

## Ground-Based Monitoring of Pinatubo Aerosols and Ozone at L'Aquila, Italy:

### II. – Calculation of Aerosol Surface Density for Photochemical Models.

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**Summary.** — A simple method for calculating the surface area density of aerosol particles starting from lidar backscattering ratios is described in this work. Surface density is the key parameter to be included in model assessments of the ozone perturbation following the eruption of Mt. Pinatubo. As an example we have used a photochemical 2D model to calculate the sensitivity of O<sub>3</sub> depletion to the surface density of volcanic aerosols. Monthly averaged profiles of ozone concentration measured with a DIAL system for ten months after the eruption are also shown.

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### 1. – Introduction.

An important role in activating heterogeneous chemistry outside the polar stratosphere could be played by sulfate aerosols, both from natural background and from large volcanic eruptions. A large ozone loss has been predicted [1] as a consequence of the enhanced-aerosol-surface areas following the eruption of El Chichón.

In this work we discuss a simple method to calculate the aerosol surface density for a given particle size distribution and starting from lidar backscattering ratios. Using a two-dimensional (2D) photochemical model of the stratosphere and including heterogeneous reactions over sulfuric-acid aerosols, we show the model-predicted ozone sensitivity to the surface area density of volcanic particles.

It has to be stressed that the ozone perturbation in the real atmosphere is the net effect of complex radiative dynamical photochemical interactions. The simple numerical experiment conducted here is only intended to study the sensitivity of the heterogeneous chemical destruction of ozone to the available aerosol surface density, which is in turn calculated from measured quantities; no attempt is made to model the real

ozone depletion (if any) resulting from coupling the physical mechanisms mentioned above.

## 2. - Surface density calculation.

Ten months of aerosol data collected at the lidar station of L'Aquila, Italy [2] (42°N, 13°E) (SLAQ from now on) are used as the first input for the following calculations.

These backscattering ratios need to be converted into surface area density in order to be used in photochemical calculations including heterogeneous reactions over sulfuric-acid aerosols. To do this, we first write the expression of surface ( $S$ ) and volume ( $V$ ) densities as a function of the aerosol size distribution ( $n$ ),

$$(1) \quad S = \int_0^{\infty} 4\pi r^2 n(r) dr, \quad V = \int_0^{\infty} \frac{4}{3} \pi r^3 n(r) dr,$$

where  $r$  is the particle radius. For a log-normal size distribution with dispersion  $\sigma$ , mode radius  $r_0$  and total particle density  $N$ , we obtain

$$(2) \quad \frac{S}{V} = \frac{3}{r_0} \exp \left[ -\frac{5}{2} \log^2 \sigma \right].$$

Using the aerosol volume backscattering  $\beta_a^\pi$ ,

$$(3) \quad \beta_a^\pi = \beta_R^\pi (R-1) = \int_0^{\infty} Q_\pi \pi r^2 n(r) dr,$$

and observing that for sulfate particles with  $r \leq 0.6 \mu\text{m}$  the Mie backscattering efficiency ( $Q_\pi$ ) is close to a linear function of the size parameter  $x = 2\pi r/\lambda_0$  with constant slope  $g$ , it is possible to relate the backscattering ratio  $R$  to  $V$  and finally to  $S$  through eq. (2). The resulting expression is the following:

$$(4) \quad S = \frac{2\lambda_0}{\pi g r_0} \exp \left[ -\frac{5}{2} \log^2 \sigma \right] \beta_R^\pi (R-1),$$

with  $\beta_R^\pi$  being the molecular backscattering coefficient.

A Mie-scattering numerical code has been used to calculate the backscattering efficiency in the case of spherical particles with a refractive index  $m = 1.43$  and negligible absorption [3] (at  $\lambda_0 \approx 0.590 \mu\text{m}$ ). A fit of  $Q_\pi$  from  $x = 0$  up to  $x = 6$  (*i.e.*  $r \leq 0.6 \mu\text{m}$ ) following the method suggested by Pinnick et al. [4] gives  $g = 0.017 \pm 0.003$ . The size distribution parameters ( $r_0, \sigma$ ) introduce the largest indetermination in eq. (4), due to the limited number of direct measurements made after the eruption of Pinatubo and also for our assumption of unimodal particle distribution. In addition,  $n(r)$  for volcanic aerosols changes with both altitude [3] and time, particularly at the early stages of the eruption. In this work we have adopted the size distributions measured at Laramie, WY (41°N) during August 1991 at (16÷17) and (22÷23) km altitude [5] and in early 1992 in the same location [6]. Because of the very limited amount of available size distribution measurements, a large uncertainty must be accounted for  $S$  ( $\leq 50\%$ ). The time series of the calculated aerosol surface density at 40°N is shown in fig. 1. Values larger than  $20 \mu\text{m}^2 \text{cm}^{-3}$  are found during January and February 1992, between (15÷20) km altitude; our calculation is in good agreement with independent measurements [7].

