Variation of Stratospheric Aerosol Content Measured by Laser Radar

Part I Seasonal Variation of Aerosol Content and Flux of Sulfur Compounds from Troposphere into Stratosphere

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(Manuscript received 25 September 1980, in revised form 3 June 1981)*

Abstract

Seasonal variation of stratospheric aerosol content is discussed on the basis of laser radar measurements at Nagoya (35°N, 137°E) Japan. Analytical results suggest that there were noticeable winter maximum of aerosol content, and this was in good agreement with other investigators' measurements (Hofmann *et al.*, 1975; Bigg, 1975; Hofmann and Rosen, 1977; Reiter *et al.*, 1979). In summer season 1977, and possibly 1976, second maximum of aerosol content was suggested in laser radar measurements at Nagoya. Maximum of stratospheric aerosol content in summer season can be found, though it is small, in the analytical results by Gras and Laby (1979) who suggested the second maximum of aerosol content in summer (southern hemisphere) 1975, and by Reiter *et al.* (1979) whose results contained small maximum of loading in summer 1977, and 1978. Summarizing various measurements, the winter maximum of aerosol content is always observed on the global scale, and summer second maximum of loading can be sometimes observed.

Present analytical results suggest that the flux of sulfur compounds of 5×10^7 (S) atoms/cm²/sec from troposphere to stratosphere is necessary to compensate the sedimentational loss of particulate matter if it is assumed that the stratospheric aerosol is composed of sulfuric acid of 75% in weight.

1. Introduction

Since Junge discovered the stratospheric aerosol layer over a decade ago (Junge *et al.*, 1961), various scientific researches have been made in order to make clear the nature of stratospheric aerosol particles. The scientific interest on this problem has been stimulated by recent suggestions that the stratospheric aerosol may exert an important influence on the climate of the earth (Pollack *et al.*, 1976a, 1976b). Consequently there presently exists a fair amount of observational data on the stratospheric aerosols, and these have shown that there exists in the lower stratosphere a world-wide highly structured layer of sulfate containing particles, probably in the form of sulfuric acid, in the 0.1 to 1.0 micron range. But there are considerably less observational informations and agreement concerning such characteristics as seasonal variations, global

distribution, sources or sinks of stratospheric aerosols (Hofmann *et al.*, 1975; Reiter *et al.*, 1979).

Laser radar observations of stratospheric aerosols have been made from August 1975 on clear nights at Nagoya (35°N, 137°E). In this paper, the first of series dealing with results of research program, we should like to present the seasonal change of stratospheric aerosol content using the data during the period from January 1976 to December 1978, relatively quiet time stratosphere, and compare these with previous investigators' measurements (Hofmann *et al.*, 1975; Hirono *et al.*, 1976; Reiter *et al.*, 1979).

The sulfur budget of stratosphere is one of the most essential factor controlling the nature of stratospheric aerosol particles, and strongly affects the density variation of stratospheric aerosols. Therefore it is very useful for the research on the stratospheric aerosol behavior to discuss the

influx of sulfur compounds into stratosphere across the tropopause. The flux of sulfur compounds with gas phase form, such as SO_2 , H_2S , COS , and so on, from troposphere to stratosphere was estimated using analytical results, and estimation shows that the flux of sulfur compounds is about 5×10^7 molecules/cm²/sec under the condition that all of large particles in stratosphere are composed with 75% sulfuric acid solution and this sulfuric acid is produced through photo-oxidational reaction of sulfur compounds transported into stratosphere from troposphere (Hofmann *et al.*, 1974; Cadle, 1975; Turco *et al.*, 1979). This estimation is showing good agreement with other investigators' results (Hofmann *et al.*, 1974; Cadle, 1975; Krey, 1970; Turco *et al.*, 1979).

