COMPARISON OF THE IMPACT OF VOLCANIC ERUPTIONS AND AIRCRAFT EMISSIONS ON THE AEROSOL MASS LOADING AND SULFUR BUDGET IN THE STRATOSPHERE

G. K. Yue and L. R. Poole
Atmospheric Sciences Division
NASA Langley Research Center
Hampton, Virginia

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Atmospheric Sciences Division
NASA Langley Research Center
Hampton, Virginia 23665-5225

Abstract

Data obtained by the Stratospheric Aerosol and Gas Experiment (SAGE) I and II were used to study the temporal variation of aerosol optical properties and to assess the mass loading of stratospheric aerosols from the eruption of volcanos Ruiz and Kelut. It was found that the yearly global averae of optical depth at 1.0 μm for stratospheric background aerosols in 1979 was 1.16×10^{-3} and in 1989 was 1.66×10^{-3}. The eruptions of volcanos Ruiz and Kelut ejected at least 5.6×10^5 and 1.8×10^5 tons of material into the stratosphere, respectively. The amount of sulfur emitted per year from the projected subsonic and supersonic fleet is comparable to that contained in the background aerosol particles in mid-latitudes from 35°N to 55°N.

Introduction

It is recognized that the "background" aerosol particles in the lower stratosphere are supercooled sulfuric acid solution droplets composed of about 75% sulfuric acid by weight. Since these aerosols can directly and indirectly influence the radiation budget of the atmosphere and, thus, affect the climate, their properties have been of increasing concern. Based on measurements by optical particle counters over Laramie, Wyoming, during the period from 1971 to 1990, Hofmann (ref. 1) suggested that the background stratospheric sulfuric acid aerosol mass at northern mid-latitudes has increased by about 5% per year during the past 10 years. He later suggested that this increase of aerosol mass may be related to the increased emission of sulfur from aircraft (ref. 2). By considering the worldwide jet fuel consumption of 153×10^9 kg in 1987, and assuming the mass concentration of stratospheric aerosol is about 0.1 μg m^{-3} with a layer thickness of 5 km, he found that the total sulfur emission per year from airline traffic is about 65% of the required source strength for background aerosols. If this estimate is true, sulfur emissions from the continuously increasing airline traffic flight may greatly enhance the mass loading of aerosol particles in the stratosphere and perturb stratospheric temperature and ozone concentrations.

In this paper, we will address three questions: (1) Are background aerosol particles in the stratosphere increasing? (2) What is the effect of volcanic eruptions on the mass loading of aerosol particles in the stratosphere? (3) How does the amount of sulfur emitted from the proposed high-speed civil transport (HSCT) fleet compare with

other sources of sulfur in the stratosphere? Our results are based on the global measurements of aerosol particles measured by the Stratospheric Aerosol and Gas Experiment (SAGE) I and II (refs. 3 and 4). The calculation of the amount of sulfur from aircraft exhaust is based on the latitudinal distribution of fuel used by supersonic and subsonic aircraft (ref. 5).

Temporal Variation of Optical Properties of Stratospheric Aerosols Measured by SAGE I and II

SAGE I was launched on February 18, 1979, on the Application Explorer Mission-II (AEM-II) satellite. Its sensors measured the intensity of solar radiation traversing the Earth's limb during each sunrise and sunset event (approximately 15 each per day) encountered by the spacecraft in its orbit. The measured data were inverted to obtain profiles of aerosol extinction at 1.0 and 0.45 μm. Due to a power failure in the spacecraft, SAGE I measured only sunset events after May 1979 and stopped collecting data entirely in December 1981. SAGE II was launched on October 5, 1984, on the Earth Radiation Budget Satellite (ERBS) and is still operational. The SAGE II instrument is in a similar orbit to SAGE I and provides aerosol extinction profiles at 1.02 μm, 0.453 μm, and two other wavelengths.

In this study, the optical depth of stratospheric aerosol is defined as the integral of the aerosol extinction coefficient at 1.0 μm or 1.02 μm from an altitude of 2 km above the tropopause to an altitude of 15 km above the tropopause. The 2 km is used because the available tropopause heights are not necessarily derived from measurements made close to the location of our profiles, and high altitude clouds will occasionally occur at heights above the assumed tropopause height (ref. 6). The SAGE I and SAGE II measurements of mean hemispheric optical depth at 1.0 μm are shown in Figures 1(a) and 1(b), respectively.

As can be seen from Figure 1(a), the optical depths in the year 1979 for both hemispheres were quite constant at about 1.16×10^{-3}. After December 1979, the hemispheric optical depths began to increase gradually due to the eruption of volcanos Sierra Negra in November 1979, Mount St. Helens in May 1980, and Ulawun in October 1980. By the end of 1980, the level of volcanic debris in the Northern Hemisphere began decaying. However the eruptions of Alaid in April 1981 and Pagan in May 1981 ejected appreciable amounts of particles and gases into the stratosphere, resulting in the increase of optical depths after May 1981. The optical depths in November 1981 for the Southern and Northern Hemispheres were 2.3×10^{-3} and 2.4×10^{-3}, respectively.

At the beginning of 1985, the optical depths of the Southern and Northern Hemispheres were 6.9×10^{-3} and 8.6×10^{-3}, respectively. The several-fold increase of optical depth from the background value measured in 1979 is due to the presence of debris from El Chichon which erupted in April 1982. The optical depths vary

seasonally with a maximum in local winter and a minimum in local summer. The temporal decay of the level of debris from El Chichon eruption was interrupted by the eruption of Ruiz in November 1985 and the eruption of Kelut in February 1990. By the end of 1989, the optical depths of the Southern and Northern Hemispheres were 1.47 x 10^{-3} and 1.51 x 10^{-3}, respectively. The yearly averaged optical depths of the Southern and Northern Hemispheres for 1989 were 1.83 x 10^{-3} and 1.50 x 10^{-3}, respectively. If we regard the aerosol properties observed in 1989 as the new "background" aerosol properties, there is a 43% increase of aerosol optical depths from 1979 to 1989.

In order to study the change of aerosol size, we have also calculated the ratio of optical depth at 0.45 (or 0.453) μm to that at 1.0 (or 1.02) μm. This ratio is a measure of the column aerosol size; the smaller the ratio, the larger the size. The monthly optical depth ratios are averaged over each hemisphere and the results are shown in Figures 2(a) and 2(b). The SAGE I data shows a much larger month-to-month variation, possibly due to the fact that larger uncertainties are associated with the aerosol extinction at 0.45 μm. Further observations are needed before any trend analysis of the aerosol size can be conclusive. However, the gradual increase in optical depth ratio for both hemispheres shown in Figure 2(b) does indicate that the effects of El Chichon are diminishing.

