Sun-photometric measurements of atmospheric turbidity variations caused by the Pinatubo aerosol cloud in the Himalayan region during the summer periods of 1991 and 1992

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Summary. — Measurements of direct solar irradiance were taken episodically on two days of September 1990 and regularly in the summer periods from July to October 1991 and from mid-July to mid-August 1992 at the Pyramid Laboratory (5050 m a.m.s.l.) situated at the foot of Mt. Everest (Nepal), using two examples of the Volz multispectral sun-photometer, model A. These sun-photometric measurements were analysed in terms of the Bouguer-Lambert-Beer law in order to determine the values of aerosol optical thickness at the three sun-photometric window-wavelengths. Examining these spectral series in terms of the well-known Ångström formula, the best-fit values of turbidity parameters α and β were calculated with great accuracy. From the measurements taken in September 1990, we found values of the aerosol optical thickness in good agreement with the mean values of the background aerosol optical depth measured at the Mauna Loa Observatory (Hawaii) during the four-year period from 1988 to 1991. The spectral values of the aerosol optical thickness determined during the summer of 1991 show that the mean daily values of parameter β increased abruptly from about 0.06 to more than 0.16 from July 26 to 28, 1991, and then varied between 0.09 and 0.16 during the rest of the measurement period. Simultaneously, parameter α was found to decrease from more than 1.25 to 0.39 towards the end of July and to vary between 0.40 and 0.78 during September. These large variations of both atmospheric turbidity parameters have been attributed to the growth of the aerosol particles and to the consequent changes in the size distribution curve of the Pinatubo aerosol particles. Analysing the variations of the stratospheric aerosol optical depth in terms of particle polydispersions consisting of a linear combination of a background aerosol monomodal model and a bimodal model representing the Pinatubo fresh aerosol particle size distribution, the vertical mass loading of stratospheric aerosol particles was estimated to vary between 0.037 and 0.047 g m^{-2} from July 24 to 27 and to increase to values ranging between 0.048 and 0.074 g m^{-2} during the rest of summer 1991, presenting a mean value of (0.063 ± 0.009) g m^{-2} for the two-month period. The measurements taken in summer 1992 show that parameter β ranged between 0.08 and 0.12, while parameter α was found to vary between 0.23 and 0.73. The strato-

spheric aerosol depth values were analysed through a best-fit procedure based on a linear combination of a background model of small aerosol particles and a trimodal model consisting of aged volcanic aerosol particles. Following this procedure, the vertical mass loading of Pinatubo aerosol particles was found to vary between 0.043 and 0.057 g m^{-2} during summer 1992, the mean value being evaluated as equal to (0.047 ± 0.004) g m^{-2}.

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1. – Introduction

Within the framework of the Everest-K2-CNR Scientific Project and as part of a cooperation agreement between the Italian National Council of Research (CNR) and the Royal Nepal Academy of Science and Technology (RONAST), the Pyramid Laboratory was erected at 5050 m MSL in the National Park of Sagarmatha (Nepal) (27° 57′ N, 86° 49′ E), on a morainic terrace situated on the right side of the Khumbu Glacier at the foot of Mt. Everest (Himalaya). This building was designed as a high-altitude laboratory suitable for developing scientific and technological research concerning geological, geodetical, glaciological, medical, biological, hydrological and environmental themes. Since high-altitude measurements of direct solar irradiance at visible and near-infrared wavelengths can be very useful in evaluating the solar radiation extinction effects produced by the aerosol particles suspended in the upper troposphere and stratosphere, we decided to take the opportunity to carry out routine multispectral sun-photometric measurements at the Pyramid Laboratory during the summer period over two or three years. However, given the difficulties in transporting fragile instruments by air from Milan (Italy) to Kathmandu (Nepal) and then to Lukla (about 2800 m MSL) and, subsequently, in carrying them by pack-animals and/or porters from Lukla to the high-altitude station of the Pyramid Laboratory, we decided to begin to record these measurements using the multispectral sun-photometer, model A, manufactured by Volz [1] several years ago. This is a portable, very robust instrument, without fragile optical and electronic elements, and can easily be carried by knapsack without fatigue or risk along the steep tracks which lead from Lukla to the Pyramid Laboratory, a trek of at least six days.

For normal transparency conditions of the atmosphere, the solar radiation extinction produced by aerosol particles at 5 km height is really very small, giving values of aerosol optical depth at visible and near-infrared wavelengths generally varying between 0.04 and no more than 0.07 [2]. However, when strong volcanic eruptions episodically inject sulphur gases into the stratosphere, dense clouds of aerosol particles can form at relatively low stratospheric altitudes within one or two months, as a result of highly efficient gas-to-particle conversion processes. Thus, the scattering effects produced by a volcanic aerosol cloud may become very intense at all

visible and near-infrared wavelengths, to such an extent that the backscattering of solar radiation increases suddenly and the atmospheric albedo is thus considerably enhanced. When these stratospheric clouds of volcanic aerosol particles extend to cover a large part of the Earth, the equilibrium conditions of the radiation balance of the planet are significantly changed so that atmospheric cooling processes of several tenths of a degree over the global scale can be generated for a period of at least one year.

Two examples of the Volz sun-photometer, model A, were carried to the Pyramid Laboratory on the first days of September 1990. The measurements were taken on two days only during the scheduled summer period of 1990 because of the unfavourable meteorological conditions. However, these few measurements were enough to provide reliable evaluations of the aerosol optical thickness corresponding to the background conditions of atmospheric transparency, associated with the presence of thin layers of aerosol particles in both the upper troposphere and stratosphere. Other more extensive sets of measurements were collected during the summer periods of 1991 and 1992. The sun-photometric measurements carried out during summer 1991 showed with great clarity that the atmospheric turbidity parameters suddenly changed as a consequence of the arrival above the Himalayan area during the first half of July of the volcanic aerosol particle cloud generated by the violent eruptions of Mt. Pinatubo (15° 08′ N; 120° 21′ E) on the island of Luzon (Philippines) between June 10 and 17, 1991. The sun-photometric measurements taken during summer 1992 clearly showed that the Pinatubo aerosol cloud remained above the Himalayan region even one year after the eruption, producing stable extinction features of direct solar irradiance. Since these measurements were correctly performed in a remote area of the planet giving good evaluations of the aerosol optical depth at selected wavelengths and of atmospheric turbidity parameters, they may be of interest to all studies aiming to define the climatic effects generated by the Pinatubo eruption. Therefore, we present here not only the results describing the evolutionary features of the spectral series of aerosol particle optical thickness and Ångström's [3] turbidity parameters, but also the criteria adopted in order to take the sun-photometric measurements during the various campaigns and analyse them, according to the sun-photometry method proposed by Shaw [4]. This serves the purpose of showing without doubt that the present results are wholly reliable, even if obtained from measurements taken without sophisticated instrumentation. A comparison of our results with those simultaneously obtained by Pueschel *et al.* [5] at the Mauna Loa Observatory (Hawaii) is also presented. Moreover, this work provides estimates of the vertical aerosol mass loading of the Pinatubo particle cloud, as derived from the sun-photometer measurements carried out during the summer periods of 1991 and 1992. Such estimates were made using the Pinatubo cloud extinction models defined by Pueschel *et al.* [5] on the basis of stratospheric aerosol samples taken at various altitudes between 10 and 20 km, from February 1991 to March 1992.

2. – Sun-photometric measurements

The Volz sun-photometer, model A (in the version used since late 1979), takes measurements of direct solar irradiance within three spectral intervals defined by interference filters with passband transmission curves having peak-wavelengths of 380, 500 and 875 nm and half-bandwidths of 11, 10 and 17 nm, respectively. The fourth

interference filter centred at 946 nm wavelength with a half-bandwidth of 16 nm is used to take simultaneous measurements of solar radiation absorption within a spectral interval occupied by the water vapour band σ. From the measurements performed at 875 nm and 946 nm wavelengths, evaluations of precipitable water can be obtained [1].

The Volz sun-photometer can be manually pointed at the Sun in the direction indicated by a pinhole sight. Since the circular field of view of the instrument has an angular diameter slightly smaller than 2°, while the solar disk has an angular diameter of about 30′, each output voltage also includes a small contribution from the diffuse solar radiation entering the instrument. Therefore, an error is certainly made when the output voltage is attributed to direct solar radiation alone. Since such an error considerably increases as a function of solar zenith angle and, hence, of the relative optical air mass m [4], we decided to avoid taking measurements for values of m larger than 3, that is for values of solar elevation angle h lower than 18°. In actual fact, the horizon around the Pyramid Laboratory is notably bounded by very high mountains, and therefore we could take the measurements only for values of h greater than 20° and, hence, for values of m smaller than 2.8.

Important errors are often made in reading the output signals on the graduated scale of the microamperometer mounted on the sun-photometer. In order to reduce such errors, each sun-photometric measurement was made by systematically recording three consecutive spectral series of output voltages, each series consisting of four monochromatic output signals corresponding to the four peak-wavelengths mentioned above. Thus, the time-interval required for each measurement was of about three minutes. Such cautious criteria were adopted with the main purpose of routinely collecting triplets of output signals for each window-wavelength within a few minutes, so as to obtain sets of homogeneous measurements relative to very similar conditions of atmospheric transparency, from which the average values of atmospheric optical depth can be reliably determined.

Moreover, in order to minimize possible errors in evaluating the relative optical air mass m, we regularly recorded the measurement time of each output signal with a precision of a few seconds. In this way, the solar elevation angle h was calculated with good precision as a function of the local time, the astronomical coordinates of the Sun and the geographical coordinates of the measurement station, making use of the formulas commonly adopted by astronomers [6, 7]. The relative optical air mass m was determined for each value of h using the well-known Kasten [8] formula for the vertical profile of air density above the 5 km height, as given in the ARDC Model Atmosphere 1959.

Another important error is certainly caused by the thermal drift of the integrated silicon detector/amplifier used as the sensor. Thus, the internal temperature of the instrument given by the small thermometer incorporated in the sun-photometer was routinely checked at the beginning and the end of each spectral measurement. Between one measurement and another, the sun-photometer was kept in the shade, so as to avoid warming due to direct sun-light. On the basis of these temperature measurements, we appropriately corrected all the sun-photometric signals for temperature deviations caused by the thermal drift of the sensor, using the correction factors proposed by the manufacturer to normalize all output voltages to an internal temperature of the instrument equal to 20 °C.

During summer 1990, the sun-photometric measurements were performed only on September 20 and 21, as very bad meteorological conditions prevented measurements

for several days during the proposed observation period. Conversely, regular measurements were taken during two distinct periods of summer 1991, that is on seven clear-sky days from July 24 to August 5, 1991, and another five clear-sky days from September 19 to October 2. Everyday, these measurements were performed at different solar elevation angles, more frequently during the morning than in the afternoon, since the formation of orographic clouds above the station was usually more extensive during the hours of most intense solar heating. Following the same criteria, an extensive measurement campaign was performed during summer 1992, from July 23 to August 11, collecting clear-sky measurements of direct solar irradiance on twelve days in all.