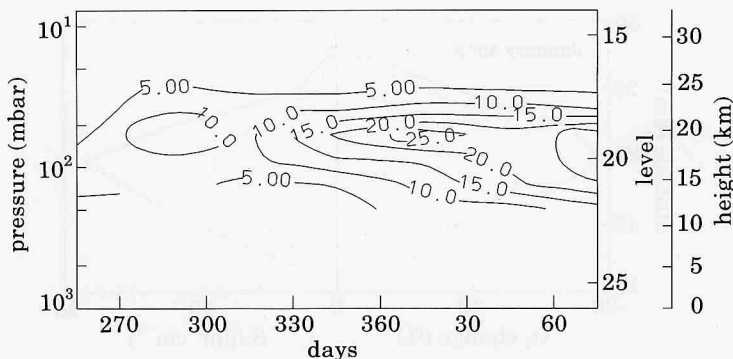
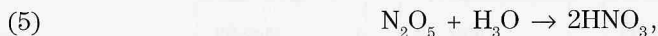


Fig. 1. - Altitude-time plot of the aerosol surface density ( $\mu\text{m}^2 \text{cm}^{-3}$ ) corresponding to the backscattering ratios measured at SLAQ (day number is on the  $x$ -axis).

### 3. - Application to a 2D model.

The results shown in fig. 1 and similar others obtained from published or prepublished lidar data [8,9] have then been used in a 2D model. The time evolution of the aerosol cloud in the tropics has been deduced from optical thickness measurements [10,11] and the volcanic cloud has been confined between  $60^\circ\text{S}$  and  $60^\circ\text{N}$ .

The 2D model adopted here includes heterogeneous chemistry over sulfuric-acid aerosols through the following reactions [12]:



For (5) the reaction probability is only slightly dependent on the amount of  $\text{H}_2\text{SO}_4$  in the aerosol and 0.1 appears a reasonable choice [13]. The probability of reaction (6), however, is a sensitive sharp function of the  $\text{H}_2\text{SO}_4$  percentage, which in turn depends on temperatures. We have adopted the recommendation given in a recent WMO assessment report [14]. We have run the model to steady state using only the WMO-recommended lower limit of background aerosols. Then we have included the aerosols injected by the eruption of Pinatubo starting from the month of August and taking into account the heterogeneous chemical perturbation only.

As a consequence of reactions (5), (6), the model predicts a sensible  $\text{NO}_2$  depletion and a large increase in ClO (up to 0.4 p.p.b.v. at 22 km altitude during winter at mid-latitudes). This ClO enhancement is in turn responsible for the model-predicted ozone depletion that reaches the largest values during winter at mid-latitudes (about 10% total-ozone reduction between  $(30 \div 60)^\circ\text{N}$  in January). In fig. 2 we show the sensitivity of the  $\text{O}_3$  depletion to the aerosol surface density: a 20% decrease of the ozone loss is found at 20 km altitude when  $S$  is lowered by 50%. We conclude that the amount of the  $\text{O}_3$  depletion due to reactions (5), (6) is rather well established in comparison to the large overall uncertainty of the potential ozone perturbation, which can also be produced by radiation and transport.

Finally, fig. 3 shows six monthly averaged ozone profiles measured at SLAQ with a DIAL system following the Pinatubo eruption. A relatively large error in the DIAL

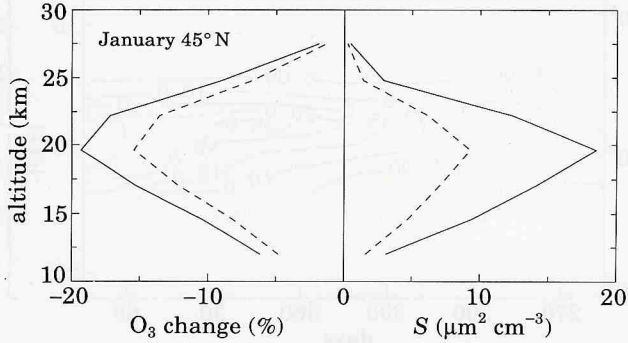


Fig. 2. – Vertical profile of model-predicted ozone depletion (left, percent) and aerosol surface density (right,  $\mu\text{m}^2 \text{cm}^{-3}$ ). Dashed lines are for a halved surface density.