Mass Loading of Volcanic Aerosols

As mentioned in the previous section, there have been two major volcanic eruptions observed by SAGE II. The first increase in optical depth is identified to be due to the eruption of Ruiz (4.89°N, 75.37°W) on November 13, 1985, and the second increase in optical depth is identified to be due to the eruption of Kelut (7.93°S, 112.31°E) on February 10, 1990. The temporal variation of optical depth for latitude bands in the tropical region from 20°S to 20°N, where these two volcanos are located, is shown in Figure 3. The dramatic increase of optical depth after November 1985 and February 1990 is obvious in this figure. In order to estimate the mass loading of volcanic aerosols we have used the corresponding SAGE II water vapor data to deduce aerosol composition and the multi-wavelength aerosol data profile to deduce aerosol size (ref. 7). From aerosol composition and size distribution, the mass loading for each latitude band and month is calculated. The mass column density is obtained by integrating the mass density from a height of tropopause +2 km to a height of tropopause +15 km. The mass column density for latitude bands from 20°S to 20°N is shown in Figure 4. The global mass loading of stratospheric aerosols can easily be obtained by integrating the aerosol mass in each latitude band, and the results are shown in Figure 5(a).

It can be seen that after November 1985, the aerosol mass loading began to increase and reached a peak value in February 1986. The difference of aerosol mass loading in February 1986 and that in November 1985 was 5.6 x 1.0^{5} metric tons. We

assume this is the amount of material ejected into the stratosphere by volcano Ruiz even though the actual amount should be higher since we are calculating only from a height of tropopause +2 km upward. The amount of material ejected by volcano Kelut was much less and was concentrated in tropical regions. We found it hard to estimate the amount of material ejected by Kelut in Figure 5(a) because the fluctuation of mass loading in high latitudes at the beginning of 1990 was comparable to that in low latitudes due to this volcanic eruption. The mass loading of aerosol particles in the stratosphere from 40°S to 40°N is shown in Figure 5(b). The difference in mass loading between January 1990 and March 1990 is 1.8 x 10^5 tons. In order to compare the strength of different volcanic eruptions and to assess the impact of volcanic eruption on the sulfur budget in the stratosphere, we have listed the estimates of mass loading from different volcanos in Table 1. In addition, the mass loading of 1979 background aerosols (ref. 6) and the mass loading of 1989 background aerosols obtained from Figure 5(a) are also listed. Both the Mount St. Helens and Ruiz eruptions ejected amounts of aerosol material close to the amount of background aerosols.

Table 1
Estimate of Aerosol Mass Loading in the Stratosphere

Date	Volcano or Background	Location	Mass Loading (10^5 tons)	Reference
1979	Background		5.7	6
1989	Background		7.5	This study
3/17/63	Agung	8.4°S, 115.5°E	160	8
			300	9, 10
10/10/74	Fuego	14.5°N, 90.9°E	60	9, 10
			30	11
1/22/76	St. Augustine	59.4°N, 153.4°W	6.0	10
4/17/79	La Soufriere	13.3°N, 61.2°W	0.023	12
11/13/79	Sierra Negra	0.8°S, 91.2°W	1.6	6
5/18/80	St. Helens	46.2°N, 122.2°W	5.5	6
10/7/80	Ulawun	5.0°S, 151.3°E	1.8	6
4/27/81	Alaid	50.8°N, 155.5°E	3.0	6
5/15/81	Pagan	18.1°N, 145.8°E	2.0	6
4/4/82	El Chichon	17.3°N, 93.2°W	200	13
			120	14
			101	15
11/13/85	Ruiz	4.9°N 75.4°W	5.6	This study
2/10/90	Kelut	7.9°S, 112.3°E	1.8	This study

Estimate of the Impact of Subsonic and Supersonic Aircraft Exhaust on Sulfur Budget

Aerosol particles in the stratosphere are formed, in large part, from precursor sulfur-bearing gases. Carbonyl sulfide (OCS) and sulfur dioxide (SO_2) are the most prominent precursors. Concentration of other sulfur-bearing gases, including carbon disulfide (CS_2) and hydrogen sulfide (H_2S), are too small to be of any direct influence on the formation and properties of sulfate particles in the stratosphere. Both carbonyl sulfide and sulfur dioxide will be oxidized through complex processes to form sulfuric acid (H_2SO_4) which directly affects the properties of sulfate particles (ref. 16). The concentration of OCS is about 0.3 to 0.5 ppbv at altitudes of about 15 km and rapidly decreases at higher altitudes (refs. 17 and 18). The concentration of SO_2 is about 0.05 ppbv at 15 km and decreases to 0.01 ppbv at 30 km (ref. 16). The concentration of H_2SO_4 has been deduced using mass spectrometer measurements of stratospheric ions. The results indicated that H_2SO_4 concentration is about 1.7×10^5 cm^{-3} in most parts of the lower stratosphere and it increases rapidly to 10^7 cm^{-3} near 34 km (refs. 19 and 20). The calculated global amounts of sulfur contained in these species in the lower stratosphere from 15 km to 30 km are listed in Table 2.

Also listed in Table 2 are the amount of sulfur in the 1979 and 1989 background aerosols and aerosols from the eruption of St. Helens and Ruiz. We assume that stratospheric aerosols are composed of approximately 75% H_2SO_4 and 25% H_2O. Our estimate of sulfur for 1979 background aerosol is more than twice the value (6.25×10^7 kg) estimated by Hofmann. The reason is that he assumed an aerosol layer of 5 km thickness and we integrated aerosol data from a height of about 15 km to 30 km.