3. – Calibration procedure

Each output voltage $J(\lambda)$ given by the sun-photometer within a certain spectral range centred at the window-wavelength λ is related to the monochromatic zero-air-mass output voltage $J_0(\lambda)$ (which is the output signal given by the sun-photometer outside the atmosphere for the mean yearly Earth-Sun distance) through the following relationship based on the well-known Bouguer-Lambert-Beer law:

$$J(\lambda) = R\, J_0(\lambda) \exp\left[-m\, \delta(\lambda)\right], \tag{1}$$

where:

1) the parameter R is the correction factor used to take into account the day-to-day variations of $J_0(\lambda)$ due to the periodical changes in the Earth-Sun distance arising from the eccentricity of the Earth's orbit [6];

2) the relative optical air mass m is given by the ratio between i) the integral of air density along the oblique atmospheric trajectory described by the incoming direct solar radiation, and (ii) the integral of air density along the vertical atmospheric path [6], so that this quantity gives in practice a measure of the relative optical length of the sun-path; and

3) the monochromatic optical thickness $\delta(\lambda)$ of the atmosphere gives a measure of the overall extinction of direct solar radiation along the vertical atmospheric path and, therefore, is the sum of various partial terms due to Rayleigh scattering, aerosol particle extinction and molecular absorption by minor gases, such as ozone, nitrogen dioxide and water vapour.

Therefore, the zero-air-mass signal $J_0(\lambda)$ must be determined with a very high precision at each window-wavelength if one wishes to obtain correct evaluations of the atmospheric optical thickness $\delta(\lambda)$. For this purpose, the Langley plot method is commonly used, applying it to monochromatic sets of sun-photometric measurements taken at different solar elevation angles (and, hence, for various values of the relative optical air mass m) on days characterized by stable features of atmospheric transmittance at visible and near-infrared wavelengths [4]. The sun-photometer measurements taken on September 20 and 21, 1990, were all examined applying the Langley plot method within the limited range of m feasible at the Pyramid Laboratory, in order to obtain reliable estimates of the calibration constants $J_0(\lambda)$ at the three sun-photometric window-wavelengths. The tern of values of $J_0(\lambda)$ determined from these measurements for one of the two sun-photometers is shown in fig. 1.

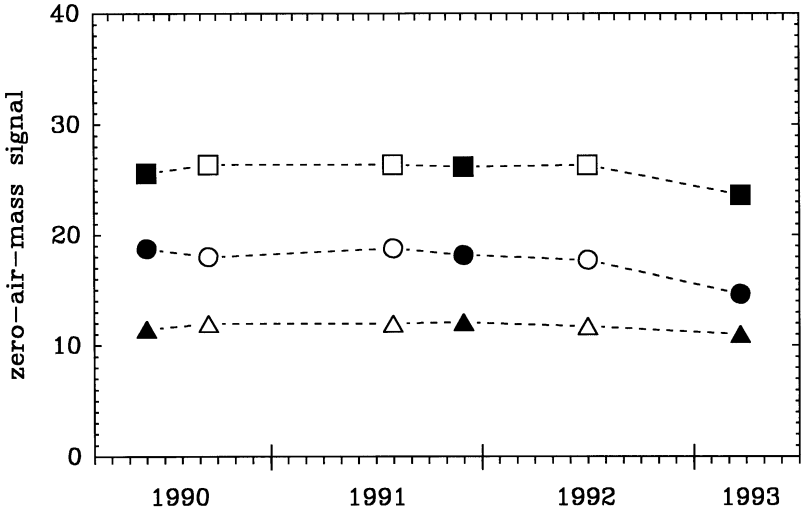

Fig. 1. – Time variations of the zero-air-mass signals $J_0(\lambda)$ determined at the three window-wavelengths of the Volz sun-photometer No. 596 over a four-year period. Circles refer to $J_0(380\ \text{nm})$, squares to $J_0(500\ \text{nm})$ and triangles to $J_0(875\ \text{nm})$. Solid simbols represent the average values of $J_0(\lambda)$ obtained through the intercomparison of measurements taken simultaneously with both Volz sun-photometers and the UVISIR-2 sun-radiometer, during the last days of May 1990 at the San Pietro Capofiume station and during the second half of November 1991 and April 1993 at the Sestola station. Open simbols refer to the average values of $J_0(\lambda)$ obtained by applying the Langley plot method to the measurements performed at the Pyramid Laboratory station on clear-sky days of September 1990, July 1991 and July-August 1992.

Two daily sets of calibration constants $J_0(\lambda)$ relative to the three window-wavelengths were determined for the measurement period in summer 1991 by applying the Langley plot method to the measurements of $J(\lambda)$ taken on July 24 and 25, 1991, corresponding to different values of m. These two days were selected because they presented the most propitious and stable atmospheric transparency conditions observed throughout the measurement period in 1991. For each triplet of monochromatic output signals $J(\lambda)$, we calculated the average values of both $J(\lambda)$ and m. Subsequently, we applied the Langley plot method to each daily monochromatic set of the above values, plotting the natural logarithms of $J(\lambda)$ as a function of m. An example of Langley plot is shown in fig. 2, relative to the measurements taken on the morning of July 24, 1991. Following this procedure, we obtained for each day a triplet of best-fit lines relative to the three window-wavelengths and the intercept values of the best-fit lines at air mass $m = 0$. In all cases where the atmospheric optical depth $\delta(\lambda)$ was stable in time during the calibration measurement period, each intercept value can be correctly considered to be equal to $R\,J_0(\lambda)$, according to eq. (1). Consequently, spectral triplets of calibration constants $J_0(\lambda)$ were determined for each calibration day by simply dividing the above intercept values by the corresponding daily value of R. The calibration constants $J_0(\lambda)$ determined for each sun-photometer on the two days selected above were found to differ very little at all three window-wavelengths. The mean values obtained for one of the two sun-photometers are shown in fig. 1.

Similarly, the Langley plot method was applied to the sun-photometric measure-

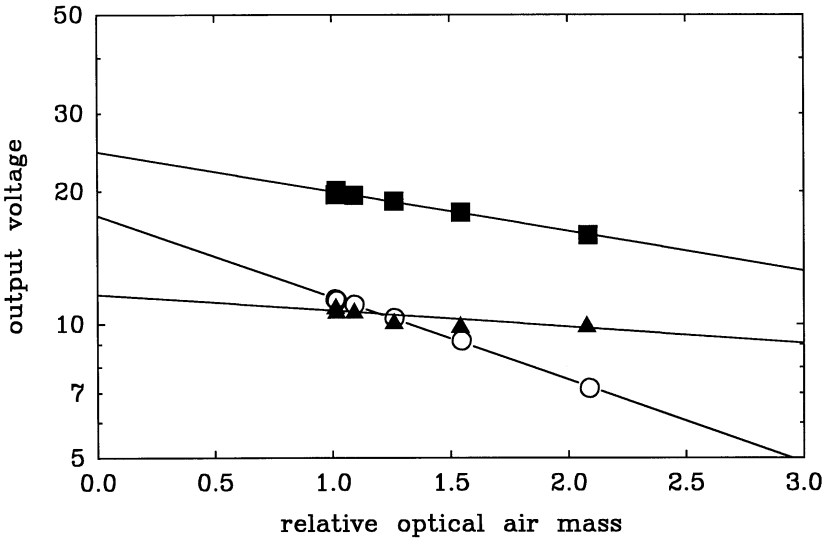

Fig. 2. – Best-fit lines drawn by applying the Langley plot method to the mean values of the output voltages $J(\lambda)$ measured at the Pyramid Laboratory (5050 m MSL) on July 24, 1991, with the Volz sun-photometer No. 596. Circles refer to J(380 nm), squares to J(500 nm) and triangles to J(875 nm).

ments taken at the Pyramid Laboratory on July 29 and August 11, 1992, so as to obtain two triplets of average calibration constants $J_0(\lambda)$ for both sun-photometers, relative to the observation period in summer 1992. Following the Langley plot procedure, we determined the average values of $J_0(\lambda)$ shown in fig. 1.

However, considering that all the above triplets of average values of $J_0(\lambda)$ were determined examining measurements taken within very small intervals of m, as clearly shown in fig. 2, we decided to further verify the reliability of the calibration results through a supplemental calibration procedure based on the intercomparison between hree sets of direct solar irradiance measurements taken with the two Volz sun-photometers and simultaneous measurements performed using the more sophisticated multispectral sun-photometer, model UVISIR-2 (portable version), designed and built at the Institute FISBAT-CNR (Bologna) [9]. The intercomparison measurements were performed in three different situations from summer 1990 to 1993: i) on May 31, 1990, at the S. Pietro Capofiume station (9 m a.m.s.l.), some 25 km North-East of Bologna (Italy), ii) on November 28, 1991, at the Sestola station (910 m a.m.s.l.) in the Apennine chain, about 55 km South-West of Bologna, and iii) on April 20, 1993, at the Sestola station. Sun-photometer UVISIR-2 is the advanced version of the multiwavelength sun-photometer (model FISBAT) built by Tomasi *et al.* [10] more than twelve years ago. It was used many times and successfully in Italy [11] and at the Terra Nova Bay station in Antarctica [9, 12, 13], where several measurement campaigns were performed hin order to determine the atmospheric turbidity parameters produced by aerosol particles in the Antarctic atmosphere. During these campaigns, several calibration tests were routinely carried out, providing excellent results thanks to the exceptional transparency conditions of the atmosphere in the Terra Nova Bay region. This multispectral sun-photometer is mounted on a cine-camera tripod and can be

manually pointed at the Sun by means of an electronic pinhole sight with a precision of less than 6′. It is equipped with:

1) Twelve interference filters with passband transmission curves presenting peak-wavelengths ranging between 320.2 and 1047.4 nm and half-bandwidths varying between 7.2 and 13.6 nm [9, 11, 12].

2) A quartz lens of 38 mm diameter and focal length varying from 149.3 to 160.8 mm as wavelength ranges from 320 to 1050 nm. This lens is coupled with a circular diaphragm of 2.5 mm diameter placed at a distance of 156 mm from the lens, so that the sun-photometer has an angular field of view of circular shape with diameter varying between 59′ and 1° 03′ throughout the above wavelength range.

3) An UDT Photop, model 555UV, utilized as a sensor and equipped with an electronic device giving amplified output voltages which are steadily proportional to the incident solar radiation at all the sun-photometric wavelengths. This sensor is housed in a thermostating box kept at a stable temperature of (50 ± 1) °C, so that errors caused by the thermal drift of the responsivity characteristics of the sensor are wholly negligible.

As mentioned above, the intercomparison measurements between the two Volz sun-photometers and the more sophisticated UVISIR-2 sun-radiometer were carried out at the end of May 1990 at the low-level station of San Pietro Capofiume and on the last days of November 1991 and during April 1993 at the mountain station of Sestola, on days characterized by clear-sky and very stable conditions of atmospheric transmittance. The measurements taken on May 31, 1990, with the UVISIR-2 sun-photometer were analysed using the calibration constants $J_0(\lambda)$ determined by applying the Langley plot method to the measurements carried out at the Italian station of Terra Nova Bay (Antarctica) in January 1990 (only a few months prior to the intercomparison) for exceptionally high transmittance conditions of the atmosphere and over a very wide range of m from 1.5 to more than 7 [12]. The measurements taken in November 1991 (only a few weeks after the return of the Volz sun-photometers in Italy) were examined using the calibration constants obtained by applying the Langley plot method to the measurements taken at the Sestola station over a wide range of m from 2.4 to more than 7 [11]. Similarly, the measurements performed in April 1993 were analysed by following the Langley plot method over a large range of m from 1.2 to about 6.5.