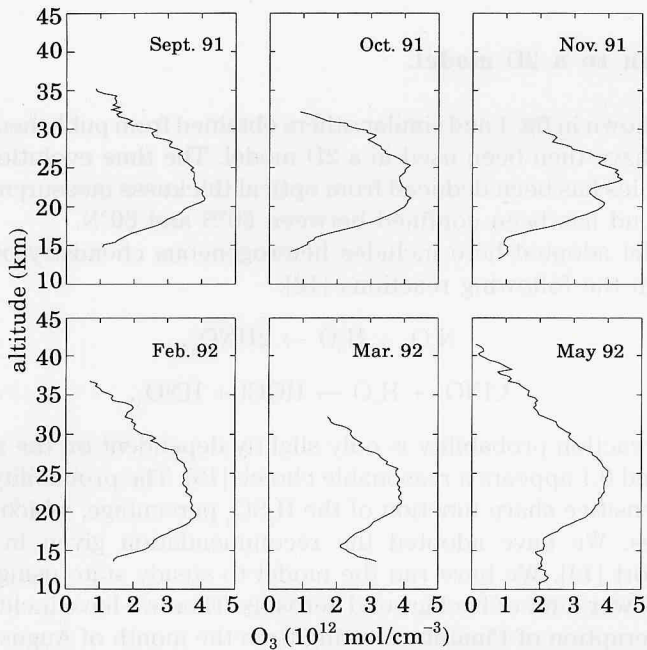


Fig. 3. – Monthly averaged vertical profiles of ozone concentration ( $10^{11} \text{mol/cm}^3$ ) measured at SLAQ (Sept.-Nov. 1991 and Feb.-May 1992).

profiles must be accounted for in the thick aerosol layer (about (15÷20)%); in this altitude range a physically meaningful retrieval technique must be adopted [15] to obtain the ozone profile from the measured backscattered signals that are partly absorbed by the volcanic aerosols.

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## REFERENCES

- [1] D. J. HOFMANN and S. SOLOMON: *J. Geophys. Res.*, **94**, 5029 (1989).
- [2] A. D'ALTORIO, F. MASCI, G. PITARI, G. VISCONTI, V. RIZI, M. CERVINO and G. GIOVANELLI: this issue p. 91.
- [3] H. JÄGER and D. HOFMANN: *Appl. Opt.*, **30**, 127 (1991).
- [4] R. G. PINNICK, S. G. JENNINGS and P. CHYLEK: *J. Geophys. Res.*, **85**, 4059 (1980).
- [5] T. DESHLER, D. J. HOFMANN, B. J. JOHNSON and W. R. ROZIER: *Geophys. Res. Lett.*, **19**, 199 (1992).
- [6] G. P. GOBBI, private communication (1992).
- [7] G. K. YUE, L. R. POOLE and L. W. THOMASON: *II HSRP/AESA meeting, Virginia Beach, VA, USA 1992*.
- [8] D. M. WINKER and M. T. OSBORN: *Geophys. Res. Lett.*, **19**, 167 (1992).
- [9] S. YOUNG, private communication (1992).
- [10] M. P. MCCORMICK and R. E. VEIGA: *Geophys. Res. Lett.*, **19**, 155 (1992).
- [11] M. P. MCCORMICK: private communication (1992).
- [12] G. PITARI, G. VISCONTI and V. RIZI: *Geophys. Res. Lett.*, **18**, 833 (1991).
- [13] D. R. HANSON, and A. R. RAVISHANKARA: *J. Geophys. Res.*, **96**, 17307 (1991).
- [14] *WMO, Scientific Assessment of Ozone Depletion - 1991*, WMO report No.25, in press (1992).
- [15] A. D'ALTORIO, F. MASCI, G. VISCONTI, V. RIZI and E. BOSCHI: *Geophys. Res. Lett.*, **19**, 393 (1992).