In the late 1980s, there has been renewed commercial interest in developing supersonic aircraft, now denoted as high-speed civil transports, or HSCTs. It is suggested that an economically feasible fleet size is about 500 aircraft, and it is estimated that the supersonic fleet will consume fuel during cruise at a rate of 70×10^9 kg/year. The ideal cruise altitude depends on the Mach number which varies from about 15.8 km at Mach 1.6 to about 22.8 km at Mach 3.2. Since most of the flights will be in mid-latitudes, we assume all the supersonic aircraft exhaust reaches the stratosphere. For the subsonic fleet, the impact is represented by emissions at two cruise altitudes: 22,000-32,000 ft and 32,000-42,000 ft. The assumed fuel consumptions are 17×10^9 kg/year and 152×10^9 kg/year, respectively. Depending on the flight altitude and latitude, some of the subsonic flights will be in the stratosphere. If we follow Hofmann's argument that 1/6 of the flights at 32,000-42,000 ft are in the stratosphere and 1/4 of the exhaust from tropospheric flights will be transported into the stratosphere through dynamic processes, then 9/24 of the exhaust from subsonic jets will reach the stratosphere. The emission index for SO_2 is 1.0 gm per 1.0 kg of fuel. The amounts of sulfur in the SO_2 from the exhaust of subsonic and

supersonic fleets per year are also listed in Table 2. Since a lifetime of 1 year is typical of volcanic stratospheric aerosols, we assume that the sulfur from aircraft engines will stay in stratosphere for 1 year.

Table 2
Amount of Sulfur in the Stratosphere

Source	Amount (10^7 kg)
Background OCS	17.4
Background SO_2	2.6
Background H_2SO_4	0.03
Background Aerosol (1979)	14.0
Background Aerosol (1989)	18.3
St. Helens Aerosol	13.5
Ruiz Aerosol	13.7
SO_2 in Exhaust from Subsonic Fleet	2.9
SO_2 in Exhuast from Supersonic Fleet	3.5

It can be seen that the SO_2 from the aircraft exhaust is comparable to the background SO_2 concentration. It should be noted that most commercial flights are in the 30°N - 50°N corridor. In these regions the yearly exhaust of SO_2 from aircraft is higher than the background value. We used the SAGE II data sets to calculate the average mass loading of aerosol particles in each latitude band for 1989. The latitudinal distributions of sulfur from the exhaust of the subsonic fleet at 32,000-42,000 ft and from the exhaust of the supersonic fleet are also calculated. The ratios of these values to the sulfur in the 1989 aerosol particles are plotted in Figures 6(a) and 6(b).

Our results show that sulfur from the exhaust of the subsonic fleet at 35° N is about 92% of that from sulfate particles. At 45°N, the exhaust of the supersonic fleet is about 82% of that from sulfate particles. Even at other latitudes between 25°N and 55°N, the amount of sulfur exhausted from aircrafts is comparable to that in the background aerosols. Since most of the flights are at altitudes below 22 km and our estimation of the background sulfur is integrated from 15 km to 30 km, the amount of sulfur from engine exhaust is more than that from background aerosols in lower altitudes. Also, the appreciable increase in the amount of sulfur in the lower stratosphere from aircraft engine exhaust may increase both the number and sizes of aerosol particles in the stratosphere. The effect on northern mid-latitudes may be similar to that after a moderate volcanic eruption such as Mount St. Helens and Ruiz. If airline traffic is doubled for a period of years and the engine exhaust remains in the stratosphere for more than a year, stratospheric temperature and ozone concentrations may be perturbed appreciably. A more accurate estimate requires detailed study of the dynamical and chemical processes involving sulfur compounds in the stratosphere.

Conclusions

Based on stratospheric aerosol properties observed by the SAGE I and SAGE II experiments and a realistic estimate of exhaust from future subsonic and supersonic aircraft fleets, we conclude:

(1) There is a 43% increase in optical depth observed by SAGE II in 1989 relative to SAGE I in 1979. Debris from volcanic eruptions and aircraft exhausts may contribute to this increase in optical depth. Unfortunately, we have not had a chance to observe the optical properties of aerosol particles for one or two years without an eruption due to the Kelut eruption in early 1990. Further observations are needed before any trend analysis can be conclusive or before the relative importance of various sources can be assessed.

(2) In general, the mass loading in the stratosphere decreased from 1985 to 1990, which indicates the diminishing influence of El Chichon material. However, the decay was interrupted by the eruption of Ruiz in November 1985, and the eruption of Kelut in February 1990. The amount of material ejected into the stratosphere by Ruiz was about 5.6×10^5 tons, which is equal to the 1979 global mass loading of background aerosols. The amount of material ejected into the stratosphere by kelut was only about 1.8×10^5 tons.

(3) If the lifetime of sulfur from aircraft exhausts is 1 year, typical of volcanic stratospheric aerosol, the amount of sulfur from aircraft exhaust is comparable to that in the ambient aerosols at latitudes from 30°N to 50°N where most commercial airline flights take place. The increase in the amount of sulfur in the stratosphere due to aircraft exhaust may equal that of a moderate volcanic eruption. If airline traffic continues to increase, aerosol mass loading may reach a level that will perturb stratospheric temperature and ozone.

References

1. Hofmann, D. F., Increase in the stratospheric background sulfuric acid aerosol mass in the past 10 years, Science, 248, 996-1000, 1990.
2. Hofmann, D. F., Aircraft sulfur emissions, Nature, 349, 659, 1991.
3. McCormick, M. P., Aerosol measurements from Earth orbiting spacecraft, Adv. Space Rev., 2, 73-86, 1982.
4. McCormick, M. P., SAGE II: An Overview, Adv. Space Res., 7, 319-326, 1987.
5. Ko, M. K., D. K. Weisenstein, N.-D. Sze, R.-L. Shia, J. M. Rodriguez, and C. Heisey, Effects of engine emission from high speed civil transport aircraft: A two-dimensional modeling study. Final report for ST Systems Corporation for period July 1, 1988 through June 20, 1989, 1990.
6. Kent, G. S., and M. P. McCormick, SAGE and SAM II measurements of global stratospheric aerosol optical depth and mass loading, J. Geophys. Res., 89, 5303-5314, 1984.