2. Seasonal change of stratospheric aerosol content

The main characteristics of laser radar system of Nagoya University are listed up in Table 1. This laser radar can emit the laser pulse at wavelength = 0.6943 μm and 1.06 μm alternatively at the repetition rate = 0.2 sec⁻¹, however, analyzed results presented here are due to only data measured at wavelength = 0.6943 μm . More detailed information on the characteristics of laser radar system has been described in other paper (Iwasaka *et al.*, 1976), and we will not repeat all of them. The analysis of measurements by laser radar using photoncounting method in receiver depends on the number of photon pulse counted. It is reasonable to assume that the pulses follow a Poisson distribution in time, so that the standard deviation of measurement corresponds to the square of the number of pulses involved, appropriate allowance being made for background noise. In present case a typical night's measurements using ruby laser are based on about one thousand laser shots, therefore the

statistical error becomes smaller than few per cent at 20 km level.

In order to derive aerosol scattering profiles the expected Rayleigh scattering from the molecular atmosphere must be subtracted from the experimental profiles. For height up to 35 km we have calculated the Rayleigh scattering on the basis of model atmosphere due to U.S. Standard Atmosphere Supplement (1966). Comparison between the lidar profiles and the calculated molecular atmosphere was made by visible curve fitting (*e.g.*, Russell *et al.*, 1976). It was sometimes possible to fit the curve both above and below the height at which any aerosol scattering present was observed, but in case where aerosol scattering was present at the lowest data point, the fitting was done only at height of 30 km or above.

The scattering ratio due to particulate matter, $R(Z)$ is defined by following equation,

$$R(Z) = [B_r(Z) + B_m(Z)] / B_r(Z) \quad (1)$$

where $B_r(Z)$ and $B_m(Z)$ are backscattering coefficient of atmospheric molecules and of aerosol particles at altitude, Z respectively. And backscattering coefficient of aerosol particles is estimated using $R(Z)$ by

$$B_m(Z) = [R(Z) - 1] b_r n(Z) \quad (2)$$

where $n(Z)$ is number density of air molecules at altitude Z , and b_r is backscattering cross-section of molecules at wavelength = 0.6943 μm . Previously some investigators discussed on the procedure to determine the scattering ratio or backscattering coefficient of stratospheric aerosols and related problems with this procedure (Sanford, 1976; Russell *et al.*, 1976; Itabe *et al.*, 1977), then this is not described here.

Laser radar measurements of stratospheric aerosols have been carried out at Nagoya approximately three times a month since August 1975, and this frequency of measurements has proven satisfactory for studying the variation of aerosol content with the time scale of season. The data measured during the period from January 1976 to December 1978 were used for analysis in this paper. This data shows the effect of Mt. Fuego eruption on the stratospheric aerosol content variation, but this is not so serious problem to discuss the seasonal change of aerosol content since we can easily distinguish seasonal component from the over all trend of backscattering coefficient measured in this period (see Figs. 1, 2, 3, and 4).

The running mean value of three months on

Table 1

Laser	Ruby (0.6943 μm), YAG (1.06 μm)
Pulse energy	1 J/pulse (maximum)
Pulse duration	20 nsec
Repetition rate of pulse	0.2 sec ⁻¹
Photomultiplier	40 cm ϕ
Receiver telescope	R649(Ruby), R139(YAG)
Filter band width	10Å(Ruby), 50Å(YAG)
Rotating shutter	24,000 rpm

* Present analysis does not include the measurements at wavelength = 1.06 μm .

Fig. 1. Backscattering coefficient at the peak of stratospheric aerosol layer. The solid curve is three-monthly averaged value and dotted curve is the decay curve of backscattering coefficient gained by least squared method.

Fig. 2. Vertically integrated value of backscattering coefficient in the range, 14 km-24 km. The curve is three-monthly average.

the backscattering coefficient at particle layer peak is shown in Fig. 1. Noticeable seasonal component on the aerosol content variation in the curve of Fig. 1 dominates the temporal variation at Nagoya (35°N , 137°E) and appears to be superimposed on a long term variation which must be connected with the decay processes of aerosol content which increased suddenly due to volcanic material loading into stratosphere by the eruption of Mt. Fuego. The long term decay curve is approximated using dotted curve, and this decay curve suggests that the decay life

Fig. 3. Vertically integrated value of backscattering coefficient in all over the stratosphere (above tropopause height). The curve is three-monthly average.