It is important to point out that the three sets of calibration constants found for the UVISIR-2 sun-photometer in the three periods are in good mutual accordance at all the window-wavelengths. Subsequently, the zero-air-mass signals of the Volz sun-photometers were determined in terms of eq. (1), imposing that the values of atmospheric optical thickness $\delta(\lambda)$ at the three wavelengths are equal to those obtained through a bilogarithmic interpolation in wavelength of the measurements given by the UVISIR-2 sun-radiometer. All the three sets of $J_0(\lambda)$ were found to present very small discrepancies from the values of $J_0(\lambda)$ obtained directly by applying the Langley plot method to the measurements taken at the Pyramid Laboratory in September 1990 and on July 24 and 25, 1991, as well as on the two most favourable days of summer 1992. In fact, the absolute values of the relative differences between the various monochromatic values of $J_0(\lambda)$ found in the various years turn out to vary within $+0.6\%$ and -4.2%, as can be clearly seen in fig. 1. The most marked discrepancies between the values of

$J_0(\lambda)$ obtained at the Pyramid Laboratory and those calculated through the intercomparison procedure were found between the set determined in summer 1992 at the Pyramid Laboratory and that of April 1993 at the Sestola station. These variations are presumably due to the ageing of interference filters caused by problems during the journey home. Therefore, since the above results indicate that the spectral responsivity characteristics of both Volz sun-photometers remained very stable at all the three window-wavelengths during the period from May 1990 to August 1992, we decided to use the three terns of mean values of $J_0(\lambda)$ found at the Pyramid Laboratory as calibration constants, in order to analyse the sets of sun-photometric measurements performed during the three field campaigns at our Himalayan station.

4. – Determination of aerosol particle optical thickness

The monochromatic values of optical thickness $\delta(\lambda)$ were calculated for all the output voltages measured in September 1990 and during the summer periods of 1991 and 1992, in terms of the following formula

$$\delta(\lambda) = \ln [R\, J_0(\lambda)/J(\lambda)]/m\,, \tag{2}$$

derived from eq. (1). We evaluated the particulate optical thickness $\delta_{\mathrm{p}}(\lambda)$ by subtracting from each value of $\delta(\lambda)$ the optical thicknesses $\delta_{\mathrm{R}}(\lambda)$ due to Rayleigh scattering, $\delta_{\mathrm{o}}(\lambda)$ due to ozone absorption, $\delta_{\mathrm{n}}(\lambda)$ due to nitrogen dioxide, and $\delta_{\mathrm{w}}(\lambda)$ due to water vapour, according to the following formula:

$$\delta_{\mathrm{p}}(\lambda) = \delta(\lambda) - \delta_{\mathrm{R}}(\lambda) - (m_{\mathrm{o}}/m)\,\delta_{\mathrm{o}}(\lambda) - (m_{\mathrm{n}}/m)\,\delta_{\mathrm{n}}(\lambda) - (m_{\mathrm{w}}/m)\,\delta_{\mathrm{w}}(\lambda)\,. \tag{3}$$

Equation (3) is written in a very careful form which takes into account that the optical air mass parameters m_{o} for ozone, m_{n} for nitrogen dioxide and m_{w} for water vapour differ appreciably from relative optical air mass m calculated through the Kasten [8] formula.

The Rayleigh optical thickness $\delta_{\mathrm{R}}(\lambda)$ was calculated at the three wavelengths according to the equation

$$\delta_{\mathrm{R}}(\lambda) = (p_0/1013.25)\,u_{\mathrm{R}}(\lambda)\,, \tag{4}$$

where p_0 is the total air pressure measured in hPa at the station level and $u_{\mathrm{R}}(\lambda)$ is the monochromatic optical depth given by Rayleigh scattering, normalized to a sea-level air pressure of 1013.25 hPa. The values of $u_{\mathrm{R}}(\lambda)$ were calculated by: i) adopting the computational programme written by Fröhlich and Shaw [14]; ii) using the correction factor equal to 1.031, proposed by Young [15] to take into account the depolarization effects due to molecular anisotropy; iii) representing the atmosphere by means of the Tropical model relative to 15° N latitude [16] in the height range from 5 to 100 km only; and iv) normalizing the monochromatic values of Rayleigh optical depth, as obtained in iii), to the sea-level air pressure of 1013.25 hPa. Following this procedure, we obtained values of $u_{\mathrm{R}}(\lambda)$ equal to 0.4468, 0.1439 and 0.0149 at the 380, 500 and 875 nm wavelengths, respectively.

As clearly shown by Leckner [17], the ozone absorption band in the visible and near-infrared spectral range, usually called the Chappuis band, exhibits relatively high values of spectral intensity within the (445 ÷ 780) nm wavelength range. Therefore, the ozone optical thickness $\delta_{\mathrm{o}}(\lambda)$ was calculated only at the 500 nm wavelength, by multi-

plying the ozone absorption coefficient $\chi(500\ \text{nm}) = 0.0294\ (\text{cm STP})^{-1}$ [18] by the daily mean value of the atmospheric vertical content of ozone. This quantity was assumed to be equal to 0.28 cm STP in summer 1990 and to vary between 0.24 and 0.26 cm STP during summer 1991 and between 0.25 and 0.27 cm STP during summer 1992. These assumptions were made i) according to the measurements of total ozone carried out at Indian stations [19] and to the general trend of this parameter measured in the tropical areas [20], and ii) taking into account that the total ozone measurements performed at low latitudes after the Pinatubo eruptions of June 1991 showed a decrease of no more than 10% with respect to unperturbed climatological conditions [21-24]. However, the ozone optical depth $\delta_o(500\ \text{nm})$ usually assumes values of no more than 0.008 at low latitudes, so that fluctuations of $10 \div 20\%$ in total ozone can only cause changes of a few percent in the overall optical depth produced by aerosols and atmospheric gases. The relative optical ozone mass m_o was calculated using the formula proposed by Robinson [25] for an altitude of maximum ozone concentration equal to 22 km: the ratio m_o/m was found to assume a value of 1.0001 for the apparent solar elevation angle $h = 65°$, 0.9935 for $h = 30°$ and 0.9831 for $h = 20°$.

The optical thickness $\delta_n(\lambda)$ due to nitrogen dioxide absorption was calculated at the first two wavelengths by multiplying the NO_2 absorption coefficients $\xi(380\ \text{nm}) = 16.9\ (\text{cm STP})^{-1}$ and $\xi(500\ \text{nm}) = 5.6\ (\text{cm STP})^{-1}$ [26, 27] by the background value of atmospheric vertical content of NO_2. The latter quantity was assumed to be equal to $5 \cdot 10^{-4}$ cm STP during the three summer periods, according to some estimates in remote areas [28, 29] and taking into account that changes in the total stratospheric content of nitrogen dioxide caused by the Pinatubo volcanic cloud were observed not to exceed -20% [30-32]. On the basis of the above remarks, the optical thickness $\delta_n(380\ \text{nm})$ was evaluated to assume a mean value of 0.009 in the absence of volcanic aerosol clouds, while the decrease of this physical quantity was estimated to be very small (of no more than -0.002) in cases of a strong depletion of total nitrogen dioxide due to the presence of volcanic particle clouds, like those observed during the summer periods of 1991 and 1992. The relative optical mass m_n was calculated using the Robinson [25] formula for a maximum NO_2 concentration height of 20 km [28]. This parameter was found to assume values very close to those of air mass m_o throughout the whole range $h > 20°$.

The water vapour optical thickness $\delta_w(\lambda)$ was calculated at 875 nm wavelength as the product of the weak absorption coefficient of water vapour by precipitable water. On the basis of the spectral evaluations given by Leckner [17] in the 865 to 885 nm wavelength range, the absorption coefficient was evaluated to be equal to $2 \cdot 10^{-3}\ \text{g}^{-1}\ \text{cm}^2$, while precipitable water w was determined from the hygrometric ratio given by the Volz sun-photometer [1]. The parameter w was found to vary between 0.5 and $2.0\ \text{g cm}^{-2}$ during summer 1991 and between 0.8 and about $2.0\ \text{g cm}^{-2}$ during summer 1992. These relatively high values of w can be reasonably explained bearing in mind that intense convective motions frequently develop along the southern sides of the Himalayan chain during the summer months, causing an intense vertical transport of warm and humid air masses up to relatively high altitudes. The relative optical water vapour mass m_w was calculated using the formula proposed by Kasten [8].

For each triplet of spectral series of $J(\lambda)$, we obtained three spectral series of $\delta_p(\lambda)$, from which we calculated the average values of $\delta_p(380\ \text{nm})$, $\delta_p(500\ \text{nm})$ and $\delta_p(875\ \text{nm})$. From these values subdivided into hourly sets for each measurement day, we determined the mean hourly values of $\delta_p(\lambda)$ at the three wavelengths. Following this procedure, we calculated the mean hourly values of $\delta_p(\lambda)$ relative to September 20

and 21, 1990, from which the mean daily values of $\delta_p(\lambda)$ were determined: δ_p(380 nm) was found to be equal to (0.014 ± 0.010) and (0.038 ± 0.014) on the two measurement days, respectively, δ_p(500 nm) equal to (0.019 ± 0.002) and (0.005 ± 0.004) and δ_p(875 nm) equal to (0.025 ± 0.007) and (0.027 ± 0.007). These values of $\delta_p(\lambda)$ are in good accordance with those determined by Pueschel *et al.* [5] at the high-level station of Mauna Loa Observatory (about 3400 m a.m.s.l.), Hawaii (19° 32′ N; 155° 34′ W), during the period from March to May 1991, giving average estimates of δ_p(380 nm) equal to (0.009 ± 0.005), of δ_p(500 nm) equal to (0.011 ± 0.004) and of δ_p(875 nm) equal to (0.010 ± 0.004). It is interesting to notice that both our measurements of $\delta_p(\lambda)$ in summer 1990 and the Mauna Loa measurements are appreciably greater than the SAGE measurements of the stratospheric aerosol optical depth, as obtained in terms of average values relative to the period from July to September in the four years from 1988 to 1991. In fact, the average SAGE values were found to be equal to about 0.007 at 380 nm wavelength, to 0.005 at 500 nm and to 0.002 at 875 nm (P. Russell, private comunication), clearly indicating that a considerable part of the background aerosol optical depth is produced by tropospheric aerosols in periods of volcanic quiescence. Our values of $\delta_p(\lambda)$ found at the Pyramid Laboratory in September 1990 agree very well with the average values of $\delta_p(\lambda)$ determined by Dutton *et al.* [33] from the sun-photometric measurements taken at the Mauna Loa Observatory during the decade 1982-1992, which give the triplet of mean values of aerosol optical depth $\overline{\delta}_p(380\ \text{nm}) = 0.014$, $\overline{\delta}_p(500\ \text{nm}) = 0.010$ and $\overline{\delta}_p(875\ \text{nm}) = 0.006$, for the three-month period from July to September. Therefore, considering that our background measurements of $\delta_p(\lambda)$ were taken on two days only, while those defined by Dutton *et al.* [33] refer to a ten-year period, we adopted these latter values of $\delta_p(\lambda)$ to represent the background aerosol extinction effects produced along the vertical path of the atmosphere by both tropospheric and stratospheric particles during periods very far from volcanic eruptions.