7. Yue, G. K., M. P. McCormick, and W. P. Chu, Retrieval of composition and size distribution of stratospheric aerosols with the SAGE II satellite experiment, J. of Atmos. and Oceanic Technology, 3, 371-380, 1986.
8. Deirmendjian, D., On volcanic and other particulate turbidity anomalies, Advan. Geophys., 16, 267-296, 1973.
9. Cadle, R. D., C. S. Kiang, and J. F. Louis, The global scale dispersion of the eruption clouds from major volcanic eruptions, J. Geophys. Res., 81, 3125-3132, 1976.
10. Cadle, R. D., F. G. Fernald, and C. L. Frush, Combined use of lidar and numerical diffusion models to estimate the quantity and dispersion of volcanic eruption clouds in the stratosphere: Vulcan Fuego, 1974, and Augustine, 1976, J. Geophys. Res., 82, 1783-1786, 1977.
11. Lazrus, A. L., R. D. Cadle, B. W. Gandrud, J. P. Greenberg, B. J. Huebert, and W. I. Rose, Jr., Sulfur and halogen chemistry of the stratosphere and of volcanic eruption plumes, J. Geophys. Res., 84, 7869-7875, 1979.
12. McCormick, M. P., G. S. Kent, G. K. Yue, and D. M. Cunnold, SAGE measurements of the stratospheric aerosol dispersion and loading from the Soufriere volcano, NASA Tech Paper 1922, November 1981.
13. Hofmann, D. F., and J. M. Rosen, Stratospheric sulfuric acid fraction and mass estimate for the 1982 volcanic eruption of El Chichon, Geophys. Res. Lett., 10, 313-316, 1983.
14. McCormick, M. P., and T. J. Swissler, Stratospheric aerosol mass and latitudinal distribution of the El Chichon eruption cloud for October 1982, Geophys. Res. Lett., 10, 877-880, 1983.
15. Mroz, E. J., A. S. Mason, and W. A. Sedlacek, Stratospheric sulfate from El Chichon and the mystery volcano, Geophys. Res. Lett., 10, 873-876, 1983.
16. Turco, R. P., R. C. Whitten, and O. B. Toon, Stratospheric aerosols: Observation and theory, Review of Geophys. and Space Phy., 20, 233-279, 1982.
17. Inn, E. C. Y., J. F. Vedder, B. J. Tyson, and D. O'Hara, COS in the stratosphere, Geophys. Res. Lett., 6, 191-193, 1979.
18. Inn, E. C. Y., J. F. Vedder, and D. O'Hara, Measurement of stratospheric sulfur constituents, Geophys. Res. Lett., 8, 5-8, 1981.
19. Arnold, F., and R. Fabian, First measurements of gas phase sulfuric acid in the stratosphere, Nature, 284, 610-611, 1980.
20. Arnold, F., R. Fabian, and W. Joos, Measurements of the height variation of sulfuric acid vapor concentrations in the stratosphere, Geophys. Res. Lett., 8, 83-86, 1981.

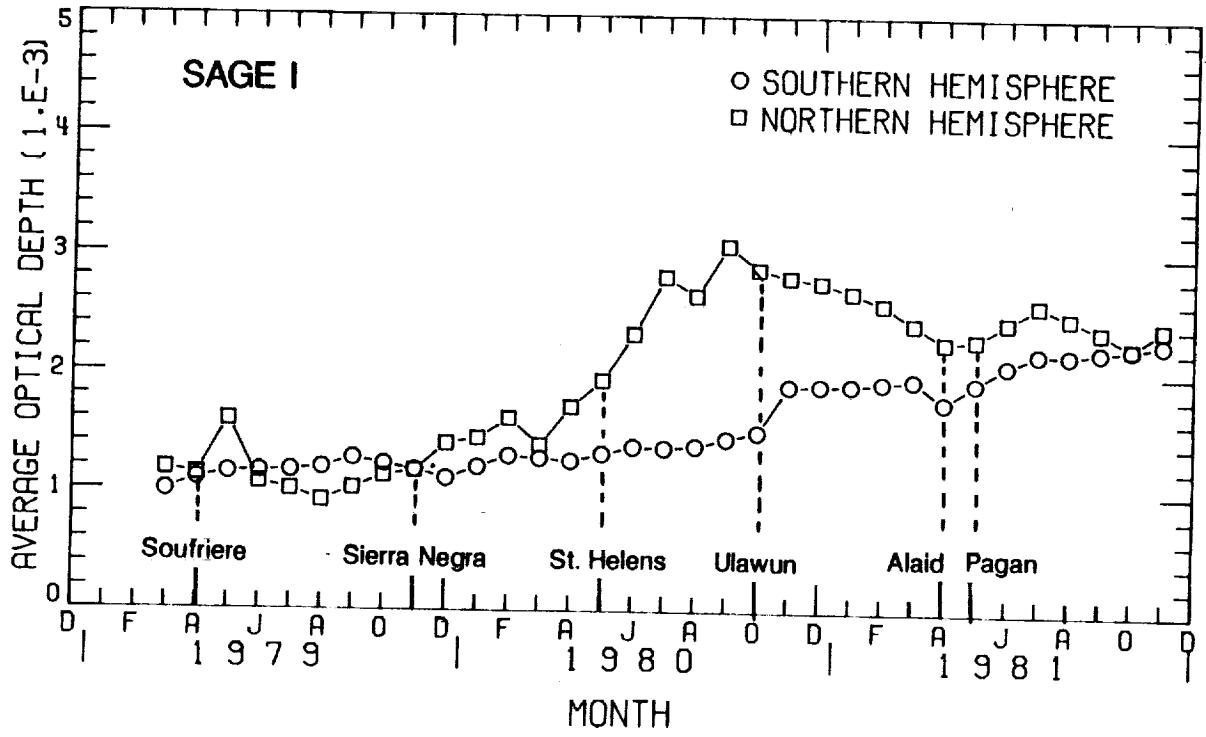

Figure 1(a)