Fig. 4. Vertically integrated value of backscattering coefficient above the height of 26 km. The curve is three-monthly average.

time of aerosol content is about 2 years. The detailed discussion on the decay of stratospheric aerosol content after the volcanic eruption will be discussed in another paper of this research program (Iwasaka, 1981). The maximum of peak values is found out in winter season '76, '77, and '78 as was already pointed out in the results of balloon soundings by the University of Wyoming's Atmospheric Physics Group during the observation 1972 and 1973 (1975). On the whole maximum in winter season is evidently found out during all observational period and summer maximum seems to decrease year by year. The winter maximum in temporal variation of stratospheric aerosol content can be seen also in integrated backscattering coefficient in the region from 14 km to 24 km (Fig. 2), in integrated value in over all stratosphere (Fig. 3),

and in integrated value above 26 km (Fig. 4). The summer maximum has not been claimed in previous investigations, but apparently found out in measurement at Garmisch-Partenkirchen (47°N, 11°E) from Oct. '76 to June '78 (Reiter *et al.*, 1979). The data due to balloon borne particle counter does not show so large peak as winter maximum in summer season, but apparently small secondary peaks are found in late spring or early summer '72, '73, and '75 in the time series of measured total stratospheric aerosol in a 1 cm^{-2} column (aerosol radius $>0.15 \mu\text{m}$) (Hofmann *et al.*, 1975; Hofmann and Rosen, 1977).

3. Discussion and conclusion

The winter maximum of stratospheric aerosol content indicated by laser radar measurements at Nagoya (35°N, 137°E) is one of the typical tendency found out in the measurement at northern mid latitude during no-disturbed stratosphere. Previously Hofmann *et al.* reported the existence of winter maximum of stratospheric aerosol content on the basis of the measurements using balloon borne optical dust counter at Laramie (41°N, 105°W) (1975). The agreement between laser radar and balloon data revealed in this manner is quite striking. According to Bigg, very similar results have been found in the measurements of number concentration of stratospheric aerosols at Mildura, Australia (34°S, 142°E) during the observational period from May 1969 to April 1974 (1976). In the measurements at Sao Jose dos Campos (23°S, 47°E) the noticeable seasonal component cannot be found during quiet stratospheric period (Clemesha and Simonich, 1978). In laser radar measurements at Garmisch-Partenkirchen (47.5°N) and Menlo Park (35°N), noticeable winter maxima of aerosol content were seen (Russell *et al.*, 1976; Reiter *et al.*, 1979). Minimum in autumn in the measurements by laser radar is noticeable at Nagoya (35°N), and found out also in the total stratospheric aerosol content measured by Hofmann and Rosen (1975). The summer maximum is very noticeable in the measurement at Nagoya, and not in other laser radar measurements. However, small summer maximum is recognized in measurements at Garmisch-Partenkirchen (laser radar measurement), and at Laramie, Wyoming (optical particle counter method). This summer maximum is rather unsteady, and decrease gradually year by year at Nagoya. As is discussed by Hofmann *et al.* (1975), and

Hofmann and Rosen (1977), the mechanism which produces above mentioned seasonal variation of aerosol content is possibly due to the variation of tropopause height, which provides an effective cleaning process of material in stratosphere. In addition to this, the air motions of global scale such as jet stream, various wave, and so on must affect on aerosol transportation, and causes difference among the measurements at various location.

The main processes which are controlling the behavior of stratospheric aerosol layer are eddy diffusion of aerosol, sedimentation, coagulation between particles, condensation of sulfuric acid vapor on to particle surface, and production of sulfuric acid vapor through photo-oxidation processes of COS, SO₂, H₂S and so on transported into from troposphere by eddy diffusion (*e.g.*, Junge *et al.*, 1961; Hamill *et al.*, 1978; Toon *et al.*, 1979).