The mean hourly values of $\delta_p(\lambda)$ were also calculated for twelve clear-sky days during summer 1991 and other twelve measurement days characterized by clear-sky conditions during summer 1992. Following this procedure, we obtained a set of forty-four triplets of mean hourly values of $\delta_p(\lambda)$ in summer 1991, with δ_p(380 nm) ranging between 0.108 and 0.294 and giving a median value of 0.216, δ_p(500 nm) between 0.077 and 0.237 with a median value of 0.155, and δ_p(875 nm) between 0.041 and 0.210 with a median value of 0.108. From the measurements performed during summer 1992, we obtained forty-three triplets of mean hourly values of $\delta_p(\lambda)$ relative to clear-sky conditions. The optical depth δ_p(380 nm) was found to vary between 0.103 and 0.214, giving a median value of 0.156, δ_p(500 nm) between 0.102 and 0.202 with a median value of 0.139 and δ_p(875 nm) between 0.084 and 0.128 with a median value of 0.101. The comparison of these median values with those obtained in summer 1991 clearly indicates that the Pinatubo aerosol cloud was still rather dense above the Himalayas, one year after its formation.

Two examples of daily time-patterns of these parameters are shown in fig. 3, relative to September 19, 1991, and August 5, 1992. On September 19, 1991, δ_p(380 nm) increased slowly from 0.216 to 0.256, while δ_p(500 nm) varied between 0.153 and 0.170 and δ_p(875 nm) between 0.122 and 0.151. On August 5, 1992, δ_p(380 nm) assumed increasing values from 0.129 to 0.189, while δ_p(500 nm) increased from 0.109 to 0.184 and δ_p(875 nm) varied between 0.085 and 0.114. The comparison between the time-patterns of two measurement days almost one year apart gives evidence of the considerable decay which occurred between summer 1991 and summer 1992 of the

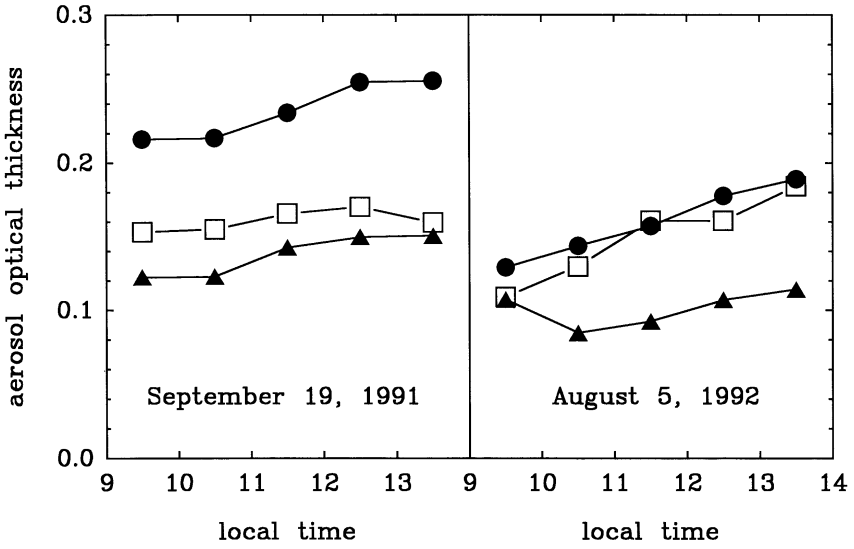

Fig. 3. – Time patterns of the mean hourly values of aerosol optical thickness $\delta_p(\lambda)$ measured on September 19, 1991, and August 5, 1992, at the three window-wavelengths $\lambda = 380$ nm (solid circles) $\lambda = 500$ nm (open squares) and $\lambda = 875$ nm (solid triangles).

extinction features produced by the Pinatubo aerosol cloud. However, all these values of $\delta_p(\lambda)$ are considerably higher than those determined by Elterman [2] at the altitude of 5 km in the free atmosphere, which are $\delta_p(380\ \text{nm}) = 0.072$, $\delta_p(500\ \text{nm}) = 0.052$ and $\delta_p(875\ \text{nm}) = 0.039$. This strong difference suggests that most of the particulate extinction was produced by the Pinatubo aerosol particles also one year after the eruption episodes.

The daily sets of mean hourly values of $\delta_p(\lambda)$ obtained in the two summer periods of 1991 and 1992 were examined in order to calculate the average daily values of $\delta_p(\lambda)$ at the three wavelengths, together with their standard deviations. The time patterns relative to summer 1991 are shown in fig. 4, which gives clear evidence of the fact that the values of optical thickness $\delta_p(\lambda)$ at the three window-wavelengths were rather stable from July 24 to 26, 1991 and then suddenly started to increase on July 27, reaching considerably higher values on July 28. They decreased slightly on July 30 and increased again on August 5. From September 19 to 21, the mean daily values of $\delta_p(\lambda)$ were found to be comparable to those determined on the last days of July, presenting the lowest values on September 24 and the highest on October 2, 1991. In particular, optical thickness $\delta_p(380\ \text{nm})$ assumed values of between 0.177 and 0.200 from July 24 to 26, 1991, while $\delta_p(500\ \text{nm})$ varied between 0.125 and 0.150, and $\delta_p(875\ \text{nm})$ between 0.068 and 0.071. Figure 4 also shows the comparison between the present results and the mean monthly values of $\delta_p(\lambda)$ determined by Pueschel *et al.* [5] from the direct solar irradiance measurements taken in July 1991 at the Mauna Loa Observatory, with the Ames autotracking sun-photometer working at five window-wavelengths. As can be seen, the agreement between our results and the Mauna Loa measurements of the aerosol optical depth at visible and near infrared wavelengths is excellent.

The average values of $\delta_p(\lambda)$ and the standard deviations obtained by us from July

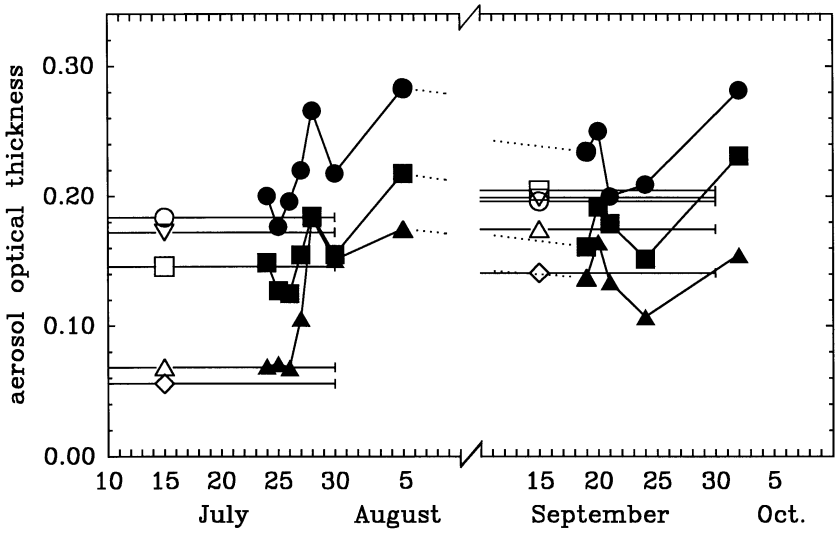

Fig. 4. – Time patterns of the mean daily values of aerosol optical thickness $\delta_p(\lambda)$ measured during summer 1991 at the Pyramid Laboratory. Solid circles give the values of δ_p(380 nm), solid squares those of δ_p(500 nm) and solid triangles those of δ_p(875 nm). The standard deviation of δ_p(380 nm) was found to range between 0.022 and 0.050, that of δ_p(500 nm) between 0.021 and 0.032 and that of δ_p(875 nm) between 0.018 and 0.027. The mean monthly values of $\delta_p(\lambda)$ found by Pueschel *et al.* (1992) at the Mauna Loa Observatory (Hawaii) in July and September 1991 are shown for comparison: open circles refer to δ_p(382 nm), open inverse triangles to δ_p(451 nm), open squares to δ_p(528 nm), open triangles to δ_p(865 nm) and open diamonds to δ_p(1060 nm).

24 to 26, 1991, are $\overline{\delta}_p(380\text{ nm}) = 0.191 \pm 0.042$, $\overline{\delta}_p(500\text{ nm}) = 0.134 \pm 0.035$ and $\overline{\delta}_p(875\text{ nm}) = 0.069 \pm 0.023$. The results in fig. 4 show very clearly that a marked increase in the solar radiation extinction by aerosols occurred at the end of July in the Himalayan region. During the second measurement period from July 27 to August 5, 1991, δ_p(380 nm) was found to vary between 0.215 and 0.283, δ_p(500 nm) between 0.149 and 0.218 and δ_p(875 nm) between 0.101 and 0.183, giving the following average values with their standard deviations: $\overline{\delta}_p(380\text{ nm}) = 0.245 \pm 0.035$, $\overline{\delta}_p(500\text{ nm}) = 0.178 \pm 0.030$ and $\delta_p(875\text{ nm}) = 0.152 \pm 0.037$. These marked variations in the aerosol optical depth $\delta_p(\lambda)$ observed within a very short period of a few days are presumably the consequence of the growth of the volcanic aerosol particles evolving into droplets of larger sizes, consisting of sulphuric acid and liquid water. In the following period from September 19 to October 2, 1991, the mean daily values of optical thickness δ_p(380 nm) were found to range between 0.201 and 0.281, δ_p(500 nm) between 0.149 and 0.231 and δ_p(875 nm) between 0.106 and 0.167, presenting the following average values: $\overline{\delta}_p(380\text{ nm}) = 0.234 \pm 0.033$, $\overline{\delta}_p(500\text{ nm}) = 0.182 \pm 0.032$ and $\overline{\delta}_p(875\text{ nm}) = 0.140 \pm 0.023$, which are thus slightly lower than those evaluated in the previous period. The mean monthly values of $\delta_p(\lambda)$ found by Pueschel *et al.* [5] at the Mauna Loa Observatory in September 1991 are also shown in fig. 4. The comparison with our results suggests that the July-to-September variations in the spectral dependence features of $\delta_p(\lambda)$, as found at the two stations, present some appreciable discrepancies,