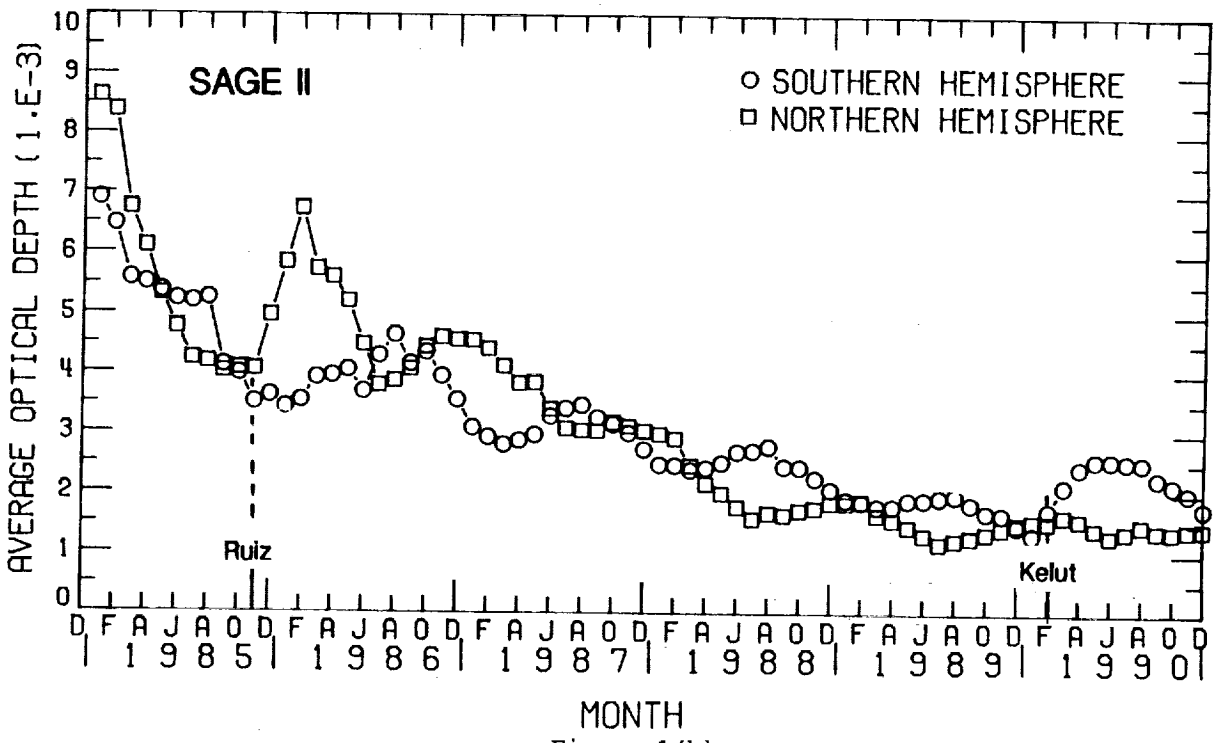

Figure 1(b)

Figure 1. Temporal variation of optical depth at 1.0 micron averaged over latitude in the Southern Hemisphere and the Northern Hemisphere.
(a) from 1979 to 1981, measured by SAGE I;
(b) from 1985 to 1990, measured by SAGE II.

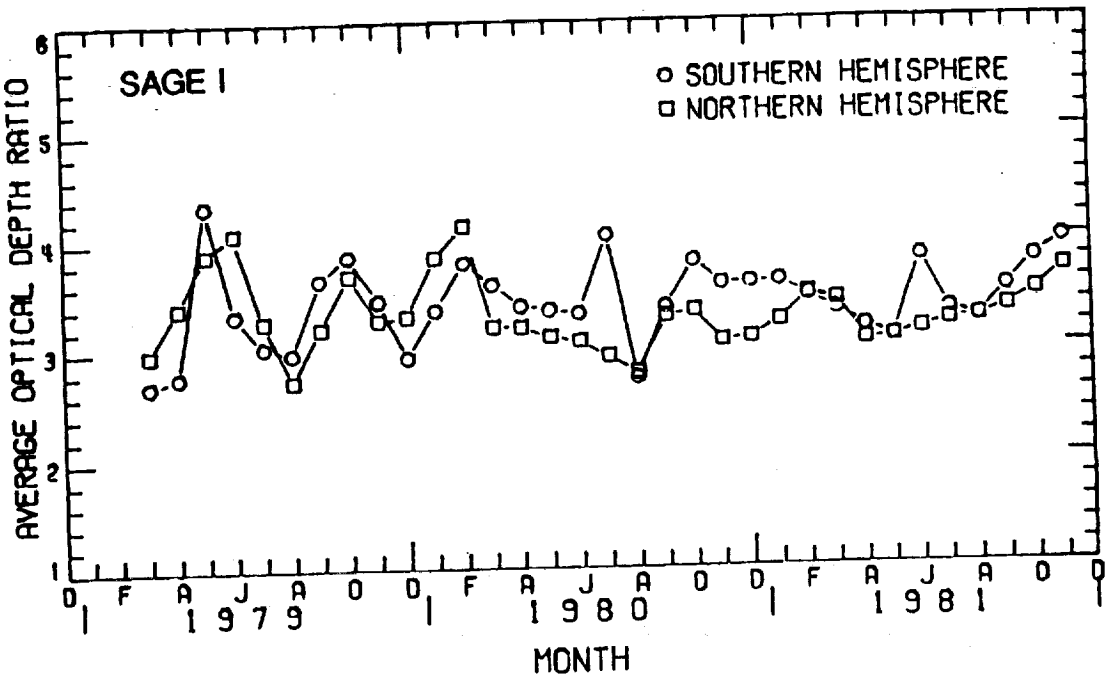

Figure 2(a)

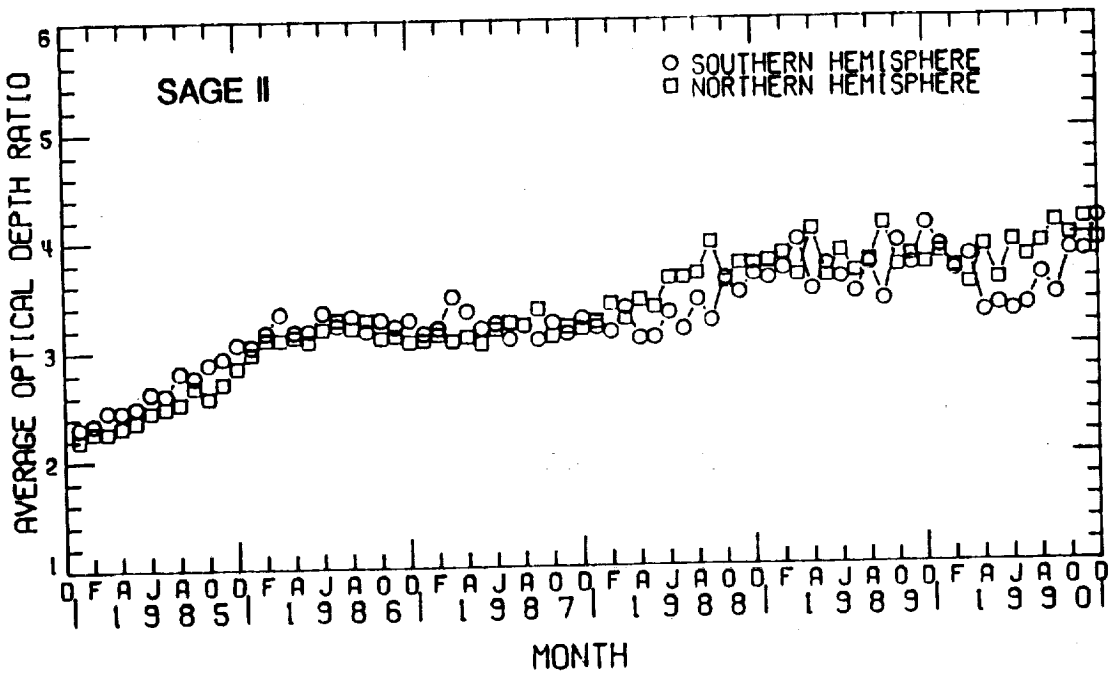

Figure 2(b)