The rate of stratospheric aerosol number density change is described as following form

$$\frac{\partial n}{\partial t} = \text{div}(F_1 + F_2) + P_1 + L_1 + P_2 + L_2 + P_3 \quad (3)$$

where n is aerosol differential number density in the radius range $R \sim R + dR$, F_1 eddy diffusion flux due to sedimentation, P_1 production rate due to condensation or evaporation of H₂SO₄ vapor, L_1 loss rate due to condensation or evaporation of H₂SO₄ vapor, P_2 production rate due to coagulation, L_2 loss due to coagulation, P_3 production rate due to homogeneous nucleation. The column density of stratospheric aerosol particles in unit of g/cm² is given by

$$N = \int_{Z_2}^{Z_1} \int_{R_2}^{R_1} \frac{4}{3} \pi R^3 \rho n(R) dR dZ \quad (4)$$

where ρ is density of particulate matter (g/cm³), Z_1 and Z_2 layer top and bottom respectively, R_1 and R_2 aerosol size limit, the number density S of aerosol is given by

$$S = \int n(R) dR \quad (5)$$

The terms controlled by coagulation are cancelled out when following integration is done

$$\int_{Z_2}^{Z_1} \int_{R_2}^{R_1} (P_2 + L_2) dR dZ = 0 \quad (6)$$

The contribution of homogeneous nucleation is possibly neglected (Toon *et al.*, 1979). The following integration (7) means the rate of particulate matter change (here 75% sulfuric

acid solution is assumed) through condensation of H_2SO_4 vapor onto aerosol surface, and is considered to be equal to rate of H_2SO_4 gas change in column,

$$P_{\text{H}_2\text{SO}_4} = \int_{Z_2}^{Z_1} \int_{R_2}^{R_1} (P_1 + L_1) \frac{4}{3} \pi R^3 \kappa dR dZ \quad (7)$$

where κ is constant determined by the chemical composition assumed here. The vertical integration of divergence term in equation (3) is rewritten as following form (only vertical fluxes are assumed)

$$\int_{Z_2}^{Z_1} \text{div}(F_1 + F_2) dZ \\ = [F_1 + F_2]_{\text{top}} - [F_1 + F_2]_{\text{bottom}} \quad (8)$$

where "top" and "bottom" mean the top of stratospheric aerosol layer and bottom of layer respectively. The flux of particulate matter is defined by

$$\phi_i = \int_{R_2}^{R_1} \frac{4}{3} \pi R^3 \rho F_i dR, \quad i=1 \text{ or } 2 \quad (9)$$

The rate of particulate matter change in column is given by

$$\frac{\partial N}{\partial t} = (\phi_1 + \phi_2)_{\text{top}} - (\phi_1 + \phi_2)_{\text{bottom}} + P_{\text{H}_2\text{SO}_4} \quad (10)$$

As is seen in equation (10), the budget of particulate matter in the stratosphere is controlled by the production due to condensation of H_2SO_4 vapor onto pre-existing particles and loss due to sedimentation or diffusion of particles. Considering that sulfuric acid contained in particles is produced through photo-oxidational reaction of sulfur compounds transported into stratosphere across the tropopause, and that most of H_2SO_4 is contained in aerosols as the form of sulfuric acid solution, the influx of sulfur compounds is necessary to compensate the production rate of H_2SO_4 75% solution. Schematically sulfur compounds budget in stratosphere is illustrated in Fig. 5.