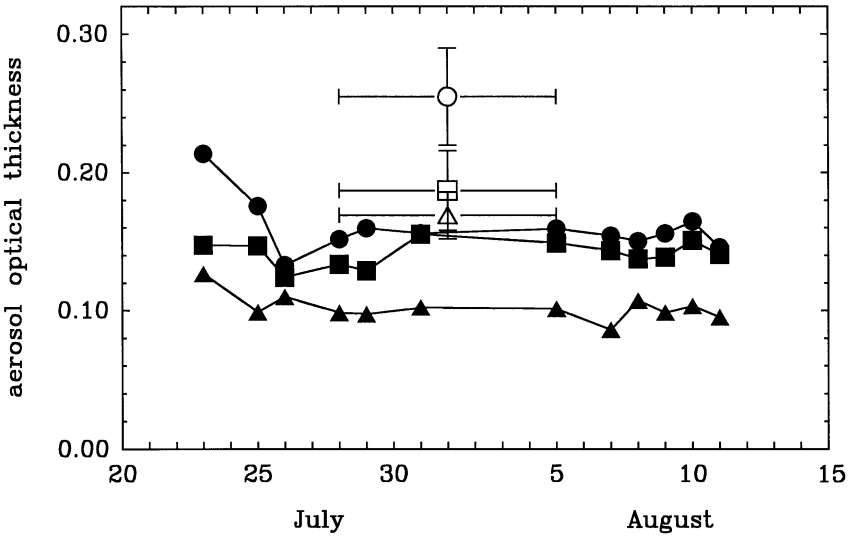

Fig. 5. – Time patterns of the mean daily values of aerosol optical thickness $\delta_p(\lambda)$ measured during summer 1992 at the Pyramid Laboratory. Solid circles refer to $\delta_p(380\ \text{nm})$, solid squares to $\delta_p(500\ \text{nm})$ and solid triangles to $\delta_p(875\ \text{nm})$. The standard deviation of $\delta_p(380\ \text{nm})$ was found to range between 0.020 and 0.060, that of $\delta_p(500\ \text{nm})$ between 0.017 and 0.034 and that of $\delta_p(875\ \text{nm})$ between 0.018 and 0.025. The mean values of $\delta_p(\lambda)$ calculated from the measurements taken during the period from July 28 to August 5 of the previous year are shown for comparison together with their standard deviations: the open circle refer to $\delta_p(380\ \text{nm})$, the open square to $\delta_p(500\ \text{nm})$ and the open triangle to $\delta_p(875\ \text{nm})$.

presumably due to the different evolutionary features of the stratospheric aerosol particle size distribution at the two sites.

Following the same procedure adopted for analysing the measurements taken during summer 1991, we calculated the mean daily values of $\delta_p(\lambda)$ and their standard deviations from the measurement set collected in summer 1992. The results are shown in fig. 5, where a comparison is made between the mean daily values of $\delta_p(\lambda)$ found in summer 1992 and the average values of $\delta_p(\lambda)$ determined during the period from July 27 to August 5, 1991, that is, one year earlier. In summer 1992, aerosol optical thickness $\delta_p(380\ \text{nm})$ was found to vary between 0.133 and 0.214, $\delta_p(500\ \text{nm})$ between 0.124 and 0.155 and $\delta_p(875\ \text{nm})$ between 0.086 and 0.127. Correspondingly, we found the following average values: $\overline{\delta}_p(380\ \text{nm}) = 0.160 \pm 0.020$, $\overline{\delta}_p(500\ \text{nm}) = 0.141 \pm 0.009$ and $\overline{\delta}_p(875\ \text{nm}) = 0.102 \pm 0.010$. The comparison clearly indicates that the aerosol optical thickness appreciably decreased (by about 30% on average) during the period of twelve months from August 1991 to August 1992.

5. – Time variations of atmospheric turbidity parameters

When applied to the scattering processes of visible and infrared radiation by aerosol particle polydispersions, the Mie theory clearly indicates that the optical depth produced by the aerosol particles suspended in the vertical atmospheric column of unit cross-section varies as a function of wavelength, presenting spectral dependence

features depending on several parameters, such as those determining the particle size distribution curve and those defining the complex refractive index of particulate matter [34]. In general, the average slope of the logarithm of aerosol optical thickness $\delta_p(\lambda)$ plotted as a function of the logarithm of wavelength within the visible and near infrared spectral range assumes values ranging between -0.3 and 2.0 for all the natural and anthropogenic aerosol particle polydispersions of different origins [35]. In particular, negative values of this slope coefficient are commonly found for aerosol particle polydispersions in which the extinction effects are predominantly produced by large and giant particles mostly consisting of liquid water, while values higher than 1.5 refer to cases where the aerosol polydispersions consist mostly of very small particles only and the predominant extinction effects are given by the Aitken nuclei. Thus, as suggested by Ångström [3], a spectral series of optical thickness $\delta_p(\lambda)$ can be realistically represented in terms of the following formula:

$$\delta_p(\lambda) = \beta \lambda^{-\alpha}, \tag{5}$$

where wavelength λ is measured in μm. The physical meaning of the two parameters α and β, usually called atmospheric turbidity parameters, is as follows:

1) parameter α gives a general idea of the spectral dependence features of particulate extinction, since it closely depends on the shape of the particle size distribution of aerosol particles, and

2) parameter β is the best-fit value of $\delta_p(\lambda)$ at 1 μm wavelength, which substantially depends on the total number of aerosol particles, the particle size distribution shape parameters and the complex refractive index of particulate matter.

Therefore, the atmospheric turbidity parameters α and β are subject to variation in all cases where the aerosol particle polydispersion in the vertical atmospheric column varies in number concentration, in the shape of the size distribution curve and/or in the physico-chemical characteristics of the particulate matter.

Thus, the large variations in $\delta_p(\lambda)$ measured from July 26 to 28, 1991 are expected to be accompanied also by marked changes in the atmospheric turbidity parameters α and β, as a consequence of the growth of the Pinatubo aerosol particles. These were already present above the Himalayan region from the second week of July. In fact, the size-distribution curve of fresh volcanic particles is usually characterized by multimodal features, with one or two modes of large particles produced by gas-to-particle conversion processes causing the growth of the aerosol particles. Thus, such modes become particularly pronounced within one or two months after the injection of sulphur gases into the stratosphere, as clearly demonstrated by Hofmann and Rosen [36] and Oberbeck *et al.* [37] who examined the aerosol size spectra and the composition characteristics of the volcanic particles forming the El Chichòn aerosol cloud. As a result of the growth of the aerosol particles, parameter α decreases considerably according to the Mie theory, while parameter β increases simultaneously to a considerable extent.

The arrival of the Pinatubo cloud over the Himalayan area in the first half of July 1991 is well confirmed by both satellite observations [38] and a simulation study of the cloud motions based on the dynamical characteristics of the low stratosphere [39]. In fact, most of the volcanic particulate matter and gaseous sulphur dioxide was posi-

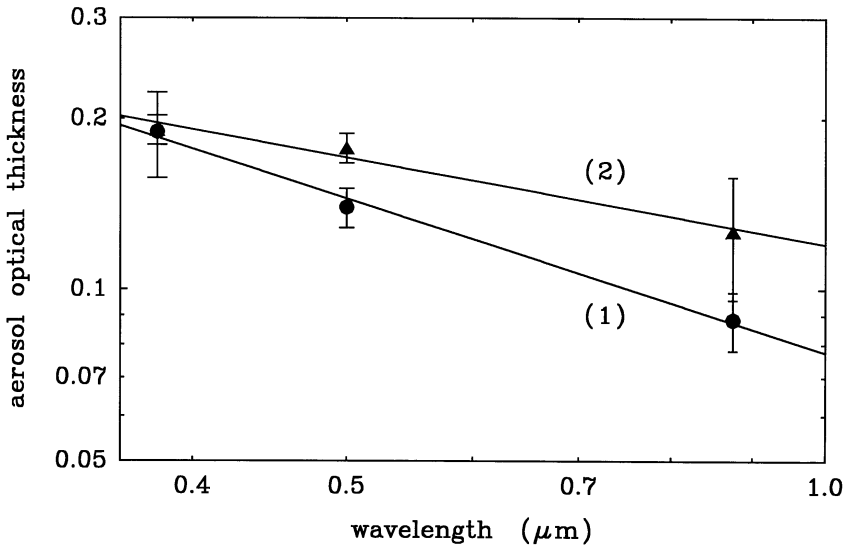

Fig. 6. – Comparison between two spectral series of aerosol optical thickness $\delta_p(\lambda)$ given with their standard deviations: the first (solid circles) was measured on July 24, 1991 (08: 28 LT) and the second (solid triangles) on September 21, 1991 (09: 24 LT). The best-fit lines found in terms of Ångström's [3] formula are defined by the following pairs of atmospheric turbidity parameters: 1) $\alpha = 0.900$ and $\beta = 0.078$ (regression coefficient equal to -0.997), and 2) $\alpha = 0.508$ and $\beta = 0.120$ (regression coefficient equal to -0.987).

tioned at altitudes of between 20 and no more than 30 km [40]. Volcanic debris and sulphur gases were transported westward during the first days after the eruptions, while the Pinatubo cloud gradually expanded reaching a meridional width of about 30° on the last days of June 1991, and the latitude of 30° N on the first days of July 1991. On July 15, the cloud occupied a zonal area of about 120° width, while gas-to-particle conversion processes probably started to take place between mid-July and mid-August, leading to the gradual formation and growth of large solution droplets. During these weeks, mainly in the altitude range from 20 to 25 km, the volcanic cloud was subject to a gradual zonal spreading, so that a fairly continuous band of dense aerosol particle clouds reached the Himalayas during July. Subsequently, the cloud of large aerosol particles circled the globe in the equatorial zone, occupying this region throughout the summer and later [40, 41]. Our measurements of $\delta_p(\lambda)$ from the last week of July to early October 1991 agree very well with the evolutionary patterns of the Pinatubo aerosol cloud described above. In order to verify whether the variations in the atmospheric transmittance characteristics are reasonably consistent with the evolutionary features of the volcanic aerosol particle cloud, we calculated the best-fit values of atmospheric turbidity parameters α and β for each spectral series of $\delta_p(\lambda)$ obtained during the summer periods of 1991 and 1992, in terms of the Ångström [3] formula given in eq. (5). Two examples of Ångström's [3] best-fit lines are shown in fig. 6. The first example refers to one of the spectral series of $\delta_p(\lambda)$ measured on the morning of July 24, 1991, giving α equal to 0.900 and β to 0.078. The second example shows a spectral series of $\delta_p(\lambda)$ measured on September 19, 1991, in the presence of a denser cloud of volcanic aerosol particles. The best-fit value of α is equal to 0.508 and hence

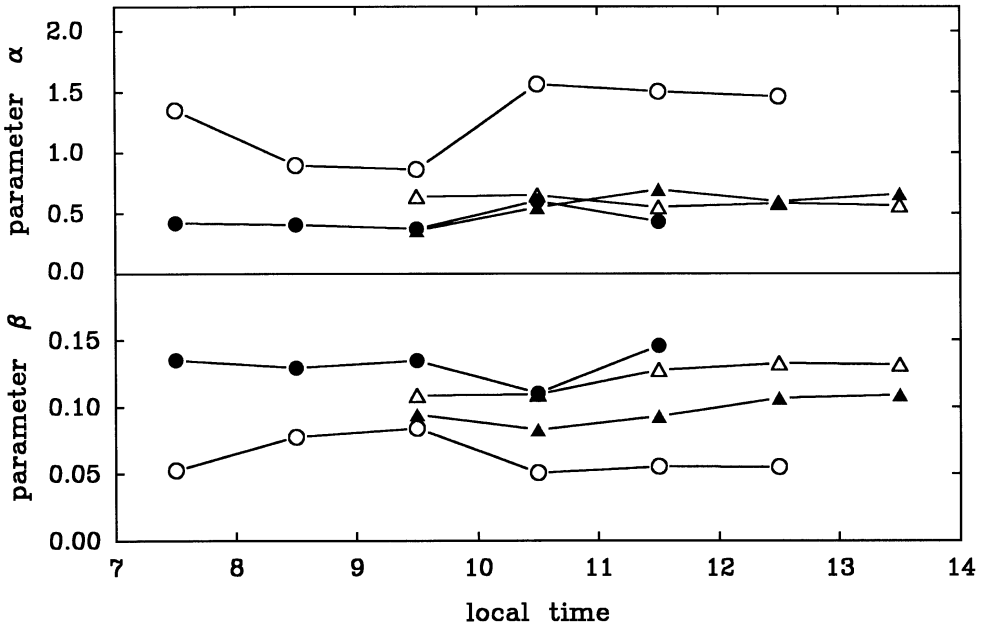

Fig. 7. – Time patterns of the mean hourly values of atmospheric turbidity parameters α and β found on July 24, 1991 (open circles) July 30, 1991 (solid circles) September 19, 1991 (open triangles) and August 10, 1992 (solid triangles). This comparison between measurements of parameters α and β carried out during different stages of the growth of the Pinatubo aerosol particle cloud provides clear evidence of the sharp changes in the atmospheric turbidity conditions which can be caused by the volcanic aerosol particle cloud within a few days as well as over a long period of about one year.

appreciably lower than that of the first example, while β is equal to 0.120, far higher than the previous one. These evaluations show that β almost doubled over the summer 1991, while α decreased very sharply as a consequence of the marked growth of the Pinatubo aerosol particles, producing gradually more intense extinction effects on the incoming solar radiation.

