^2 \, \sigma \right) \qquad (3)$$

$$M = \rho_a \, \frac{4}{3} \pi r_o^3 \; N \; exp\left(\frac{9}{2} \, ln^2 \, \sigma \right) \qquad (4)$$

Both these quantities (Fig.4) are affected by large errors due to the indetermination of the aerosol mode radii. The standard deviations range between $\pm 50\%$ and $\pm 65\%$ for the surface area density and between $\pm 60\%$ and $\pm 90\%$ for the aerosol mass mixing ratio. The values of surface area density strongly deviate from the mid-latitude conditions before the Pinatubo eruption (i.e. S$\sim 1 \mu m^2$ cm^{-3}). Our observations show that in the altitudes from 17 to 22km S reached values between 5 and $20 \mu m^2$ cm^{-3}. The aerosol mass content ranges from 15 to 60ppbm. In spite of the large indetermination, these data compare quite well with those reported by Deshler et al., 1993, which have been derived from in situ measurements of the aerosol size distribution taken at Laramie (Wyoming) during 1991 and 1992.

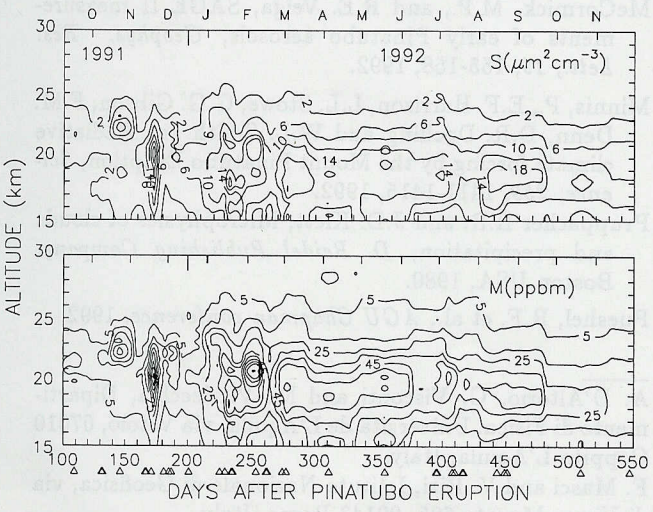

Fig.4. From the top, contour maps of the surface area density (S) and the aerosol mass mixing ratio (M; ρ_a, the aerosol mass density in Eq.(4), has bee taken equal to 1.6 $gr \; cm^{-3}$). The contour level have been obtained with 5 days step along the abscissa and 0.5km step for ordinate. The little triangles on the time scale indicate the days used for the mapping.

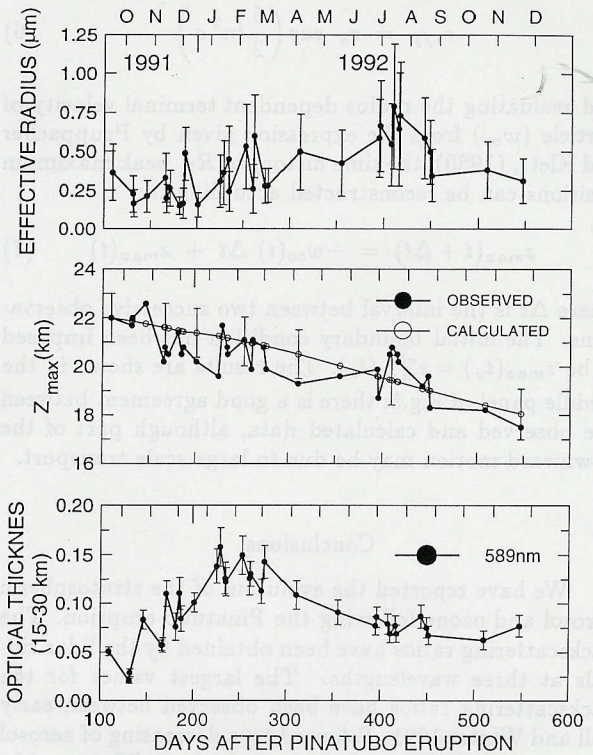

Fig.5. The effective radii of the size distributions are shown at the top. In the middle panel the maximum positions in the backscattering ratio profiles are compared to the reconstructed ones, see text for details. The optical thickness from 15 to 30km at 589nm wavelength is shown in the bottom panel. The error bars indicate 1σ standard deviations.

Also the aerosol optical thickness has been calculated in an altitude range between $z_b = 15$km and $z_t = 30$km:

$$\tau_{\lambda_i} = C_{\lambda_i} \int_{z_b}^{z_t} dz \; \beta^R_{\lambda_i} \left[R_{\lambda_i} - 1 \right] \qquad (5)$$

The results are shown in the bottom panel of Fig.5 for the 589nm wavelength. The optical thickness behavior shows two stage of the volcanic aerosol cloud. In the first phase, τ increases with a rate of 0.025 per month, reaching a maximum of 0.15 in December 1991. This is probably due to the transport of aerosols from lower latitudes. Since the beginning of 1992 τ decreases with a slower rate, which is produced mainly by sedimentation processes.