Considering that the stratospheric aerosol is essentially large particle produced through condensation of H_2SO_4 vapor and coagulation of particles, as is pointed out by Hamill *et al.* (1978), and Toon *et al.*, (1979), it is assumed that the increase of vertically integrated backscattering coefficient in winter season is mainly due to gas-to-particle conversion process such as condensation of sulfuric acid vapor on aerosol surface. The ratio, G_p , of particle backscattering

Fig. 5. Schematic illustration on the cycle of sulfur compounds. Various gas phase sulfur compounds are converted to liquid phase matter through so-called "gas to particle conversion" on stratosphere. All aerosol particles are assumed to be composed with 75% sulfuric acid solution according to many investigations (*e.g.*, Toon *et al.* 1979).

coefficient to particle number density measured by balloon borne particle counter is already discussed (Russell *et al.*, 1975; Remsberg and Northam, 1975). If we use the value $G_p = 4.7 \times 10^{-11} \text{ cm}^2 \text{ str}^{-1}$ ($\lambda = 0.6943 \text{ \AA}$), as is presented by Russell *et al.* (1975) and Remsberg and Northam (1975), and assumption that all particles are composed with sulfuric acid solution of 75% (weight) and density of particle $\rho = 1.6 \text{ g/cm}^3$, then the increase of mass of particulate matter, ΔM is given by

Table 2

Flux of sulfur	Constituent	Comment
6×10^6	COS	Model calculation (Crutzen, 1976)
5×10^6	SO_2	Analysis of residence time (June, 1974)
2×10^7	SO_2	Seasonal variation of aerosol content (Hofmann, <i>et al.</i> , 1974)
9×10^6	SO_2	Model calculation (Lazarus and Gundred, 1974)
1.8×10^7	SO_2	Analysis of volcanic activity observation (Cadle, 1975)
3×10^8	SO_2	Model calculation (Harrison and Larson, 1974)
5.4×10^7	SO_2 , COS, H_2S (not identified)	Analysis of seasonal variation of aerosol content (present research)

* Unit of flux of sulfur: (atoms/cm²/sec)

$$\Delta M = \rho \iint (B_m(Z)/G_p)(4/3\pi \bar{R}^3) dD dt \quad (11)$$

where ΔZ means the vertical range from layer bottom to layer top, Δt means time span of winter season, and R is average radius of stratospheric aerosol ($0.25 \mu\text{m}$). On the basis of results in Fig. 3, ΔM in winter season is about $1.8 \times 10^{-7} \text{ g/cm}^2$ (column). This indicates that average production rate of H_2SO_4 solution in winter season is $8.5 \times 10^{15} \text{ g/cm}^2/\text{sec}$ for the time scale, $1.5 \times 10^7 \text{ sec}$ (about 6 months), and corresponding to flux of sulfur atoms into stratosphere $= 5.4 \times 10^7 \text{ atoms (S)/cm}^2/\text{sec}$. In table 2, influx of sulfur compounds estimated here is compared with other investigators' results. Present value has good agreement with other investigations, though the estimation was done very roughly.

Acknowledgements

The author would like to thank Prof. A. Ono, Nagoya University for useful discussion. He also thanks Mr. A. Mita, Mr. K. Nagaya, Nagoya University, and Mr. M. Wada, National Institute of Polar Research for helpful assistance of laser radar observation.

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レーザレーダによって観測された成層圏エアロゾルの濃度変化

第 I 部 季節変化と、対流圏から成層圏へのイオウ化合物の流束

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名古屋におけるレーザレーダ観測の結果をもとに、成層圏エアロゾル濃度の季節変化について議論した。結果は、顕著な冬季の極大がみられ、他の研究者の結果と同様なものである (Hofmann et al., 1975; Hofmann and Rosen, 1977; Bigg, 1975; Reiter et al., 1979)。名古屋での結果は、1977年さらには1976年の夏季にも第の極大が見られる。小さな夏季の極大は、他の研究者の結果にも見出されており、1975年の夏の極大は Gras and Laby (1979) の結果に、又 1978 年及び1977年の夏の極大は、Reiter 等の観測に見られる。種々の結果をまとめてみる期と、冬の極大は定常的に地球規模で観測され、小さい冬の極大が時々見られる。

もし成層圏エアロゾルが、75%の硫酸液滴からなっていると考えるなら、粒子状物質が沈降によって失われる量とつりあうためには、イオウ化合物の対流圏から成層圏への flux は、 5×10^7 (s) atoms/cm²/sec である。