The mean hourly values of α and β were calculated for all the clear-sky days in the summer periods of 1991 and 1992. Figure 7 shows the time patterns of both parameters, as found on four measurement days, July 24, July 30 and September 19 of 1991 and August 10, 1992. These results clearly confirm that large variations in the particulate extinction features were produced within relatively short time intervals of a few days by the Pinatubo aerosol cloud as well as over longer time intervals.

The mean daily values of atmospheric turbidity parameters were also calculated for the two field measurement periods in 1991 and 1992. The results obtained during summer 1991 are shown in the left part of fig. 8. Parameter β varied between 0.057 and 0.061 from July 24 to 26, 1991, and increased rapidly during the subsequent days, reaching values of 0.093 on July 27, 0.165 on July 28, 0.138 on July 30 and 0.160 on August 5. Correspondingly, α varied between 1.13 and 1.36 from July 24 to 26 and suddenly decreased to values of 0.85 on July 27, 0.39 on both July 28 and 30 and 0.57 on August 5. Both turbidity parameters appear to be more stable during the second half of

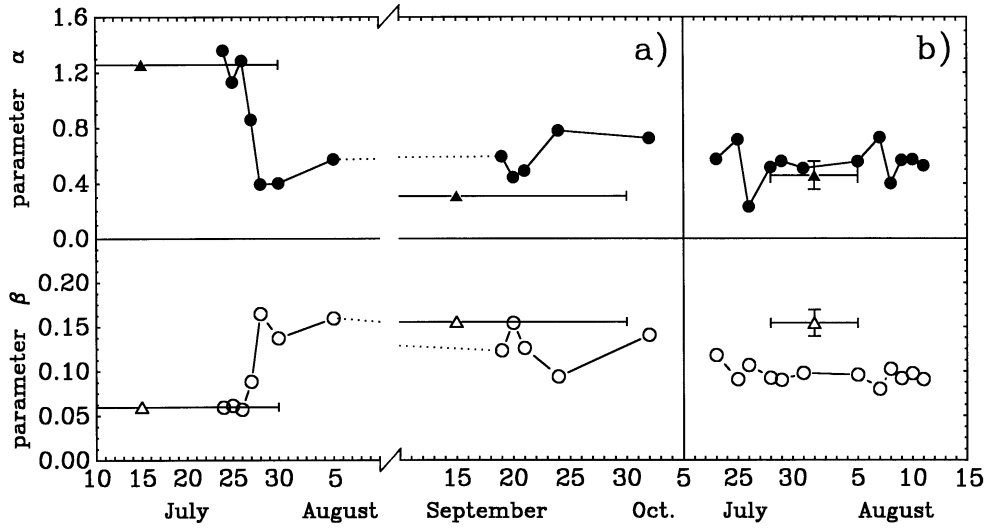

Fig. 8. – Comparison between the time patterns of the mean daily values of atmospheric turbidity parameters α (solid circles) and β (open circles) found during summer 1991 (in the left part) and summer 1992 (in the right part) at the Pyramid Laboratory. In part *a*), the present results are compared with the mean monthly values of α and β (triangles) as obtained from the spectral series of the mean monthly values of $\delta_p(\lambda)$ determined by Pueschel *et al.* [5] in July and September 1991 at the Mauna Loa Observatory. In part *b*), the mean daily values of α and β found during summer 1992 are compared with the mean values $\alpha = 0.456 \pm 0.102$ and $\beta = 0.154 \pm 0.015$ determined from the measurements taken by us in the previous year, during the period from July 28 to August 5.

September, since β fluctuated between 0.10 and 0.15 from September 19 to October 2, 1991, giving an average value of 0.13 ± 0.02, and α varied between 0.47 and 0.80, presenting an average value of 0.62 ± 0.14.

Examining the spectral series of the average values of $\delta_p(\lambda)$ for the period from July 24 to 26, 1991, we obtained the best-fit values $\alpha = 1.207$ and $\beta = 0.059$. These results are in good agreement with the best-fit values $\alpha = 1.257$ and $\beta = 0.060$, obtained from the mean monthly values of $\delta_p(\lambda)$ at five window-wavelengths, as measured by Pueschel *et al.* [5] in July 1991 at the Mauna Loa Observatory. The comparison reveals that very similar turbidity features were present in the atmospheric transparency spectrum at the two high-level stations during the first stage of formation of the Pinatubo cloud. Examining the average values of $\delta_p(\lambda)$ from July 28 to August 5, 1991, we found the best-fit values $\alpha = 0.456$ and $\beta = 0.154$. Moreover, from the spectral series of the average values of $\delta_p(\lambda)$ for the period from September 19 to October 2, 1991, we obtained the best-fit values $\alpha = 0.605$ and $\beta = 0.126$. By way of comparison, the best-fit values determined from the spectral series of the average monthly values of $\delta_p(\lambda)$ given by Pueschel *et al.* [5] in September 1991 are $\alpha = 0.308$ and $\beta = 0.156$. The comparison between our results and the mean monthly values of α and β found at the Mauna Loa Observatory is also shown in fig. 8, suggesting that the daily values of α at the Pyramid Laboratory were appreciably higher and those of β considerably smaller than the average values obtained at the

Mauna Loa Observatory. On the basis of these remarks, we can reasonably state that the Pinatubo aerosol cloud above the Himalayan area in general consisted of a polydispersion of solution droplets presenting appreciably smaller sizes.

Following the same procedure adopted for the data-set of summer 1991, we also examined the spectral measurements of $\delta_p(\lambda)$ carried out during summer 1992. The time patterns of the mean daily values of α and β are shown in the right part of fig. 8, together with the mean values of α and β found at the Pyramid Laboratory in the period from September 19 to October 2 of the previous year. Parameter β was found to vary between 0.087 and 0.112 from July 23 to August 11, 1992, presenting a mean value of 0.099 with a standard deviation of 0.008. Correspondingly, parameter α was found to vary between 0.41 and 0.74 with a mean value of 0.55 and a standard deviation of 0.09. The comparison between the time patterns of α and β relative to the two summer periods clearly shows that β decreased slowly from September 1991 to August 1992 by more than 50% over one year, while parameter α changed on the average by less than 20%. This behaviour suggests that no substantial modifications in the size distribution curve of the large particles occurred during such a long period, while the number concentration was considerably diminished.

6. – Discussion of the particulate extinction measurements

In order to give a realistic measure of the strength of the extinction effects caused during the summer periods of 1991 and 1992 by the Pinatubo aerosol cloud in the Himalayan region, it may be useful to compare the atmospheric turbidity measurements taken at the Pyramid Laboratory with those carried out by Tomasi [42] at the Italian station of Mt. Cimone (2165 m a.m.s.l.) during autumn 1982, when the El Chichòn aerosol cloud was present over Southern Europe. In this context, we defined the ranges of both parameters α and β to show that a similarity exists between the spectral features of particulate extinction presented by the two volcanic aerosol clouds after the growth of the aerosol particles. Thus, we calculated the relative frequency histograms of the mean hourly values of parameters α and β for the following four measurement sets:

a) the first set, consisting of the measurements taken at the Pyramid Laboratory from July 24 to 26, 1991, that is on the three days preceeding the marked growth of the Pinatubo aerosol particles;

b) the second set, consisting of the sun-photometric measurements taken at the Pyramid Laboratory from July 28 to October 2, 1991, in the presence of the Pinatubo cloud containing grown solution droplets of liquid water and sulphuric acid;

c) the third set, consisting of all the measurements taken at the Pyramid Laboratory from July 23 to August 11, 1992, in the presence of the aged cloud of Pinatubo aerosol particles;

d) the fourth set, consisting of the measurements carried out by Tomasi [42] using a pair of Volz sun-photometers during autumn 1982 in the presence of the El Chichòn cloud.

The relative frequency histograms of parameters α and β, as obtained for these four sets of measurements are shown in fig. 9, while the quartiles of the histograms are given in table I. The shape of the histogram of parameter α in part *a*) of fig. 9 greatly

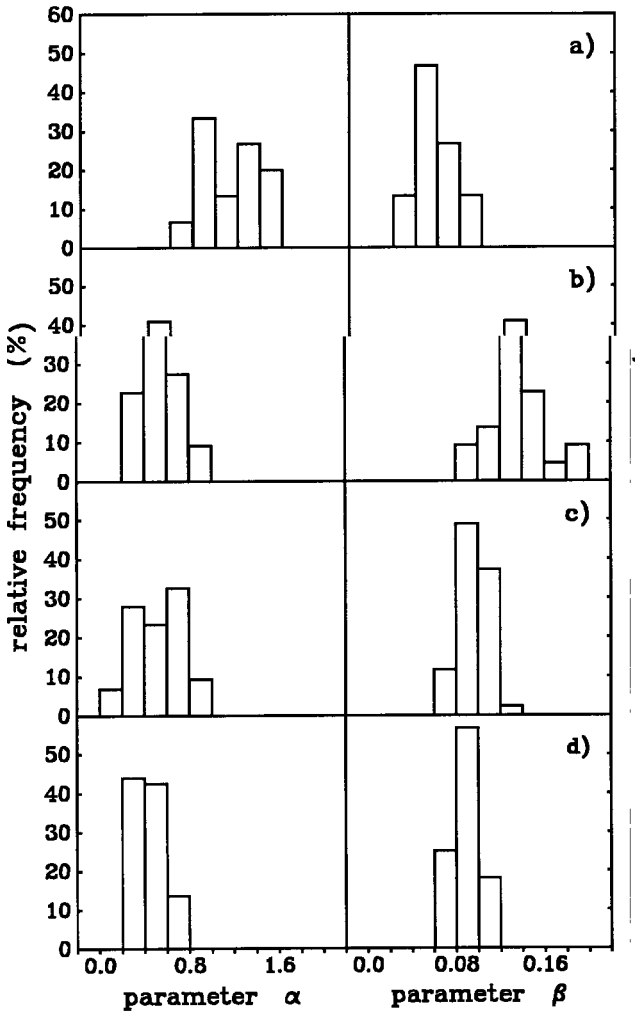

Fig. 9. – Relative frequency histograms of the mean hourly values of atmospheric turbidity parameters α and β, found for three different sets of measurements taken at the Pyramid Laboratory. The histograms in part *a*) refer to the measurements carried out from July 24 to 26, 1991, before the growth of the volcanic aerosol droplets clearly observed at the end of July. The histograms in part *b*) refer to the measurements taken from July 28 to October 2, 1991. The histograms in part *c*) refer to the measurements taken during summer 1992, that is in the presence of the aged cloud of Pinatubo aerosol particles. Part *d*) shows for comparison the histograms obtained from the measurements carried out at the Mt. Cimone (Italy) during autumn 1982, in the presence of the El Chichòn cloud [42].

differs from the others. In fact, α covers the intervals from 0.6 to 1.6 in set *a*), from 0.2 to 1.0 in sets *b*) and *c*), and from 0.2 to 0.8 in set *d*). This clearly demonstrates that set *a*) of measurements concerns the initial stage of formation of the Pinatubo solution droplets, while the other sets refer to polydispersions of larger aerosol particles.