In September 1991, the backscattering ratio profiles start to show a decrease in the altitude of backscattering ratio maximum position (z^{obs}_{max}). Then, assuming the effective radius of the size distribution as the more representative dimension of the particles (top panel in Fig.5):

$$r_{eff} = r_o \, exp\left(\frac{5}{2} ln^2 \sigma\right) \qquad (6)$$

and evaluating the radius dependent terminal velocity of particle (w_∞) from the expression given by Pruppacher and Klett, (1980), the time history of R_{λ_i} peak maximum positions can be reconstructed according to:

$$z_{max}(t + \Delta t) = -w_\infty(t) \, \Delta t + z_{max}(t) \qquad (7)$$

where Δt is the interval between two successive observations. The initial boundary condition has been imposed to be $z_{max}(t_o) = z^{obs}_{max}(t_o)$. The results are shown in the middle panel of Fig.5; there is a good agreement between the observed and calculated data, although part of the downward motion may be due to large scale transport.

Conclusions

We have reported the evolution of the stratospheric aerosol and ozone following the Pinatubo eruption. The backscattering ratios have been obtained by the lidar signals at three wavelengths. The largest values for the backscattering ratios have been observed between early Fall and Winter 1991. Followed by a decreasing of aerosol load and of maximum altitude with time. The trend in altitude of the backscattering ratio maximum can be fitted with an average sedimentation velocity, which is consistent with the expected size distribution. Conversely the optical thickness shows the maximum values in the Spring of 1992.

The fitting procedure gives a quite accurate estimation of the errors on all the parameters which characterize the aerosol size distribution. The optical thickness is determined with a ±15% of uncertainty; similar or larger (up to ±40%) standard deviations must be attributed to the dispersion parameter (the σ of size distribution) and the mode radius (r_o). Although the mode radius is affected by considerable uncertainty, it shows an increasing trend up to one year from the eruption as observed for El Chichòn. The extinction to backscattering ratio is retrieved with a better precision and for the visible wavelength it shows a range of variation between 35 and 55sr.

Some comparisons with the data available in literature have been discussed. There is a general agreement both in the optical properties of the aerosols (i.e. the extinction to backscattering ratios) and in the range of the lognormal aerosol size distribution mode radii. Although affected by large errors, the retrieved aerosol number density, surface area density and mass mixing ratio are also consistent with other experimental data.

The indetermination in the ozone number density may reach ±35% within the aerosol layer but our record shows a consistent seasonal trend with larger values at lower altitude in Winter. No conclusion on the effect of heterogeneous chemistry could be drawn at this time considering the shortness of our data record.

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References

Bluth, G.J.S., S.D. Doiron, C.C. Schnetzler, A.J. Krueger and L.S. Walter, Global tracking of the SO_2 clouds from June 1991 Mount Pinatubo Eruptions, *Geophys. Res. Lett., 19*, 151-154, 1992.

D'Altorio, A., F. Masci, V. Rizi, G. Visconti and E. Boschi, Continuous lidar measurements of stratospheric aerosols and ozone after the Pinatubo eruption. PART I: DIAL ozone retrieval in presence of stratospheric aerosol layers, *Geophys. Res. Lett., this issue*, 1993.

Deshler, T, B.J. Johnson and W.R. Rozier, Balloonborne measurements of Pinatubo aerosol during 1991 and 1992 at 41°N: vertical profiles, size distribution and volatility, *Geophys. Res. Lett., 20*, 1435-1438, 1993.

Ferrare, R.A., S.H. Melfi, D.N. Whiteman and K.D. Evans, Raman lidar measurements of Pinatubo aerosols over Southeastern Kansas during November-December 1991, *Geophys. Res. Lett., 19*, 1599-1602, 1992.

Hofmann, D.J., Perturbations to the global atmosphere associated with El Chichòn eruption of 1982, *Rev. Geophys., 25*, 743-759, 1987.

Jäger, H. and D.J. Hofmann, Midlatitude lidar backscatter to mass, area and extinction conversion model based on *in situ* aerosol measurements from 1980 to 1987, *Appl. Optics, 30*, 127-138, 1991.

Larsen, N., Backscatter measurements from Thule EASOE, *Danish Meteorological Institute, Scientific Report 92-1*, Copenhagen, 1992.

McCormick, M.P., and R.E. Veiga, SAGE II measurements of early Pinatubo aerosols, *Geophys. Res. Lett., 19*, 155-158, 1992.

Minnis, P., E.F. Harrison, L.L. Stowe, G.G. Gibson, F.M. Denn, D.R. Doelling and W.L. Smith Jr., Radiative climate forcing by the Mount Pinatubo Eruption, *Science, 259*, 1411-1415, 1992.

Pruppacher H.R. and J.D. Klett, Microphysics of clouds and precipitation, *D. Reidel Publishing Company*, Boston USA, 1980.

Pueshel, R.F. et al., *AGU Chapman conference*, 1992.

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