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

I. – Evidences for Ozone Depletion during 1991/92 Winter Months.

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Summary. — Possible evidence for stratospheric ozone depletion at northern mid-latitudes as a consequence of the Pinatubo aerosol cloud is presented in this work. A ten-month record of aerosol and ozone measurements taken at the lidar station of L'Aquila, Italy, is compared to the 1985-1986 SBUV and SAGE II ozone data. Ozonesonde data collected in the station of Hohenpeissenberg, Germany, and S. Pietro Capofiume, Italy, are used to validate the DIAL measurements corrected for the aerosol presence.

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1. – Introduction.

The discovery of the ozone hole over Antarctica, and the validation of the chemical theory through the widespread measurements collected during the AAOE-1987, have suggested the possibility that large ozone depletions could occur not only in the polar regions but also at mid-latitudes [1,2]. 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 [3] as a consequence of the enhanced aerosol surface areas following the eruption of El Chichón. More recent assessments of this mechanism [4,5] have been made by taking advantage of an updated knowledge of heterogeneous chemistry over sulfuric-acid aerosols.

A careful analysis of the ozone trend during 1980s has been carried out [6] in order to isolate the local volcanic effects from large-scale transport perturbations. Jäger and Wege estimate that 30% to 40% of the ozone deficiency between 17 to 20km is related to the aerosol perturbation, which implies that the (6-7)% loss on the total column

predicted by 2D models is probably too large. These models, on the other hand, have predicted an even stronger impact on ozone for an eruption comparable to El Chichón, but taking place in the presence of a larger background chlorine amount. The eruption of Pinatubo may offer an opportunity to test these theories.

In this work we show O_3 measurements collected from DIAL observations at L'Aquila (42°N, 13°E) (SLAQ from now on), along with ozonesonde data collected at S. Pietro Capofiume (45°N, 11°E) and Hohenpeissenberg (48°N, 11°E) (EASOE 1991-92 campaign). These ozone measurements are compared to monthly-averaged SBUV data for 1985-1986 [7] and SAGE II data for the same period.

2. - Aerosol and ozone measurements.

In fig. 1 we present ten months of data collected at SLAQ at 590nm (August 1991 - May 1992). The main feature of SLAQ is the possibility of making simultaneous measurements of both aerosols and ozone (DIAL system). In a previous work [8] we have discussed how to invert DIAL signals in the presence of intense aerosol layers in order to obtain reasonable ozone density profiles. That approach has been studied at SLAQ with the combined use of data from both O_3 DIAL and aerosol lidar. The DIAL system works at 308nm and 351nm; in clear-sky conditions, an ozone profile can be obtained at night in three hours, covering an altitude range between 12 to 35km and with an overall indetermination in the $(2 \div 15)\%$ range.

Ozone profiles retrieved form DIAL measurements taken during 1991 and early 1992 are compared to ozonesonde measurements collected in the same period, and to 1985-1986 SBUV and SAGE II satellite data as representative of the «unperturbed» atmosphere. We are aware of the intrinsic large uncertainty in this approach, mainly due to the natural fluctuations in the stratospheric-ozone transport which makes it difficult to isolate an «unperturbed, control» situation, and also to the large perturbations in the ozone retrieval from DIAL measurements when a thick aerosol layer is present. In order to reduce these uncertainties we have used independent ozonesonde data to

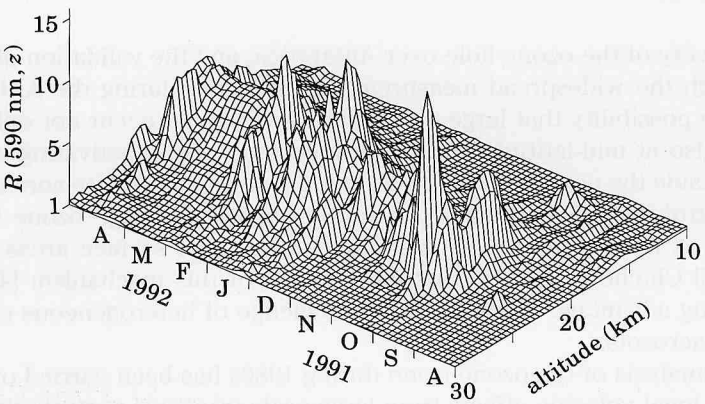

Fig. 1. Three-dimensional plot of lidar backscattering ratios measured at SLAQ, as a function of altitude and day number.

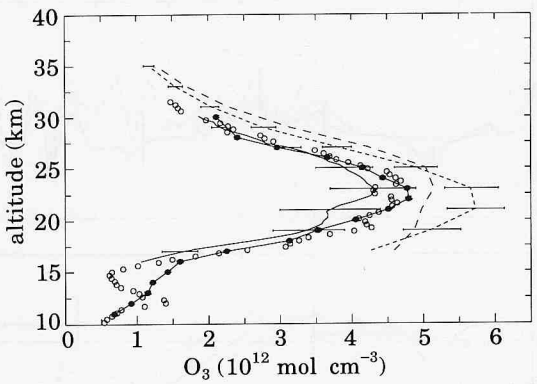

Fig. 2. Profiles of O_3 number density (10^{12} mol/cm^3). SBUV (– – –) and SAGE II (- - -) data are January averages at (40÷50)°N for 1985-1986, ozonesonde profiles are averaged over January 1992 (Hohenpeissenberg) (•) and December 1991 (S. Pietro Capofiume) (°), and finally the SLAQ DIAL profile (—) is averaged over January 1992.

validate the DIAL ozone retrieval and we have averaged over two years the monthly means of satellite data in order to smooth out the QBO signal. On the other hand, no correction has been made for the solar cycle and for the decadal ozone trend related to the atmospheric composition (from TOMS data this trend can be estimated as (3÷4)% decrease in 5 years of Northern mid-latitudes during the winter season).