TABLE I. – *Quartiles of the relative frequency histograms of atmospheric turbidity parameters α and β for the three data sets relative to different growth stages of the Pinatubo aerosol cloud and the fourth data set pertinent to the El Chichòn aerosol cloud observed in Italy during the autumn 1982.*

	Pinatubo cloud growth stages			*d*) El Chichòn cloud
	a) July 24-26 1991	*b*) July 28-October 2 1991	*c*) summer 1992	
Parameter α:				
1st quartile	0.91	0.41	0.35	0.30
2nd quartile	1.15	0.53	0.55	0.45
3rd quartile	1.36	0.68	0.67	0.54
Parameter β:				
1st quartile	0.049	0.121	0.086	0.080
2nd quartile	0.057	0.133	0.096	0.089
3rd quartile	0.074	0.146	0.106	0.098

Accordingly, table I shows that all the quartiles of α for sets *b*), *c*) and *d*) are appreciably smaller than those relative to set *a*).

Figure 9 also shows that parameter β ranges between 0.02 and 0.10 in set *a*), between 0.08 and 0.20 in set *b*), in the presence of grown volcanic aerosol particles, and between 0.06 and 0.14 in set *c*), relative to the aged cloud of Pinatubo aerosol particles. The histogram of β found for the El Chichòn cloud presents values ranging between 0.06 and 0.12. The comparison in fig. 9 clearly indicates that a strong decay in the optical density of the Pinatubo aerosol cloud occurred from summer 1991 to summer 1992 above the Himalayan region. Moreover, it can be pointed out that the turbidity conditions produced by the Pinatubo aerosol cloud in summer 1991 were considerably denser than those produced by the El Chichòn aerosol cloud over Southern Europe about six months after the eruption. As a result of ageing and decay processes during summer 1992, the Pinatubo aerosol cloud showed features of atmospheric turbidity comparable with those observed in Italy in the autumn of 1982 in the presence of the El Chichòn aerosol cloud. These considerations are confirmed by the results in table I, which show that the quartiles of the histogram of β defined for measurement set *a*) are considerably smaller than the corresponding values found for the other three sets, indicating that the scattering effects associated with the Pinatubo cloud became important only after the marked growth of the aerosol particles. Moreover, table I shows that the quartiles of β relative to set *b*) are all greater than those determined for sets *c*) and *d*), demonstrating that the extinction effects produced by the Pinatubo aerosol cloud during its first year of life were considerably stronger than those caused by the El Chichòn cloud. The quartiles of β defined for sets *c*) and *d*) were found to be very similar, confirming that the extinction effects produced by the Pinatubo cloud during the second summer above the Himalayan region were of comparable intensity to those caused by El Chichòn cloud during summer and autumn 1982 over Italy. Thus, we can state that the Pinatubo aerosol cloud present in summer 1991 above the Himalayas produced more intense extinction effects of the visible solar radiation than the El

Chichòn aerosol cloud over Southern Europe during the second half of 1982 [42, 43]. The results shown in fig. 9 and table I give a clear picture of the variations in α and β produced by the ageing of the volcanic aerosol particles suspended in the atmosphere, such as the growth of the aerosol droplets, the decay in the particle number density and the decrease in the cloud depth. The ageing of the Pinatubo cloud was certainly accompanied by variations in the mass loading of the volcanic aerosol particles. This matter is examined in the following section.

7. – Evaluations of vertical aerosol mass loading

As mentioned above, we determined the values of the aerosol optical thickness $\delta_p(\lambda)$ at the Pyramid Laboratory on two clear-sky days of summer 1990, in the absence of volcanic aerosol particle layers suspended in the stratosphere. These results indicate that $\delta_p(\lambda)$ varied between 0.005 and 0.038 at the three window-wavelengths of the Volz sun-photometers, when the background aerosol particles present in the stratosphere form very thin layers producing aerosol optical thicknesses of less than 0.01 at visible wavelengths. In fact, the background particle population in the vertical atmospheric column mainly consists of continental and wind-borne particles and, to a much smaller extent, stratospheric particles of non-volcanic origin. The latter form the so-called Junge aerosol layers at altitudes usually ranging from 16 to 22 km in the mid-latitude areas. These rather small particles are largely composed of sulphate substances (producing only slight absorption of solar radiation) and present very low number concentrations of no more than $5\,\mathrm{cm}^{-3}$ [5]. Therefore, they generally produce optical depths smaller than 0.01 at all the visible wavelengths. We decided to assume that the values of the background aerosol optical thickness at the Pyramid Laboratory without the Pinatubo aerosol cloud are equal to those measured on the average at the Mauna Loa Observatory during the decade 1982-1992 [33], *i.e.* $\delta_p(380\ \mathrm{nm}) = 0.014$, $\delta_p(500\ \mathrm{nm}) = 0.010$ and $\delta_p(875\ \mathrm{nm}) = 0.006$. Thus, we evaluated the mean daily values of the aerosol optical thickness $\delta_s(\lambda)$ due only to the Pinatubo aerosol particles present in the stratosphere as differences between the mean daily values of $\delta_p(\lambda)$ measured during the summer periods of 1991 and 1992 at the Pyramid Laboratory and the Mauna Loa [33] background values mentioned above. Since the background values of $\delta_p(\lambda)$ are very small compared to the total values of $\delta_p(\lambda)$, the values of $\delta_s(\lambda)$ were found to be slightly smaller than those of $\delta_p(\lambda)$ determined directly from the sun-photometric measurements. Therefore, the spectral series of volcanic aerosol optical thickness $\delta_s(\lambda)$ were found to present extinction features characterized by atmospheric turbidity parameters very similar (almost identical) to those shown in fig. 8. Thus, the spectral series of the mean daily values of $\delta_s(\lambda)$ obtained during summer 1991 was characterized by values of α ranging between 0.39 and no more than 1.36. In order to obtain size distribution models of the volcanic aerosol particles fitting the spectral extinction features of $\delta_s(\lambda)$ observed during summer 1991, we decided to determine for each spectral series the best-fit solution based on a linear combination of the two following stratospheric particle models defined by Pueschel *et al.* [5]:

1) The background aerosol model, which was determined from particle samples collected at 18 km altitude. It is based on a log-normal size distribution curve of spherical particles presenting the particle number concentration of $3.5\,\mathrm{cm}^{-3}$, mode radius of $0.07\,\mu$m, standard deviation equal to the natural logarithm of 1.8 and particle volume equal to $0.024\,\mu\mathrm{m}^3$ per cubic centimeter of air. Using the volume

extinction coefficients calculated by Pueschel *et al.* [5] (Pueschel, private communication), we determined the volume extinction coefficient $c(\lambda)$ at the three sun-photometric wavelengths, normalized to the unit particle concentration. Thereupon, we calculated three coefficients $b(\lambda)$ as products of $c(\lambda)$ by the unit volume of air, which have in practice the dimensions of the extinction cross-section (relative to the mean particle) and are measured in cm^2. Their values are $b_1(380\ \mathrm{nm}) = 5.734 \cdot 10^{-10}\ \mathrm{cm}^2$, $b_1(500\ \mathrm{nm}) = 4.043 \cdot 10^{-10}\ \mathrm{cm}^2$ and $b_1(875\ \mathrm{nm}) = 1.466 \cdot 10^{-10}\ \mathrm{cm}^2$. Moreover, assuming that the mean mass density of stratospheric aerosol particles is equal to $1.65\ \mathrm{g\ cm}^{-3}$, as proposed by Hofmann and Rosen [36] for spherical aerosol particles consisting of 75% sulphuric acid and 25% liquid water, we found that the average particle mass of this background model is $m_1 = 1.131 \cdot 10^{-14}\ \mathrm{g}$.

2) The bimodal model for fresh Pinatubo aerosol particles, which was defined taking into account the size-distribution shape, the morphological characteristics and the physico-chemical features of the volcanic particles sampled with wire impactors mounted on aircrafts flying in August 1991 over a very wide area and between 16.5 and 20.1 km heights [5]. The size distribution curve is given by two log-normal curves, having particle number concentrations of 1.7 and $1.2\ \mathrm{cm}^{-3}$, mode radii of 0.09 and 0.31 μm, standard deviations both equal to the natural logarithm of 1.5 and particle volumes equal to 0.013 and $0.32\ \mu\mathrm{m}^3/\mathrm{cm}^3$, respectively. From the volume extinction coefficients given by Pueschel (private communication) (see also [5]), we calculated the values of the three coefficients $b(\lambda)$ for the bimodal size distribution curve normalized to an overall particle concentration of $1\ \mathrm{cm}^{-3}$, that is $b_2(380\ \mathrm{nm}) = 5.155 \cdot 10^{-9}\ \mathrm{cm}^2$, $b_2(500\ \mathrm{nm}) = 5.562 \cdot 10^{-9}\ \mathrm{cm}^2$ and $b_2(875\ \mathrm{nm}) = 4.597 \cdot 10^{-9}\ \mathrm{cm}^2$. Adopting a mean value of particulate matter density equal to $1.65\ \mathrm{g\ cm}^{-3}$ [36], we found that the average particle mass m_2 of the bimodal model is equal to $1.895 \cdot 10^{-13}$ g.