Figure 2. Temporal variation of the ratio of optical depth at 0.45 μm to optical depth at 1.0 μm averaged over latitude in the Southern Hemisphere and Northern Hemisphere.
(a) from 1979 to 1981, measured by SAGE I;
(b) from 1985 to 1990, measured by SAGE II.

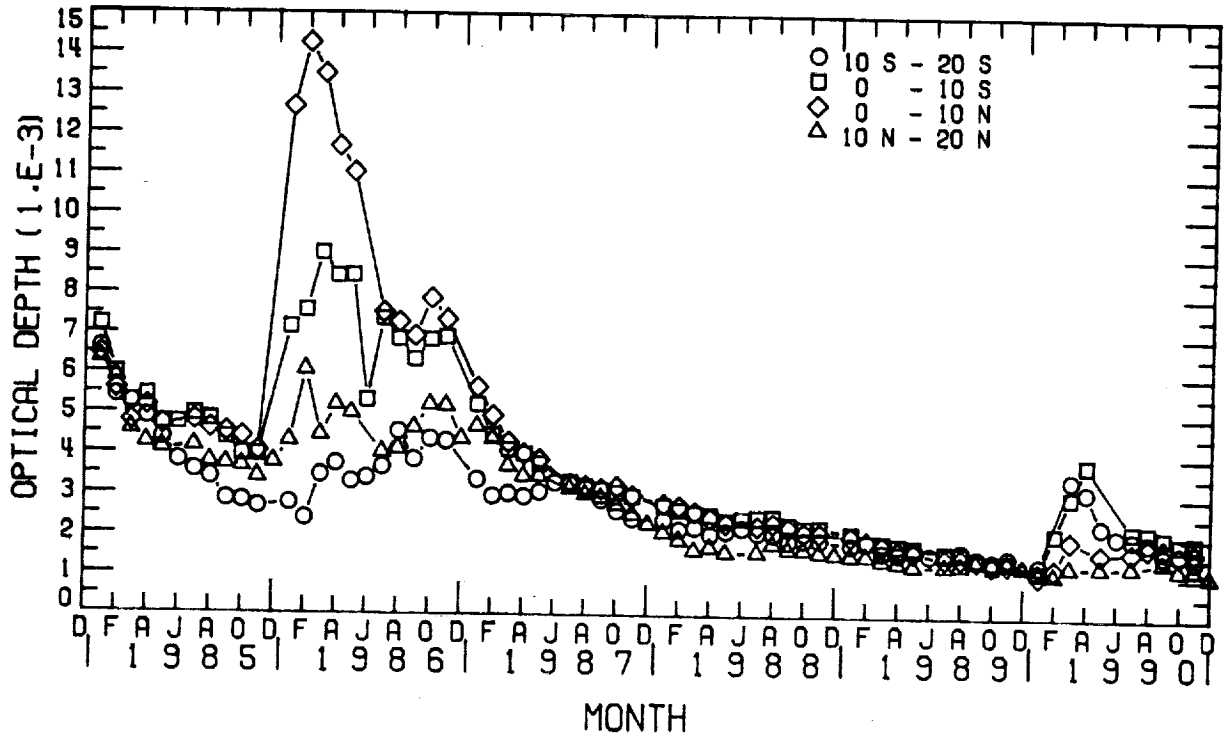

Figure 3. Temporal variation of optical depth at 1.02 μm measured by SAGE II from 1985 to 1990 for latitude bands from 20°S to 20°N.

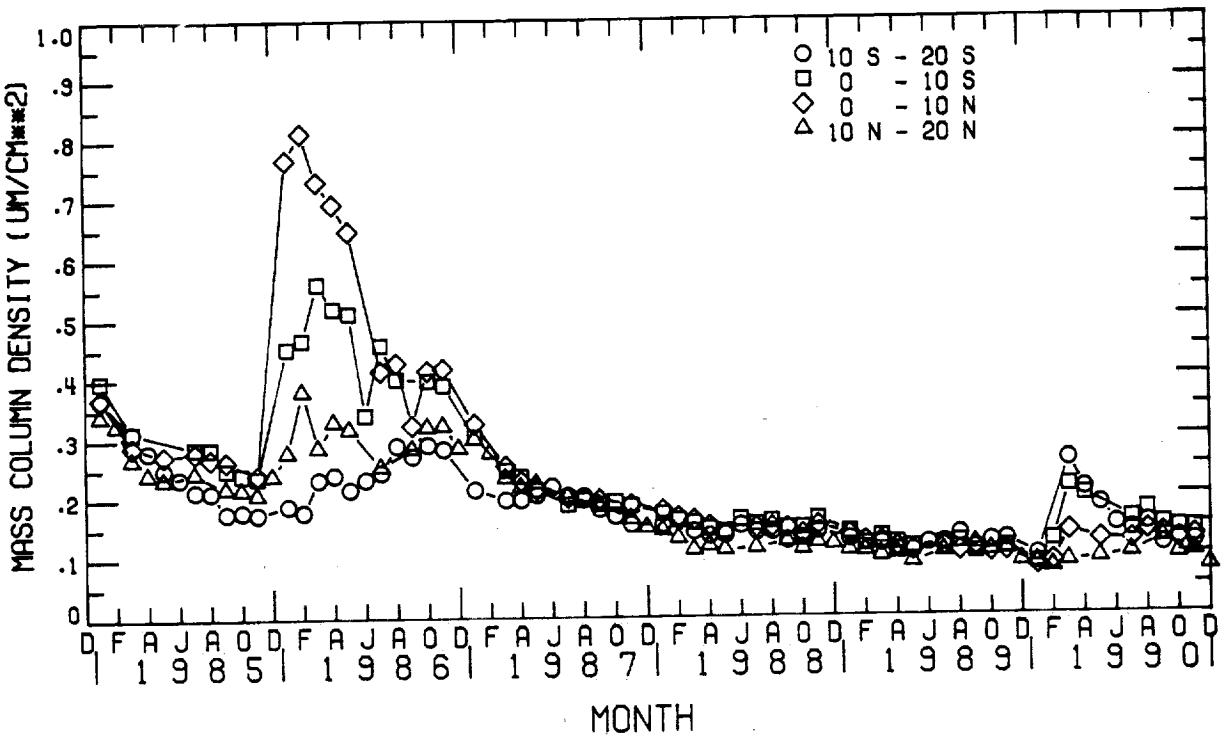

Figure 4. Temporal variation of mass column density deduced from SAGE II multiwavelength data from 1985 to 1990 for latitude bands from 20°S to 20°N.