In fig. 2 the January-averaged DIAL ozone profile corrected for the presence of aerosols seems to be in reasonable agreement with the ozonesonde data except for an average 14% underestimation in a 3km layer centred at 20.5km height. This may be attributed to the correction for the aerosol presence. Error bars on the DIAL ozone data take into account both the overall uncertainty in the measured backscattered signal (< 5% between (15÷30)km height) and the uncertainty in the correction for the aerosol presence (estimated to be $\simeq$ (10÷15)% for large values of the backscattering ratio). Uncertainty on SAGE II data is taken from a recent WMO assessment report [9]. The ozone underestimation shown by DIAL and sondes with respect to SAGE II and SBUV above 25km may be partly attributable to the chemically driven decadal ozone trend.

The time series of DIAL and ozonesonde data is shown in fig. 3 in terms of O_3 column between 18 and 28 km altitude. A comparison with the SBUV column data shows a good fit up to the end of November 1991, while a sharp column difference is experienced during December 1991 and January 1992. An average 8% to 10% loss is found for ozonesonde data averaged during this time period (the depletion is larger of about 4% when using the SAGE II data as the unperturbed reference). This O_3 loss has to be compared to an average (15±10)% loss deduced from lidar measurements: the larger column difference measured by DIAL is consistent to what shown by fig. 2.

3. - Discussion and conclusions.

Considerable caution should be used to interpret the data shown so far, and additional measurements need to be accumulated for a longer period. The assumption

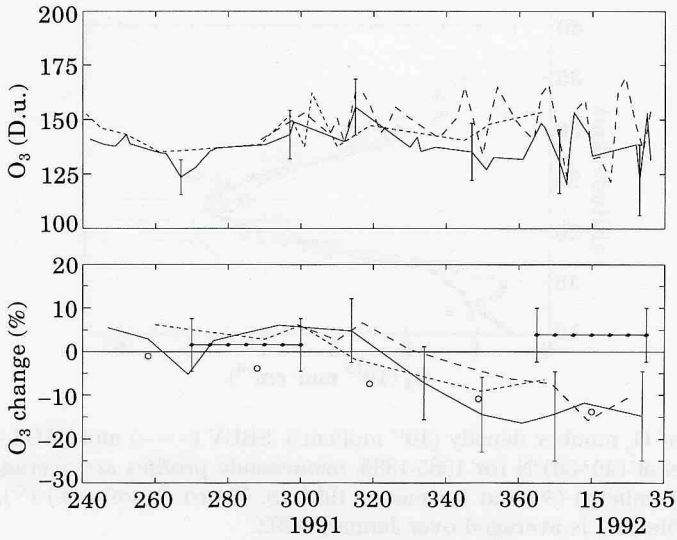

Fig. 3. Top panel shows the time series of DIAL *vs.* ozonesonde integrated O_3 column between 18 and 28 km (Dobson units); day number is on the *x*-axis. Bottom panel shows the percent differences for DIAL and ozonesonde O_3 columns ((18÷28)km) with respect to the monthly averaged SBUV data at (40÷50)°N for 1985-1986; ozonesonde and DIAL data are time-averaged over 15 days. January and October percent deviations of SAGE II data are also shown. Hohenpeissenberg (– – –), SLAQ (—), S. Pietro Capofiume (- - -), SAGE II (•••). Open circles are the corresponding percent differences as predicted by a photochemical 2D model including heterogeneous chemistry over sulfate aerosols (the differences are calculated with respect to an unperturbed control run without Pinatubo aerosols).

that the observed ozone trend in the last few months may be attributed to a volcanic effect is supported from heterogeneous chemical considerations. The following reaction can be significant over sulfuric-acid aerosols [10]:

(1) $$N_2O_5 + H_2O \rightarrow 2HNO_3 ,$$

(2) $$ClNO_3 + H_2O \rightarrow HOCl + HNO_3 .$$

In the presence of volcanic aerosols, the total particle surface available for these reactions is so large that ClO can increase of about an order of magnitude. This is a consequence of the NO_x decrease forced by reactions (1) and (2): in these conditions the ClO-$ClNO_3$ equilibrium is shifted toward ClO and the chlorine catalytic cycle for O_3 destruction becomes faster (more than the corresponding slowing-down of the odd nitrogen cycle).

Calculations carried out with a photochemical 2D model available at our research group predict a ClO mixing ratio consistent with unperturbed measurements in the lower stratosphere [11], and a ClO increase of about one order of magnitude during the winter after the Pinatubo eruption. This seems consistent with measurements collected during AASE II and EASOE 1991-92. The corresponding large NO_2 depletion has also been measured [12] and its order of magnitude is consistent with our model predictions. Model results for O_3 are included in fig. 3.

A final remark should be made on the existence of other important mechanisms for ozone interaction with volcanic aerosols (*i.e.* radiative and dynamical [13]).

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