Thus, for each spectral series of $\delta_s(\lambda)$, we considered the following system of three equations:

$$\begin{cases} N_1 b_1(380\ \mathrm{nm}) + N_2 b_2(380\ \mathrm{nm}) = \delta_s(380\ \mathrm{nm}), & (6a) \\ N_1 b_1(500\ \mathrm{nm}) + N_2 b_2(500\ \mathrm{nm}) = \delta_s(500\ \mathrm{nm}), & (6b) \\ N_1 b_1(875\ \mathrm{nm}) + N_2 b_2(875\ \mathrm{nm}) = \delta_s(875\ \mathrm{nm}) & (6c) \end{cases}$$

and determined the best-fit values of N_1 and N_2, which are the total particle numbers of the background aerosol model and the bimodal model in the vertical atmospheric column of cross-section equal to $1\ \mathrm{cm}^2$. Following this best-fit procedure, we found that N_1 varied between $2.2 \cdot 10^8$ and $2.8 \cdot 10^8\ \mathrm{cm}^{-2}$ from July 24 to 27, 1991, between $1.1 \cdot 10^8$ and $2.0 \cdot 10^8\ \mathrm{cm}^{-2}$ from July 28 to August 5, 1991, and between $1.1 \cdot 10^8$ and $2.5 \cdot 10^8\ \mathrm{cm}^{-2}$ from September 19 to October 2, 1991. Correspondingly, N_2 was found to vary between $2.8 \cdot 10^6$ and $1.1 \cdot 10^7\ \mathrm{cm}^{-2}$ from July 24 to 27, 1991, between $2.4 \cdot 10^7$ and $2.8 \cdot 10^7\ \mathrm{cm}^{-2}$ from July 28 to August 5, 1991, and between $1.3 \cdot 10^7$ and $2.8 \cdot 10^7\ \mathrm{cm}^{-2}$ from September 19 to October 2, 1991. Thereupon, the mean daily values of the Pinatubo aerosol mass loading M_s were calculated very simply as the sum of the two contributions given by the two models, that is

$$M_s = N_1 m_1 + N_2 m_2 . \qquad (7)$$

Following this procedure, we obtained the values of M_s shown in the left part of fig. 10. As can be seen, M_s was found to range i) between 0.037 and $0.047\ \mathrm{g\ m}^{-2}$ in the

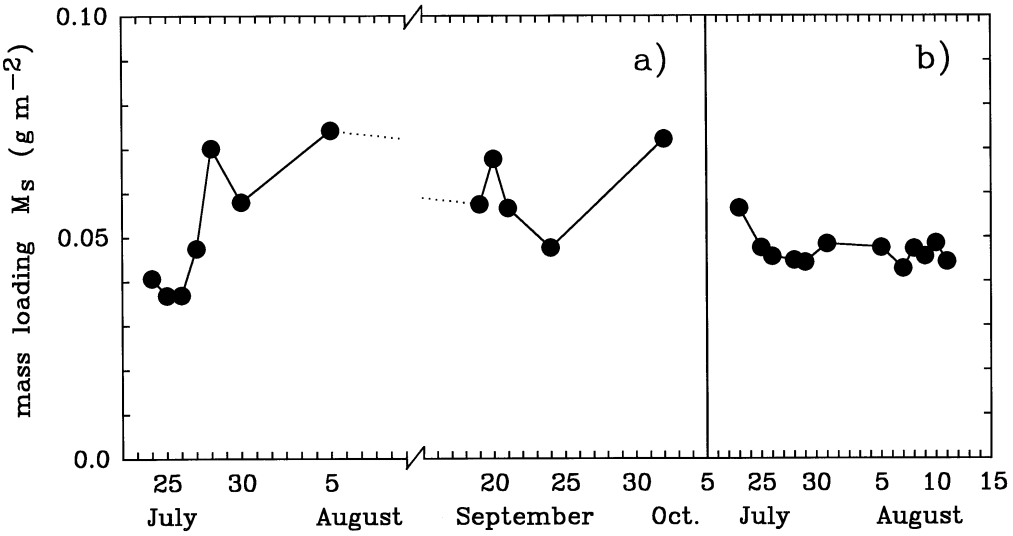

Fig. 10. – Time patterns of the mean daily values of the Pinatubo aerosol mass loading M_s, as determined from the spectral values of aerosol optical depth $\delta_s(\lambda)$ found during summer 1991 (in the left part) and summer 1992 (in the right part). In part *a*), the best-fit values of M_s were determined in terms of a variable linear combination of the background aerosol model and the bimodal extinction model defined by Pueschel *et al.* [5] for the Pinatubo aerosol cloud of a few-month age. In part *b*), the best-fit values of M_s were determined in terms of a variable linear combination of the background aerosol model and the trimodal model proposed by Pueschel *et al.* [5] for an aged population of Pinatubo aerosol particles.

period from July 24 to 27, 1991, with an average value of $(0.042 \pm 0.006)\ g\ m^{-2}$; ii) between 0.058 and $0.074\ g\ m^{-2}$ in the period from July 28 to August 5, 1991, with an average value of $(0.067 \pm 0.008)\ g\ m^{-2}$; and iii) between 0.048 and $0.072\ g\ m^{-2}$ from September 19 to October 2, 1991, with an average value of $(0.060 \pm 0.010)\ g\ m^{-2}$. These results agree very well with the values of M_s found by Valero and Pilewskie [44] to range between 0.035 and $0.080\ g\ m^{-2}$ in July 1991 over the Caribbean area and those found by Russell *et al.* [45] at the Mauna Loa Observatory, giving average monthly values of M_s equal to $(0.048 \pm 0.007)\ g\ m^{-2}$ in July 1991 and $(0.091 \pm 0.027)\ g\ m^{-2}$ in August-September 1991.

The spectral series of the mean daily values of $\delta_s(\lambda)$ measured during summer 1992 were found to be characterized by values of α ranging between 0.23 and 0.73, with a median value of 0.56. The best-fit procedure described above was applied using a linear combination of the background aerosol model [5] normalized to the unit particle concentration and described above and the trimodal model for aged Pinatubo aerosol particles proposed by Pueschel *et al.* [5] on the basis of volcanic particle samples collected during the first months of 1992 at altitudes ranging between 10.5 and 12 km height. The size distribution curve of this aerosol extinction model was found to be given by a fixed linear combination of three modes, which had particle number concentrations of 21.9, 6.2 and $0.8\ cm^{-3}$, mode radii of 0.06, 0.18 and $0.75\ \mu m$, standard deviations equal to the natural logarithms of 1.4, 1.5 and 1.2, and particle volumes equal to 0.036, 0.36 and 1.21 $\mu m^3/cm^3$, respectively. Normalizing the volume extinction

coefficients of this trimodal model [5] to the unit particle concentration, we calculated the following values of the extinction coefficient: $b_3(380\ \text{nm}) = 2.333 \cdot 10^{-9}\ \text{cm}^2$, $b_3(500\ \text{nm}) = 2.163 \cdot 10^{-9}\ \text{cm}^2$ and $b_3(875\ \text{nm}) = 2.162 \cdot 10^{-9}\ \text{cm}^2$. On this basis and assuming the average particle mass density to be equal to $1.65\ \text{g cm}^{-3}$ [36], we found a value of the average particle mass $m_3 = 9.169 \cdot 10^{-14}$ g. Following the best-fit procedure adopted above and based on the eqs. (6*a*), (6*b*) and (6*c*), where subscript 2 has been substituted by subscript 3 relative to the trimodal model, we obtained values of N_1 ranging between $1.9 \cdot 10^7$ and $1.6 \cdot 10^8\ \text{cm}^{-2}$ in the summer 1992, while N_3 was found to range correspondingly between $3.0 \cdot 10^7$ and $4.7 \cdot 10^7\ \text{cm}^{-2}$. The values of M_s were then calculated by summing the mass contributions given by the background aerosol model and the trimodal model. The results are shown in the right part of fig. 10. They present very stable features since M_s was found to range between 0.043 and $0.057\ \text{g m}^{-2}$, with a median value of $0.47\ \text{g m}^{-2}$. The average seasonal value of M_s resulted to be equal to $(0.047 \pm 0.004)\ \text{g m}^{-2}$ and, hence, in close agreement with the mean value of $(0.055 \pm 0.020)\ \text{g m}^{-2}$ found by Russell *et al.* [45] at Mauna Loa, Hawaii, during July 1992.

8. – Conclusions

The Pinatubo cloud consisting of sulphur gases and small aerosol particles probably reached the Himalayan region during the first two weeks of July 1991. The measurements of the aerosol optical thickness carried out at the Pyramid Laboratory from July 24 to 26 show that the aerosol optical depth presented values ranging between 0.07 and 0.20 at the three sun-photometric wavelengths. On the following days, the aerosol optical thickness $\delta_p(\lambda)$ was found to increase considerably, presumably as a result of the rapid growth of volcanic aerosol particles into solution droplets of large sizes. In fact, $\delta_p(\lambda)$ assumed values ranging between 0.10 and 0.26 during the end of July and early August and values ranging between 0.11 and 0.28 during September. The sun-photometric measurements carried out during the second half of July and the first half of August 1992 gave values of the aerosol optical thickness varying between 0.09 and 0.21 at the three window-wavelengths of the Volz sun-photometers. Correspondingly, the time patterns of atmospheric turbidity parameters α and β determined from these sun-photometric measurements of $\delta_p(\lambda)$ show that parameter α assumed values ranging between 1.13 and 1.36 from July 24 to 26, 1991, and started to decrease rapidly on July 27 from 1.28 to 0.85, varying between 0.39 and 0.57 on the subsequent days. Simultaneously, β was found to assume the value of 0.06 on the first three measurement days and then increased to 0.09 on July 27, passing to mean daily values of between 0.14 and more than 0.16 on the following days. During the second half of September, parameter α varied between 0.44 and 0.78 and β between 0.09 and 0.15. The persistence of Ångström's parameter α around relatively low values and the corresponding increase of β are clear proof of the marked extinction effects associated with the growth of the volcanic aerosol particles forming the Pinatubo cloud.

During the summer period from July 23 to August 11, 1992, parameter α ranged between 0.23 and 0.73 and parameter β between 0.08 and 0.12, clearly showing that the extinction features produced by the aged cloud of Pinatubo aerosol particles were still very stable in time after more than a year.

On the basis of linear combinations of the background aerosol model and the bimodal model for a Pinatubo aerosol particle polydispersion, both defined by Pueschel

et al. [5], the vertical mass loading of stratospheric volcanic particles was estimated to be of between 0.037 and 0.041 g m^{-2} during the period from July 24 to 28, 1991, and to increase considerably from a mean daily value of about 0.047 g m^{-2} on July 27 to a value of 0.074 g m^{-2} on August 5, 1991, fluctuating between 0.048 and more than 0.072 g m^{-2} during the second half of September 1991. Using a particulate extinction model, based on linear combinations of the background aerosol model and the trimodal model for aged volcanic aerosol particles [5], the vertical mass loading of the Pinatubo aerosol particles was calculated to vary between 0.043 and 0.057 g m^{-2} during the period from mid-July to mid-August 1992.

The present results show that about six months after the Pinatubo eruption, the vertical mass loading of stratospheric aerosol particles above the Himalayan region can be estimated to have been about 0.05 g m^{-2} and, hence, considerably higher than the vertical aerosol mass loading of around 0.025 g m^{-2} [42], as calculated for the El Chichòn cloud of six months observed above the Italian area in autumn 1982.

The present evaluations of aerosol optical thickness at different wavelengths, turbidity parameters and vertical mass loading of Pinatubo aerosol particles turned out to be in good agreement with the measurements of the same parameters obtained at the Manua Loa Observatory during the summer periods of 1991 and 1992 [5, 33, 45] and in other regions [44]. Therefore, our results can be considered as being of reliable use in calculations of the radiative forcing caused by albedo changes due to the presence of the Pinatubo aerosol cloud in the Himalayan region, during the period from summer 1991 to summer 1992.

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