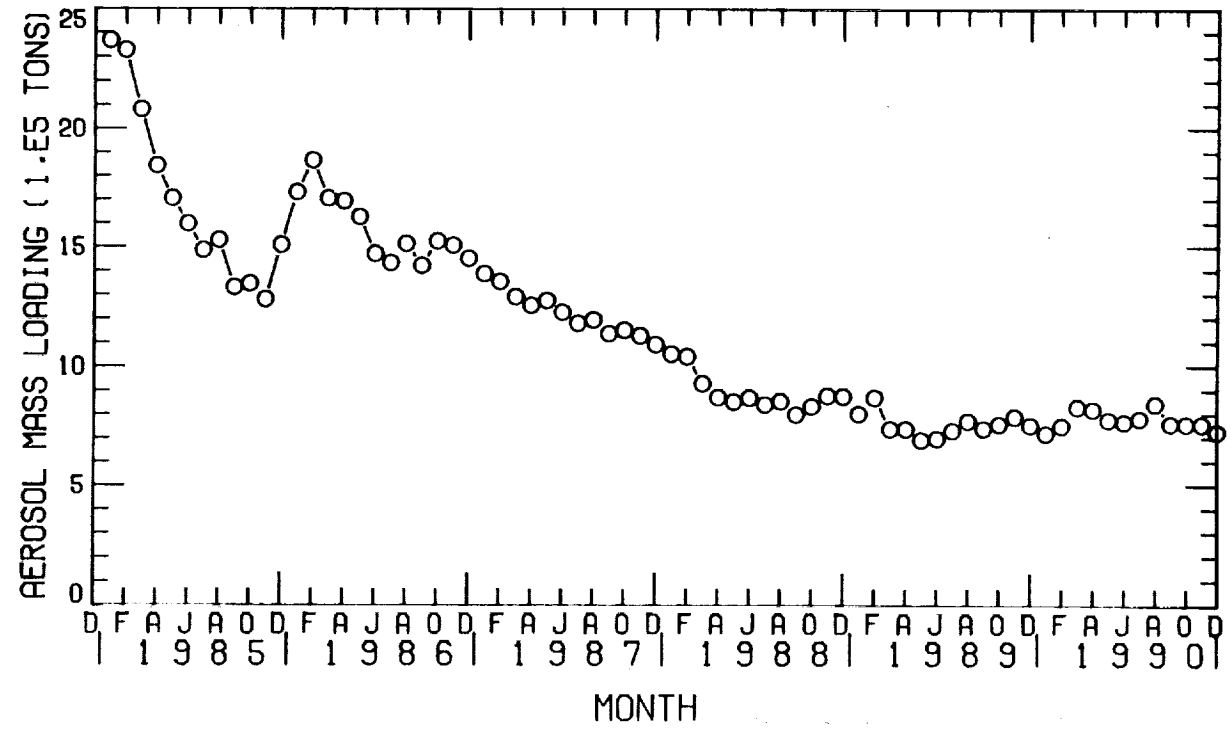

Figure 5(a)

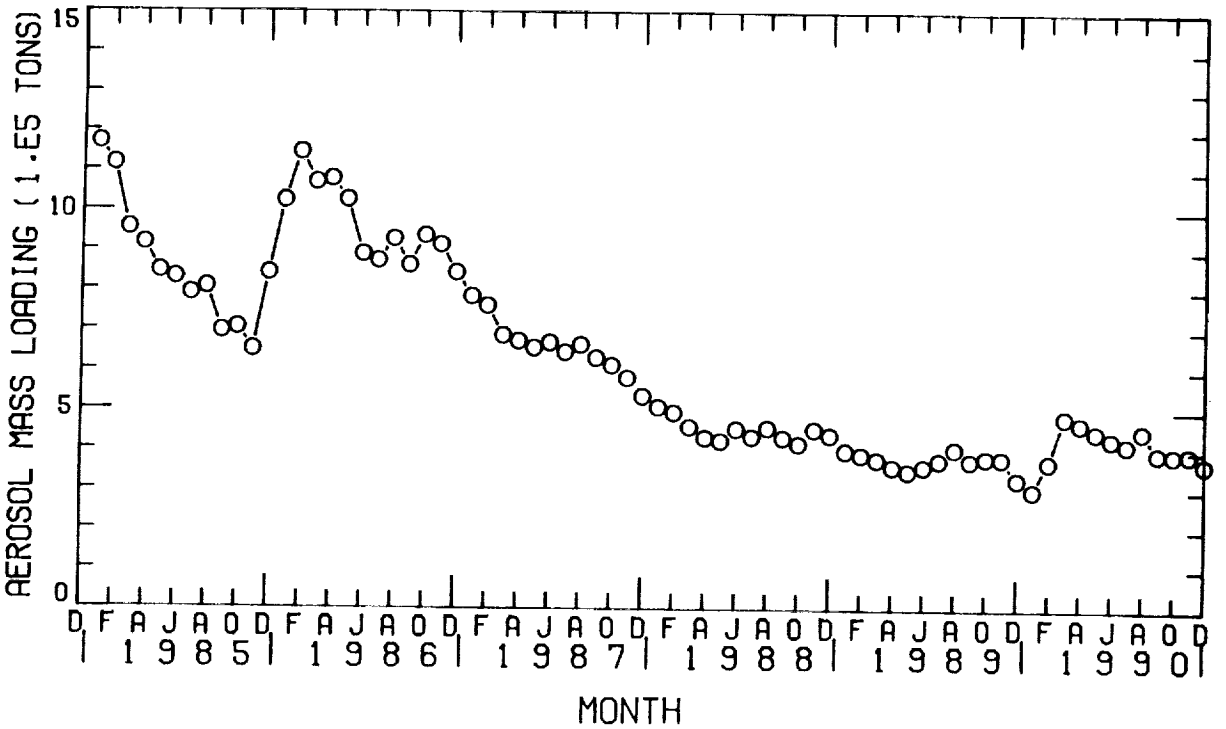

Figure 5(b)

Figure 5. Temporal variation of total mass loading for aerosols in the stratosphere.
(a) Integrated over the whole globe;
(b) Integrated from 40°S to 40°N.

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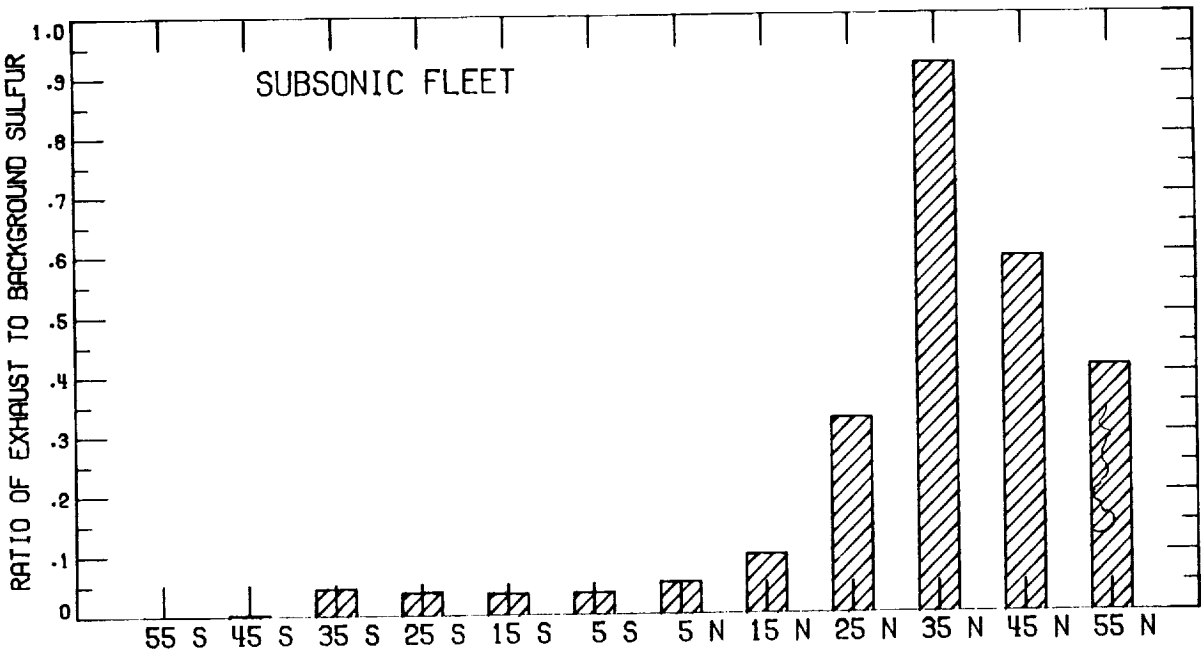

Figure 6(a)

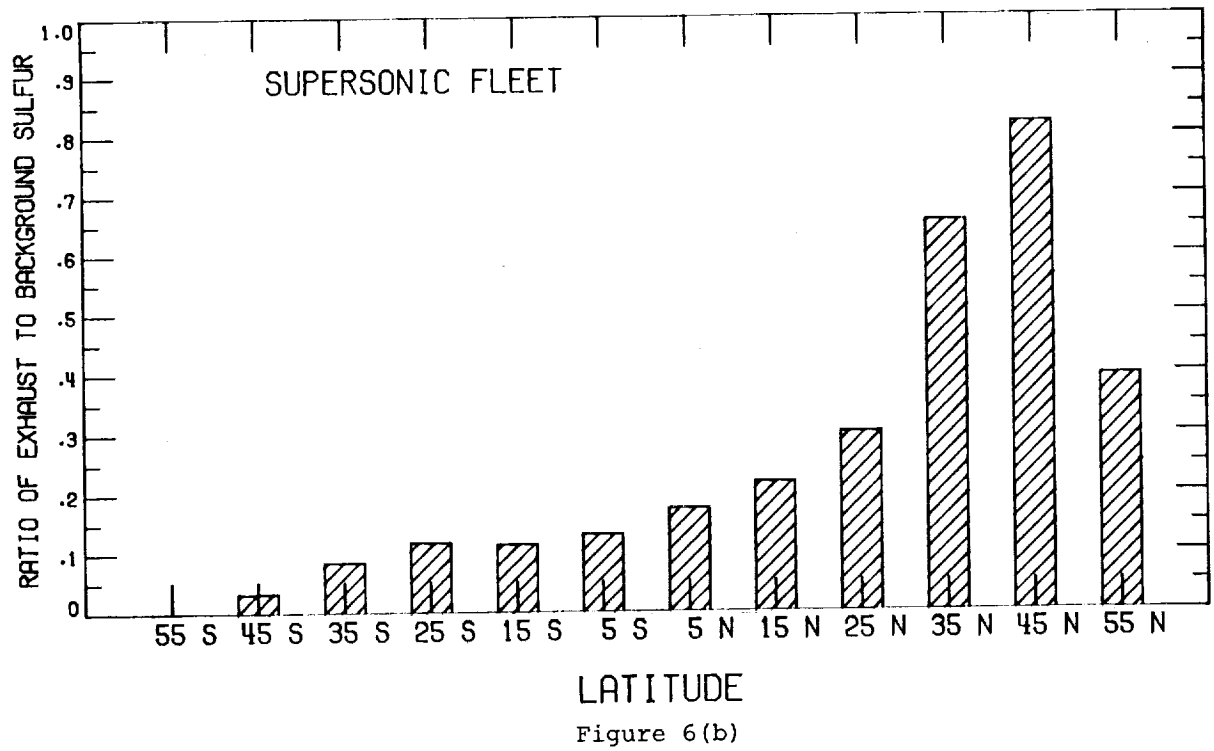

Figure 6(b)

Figure 6. Latitudinal distribution of the ratio of sulfur in aircraft exhaust to that in the 1989 background aerosol particles.
(a) Subsonic fleet;
(b) Supersonic fleet.

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