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NASA Technical Memorandum 83217

NASA-TM-83217 19820006818

STRATOSPHERIC AEROSOLS

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INTRODUCTION

Background

The stratospheric aerosol layer, or Junge Layer, is a region of the lower stratosphere where the large particle ($r>0.15$ m) mixing ratio reaches a maximum. This mixing ratio peak, of about 10 particles per mg of air in nonvolcanic periods, occurs at about 10 km above the local tropopause. Likewise, peak concentrations in this layer of about 0.5 particles/cm^3 ($r>0.15$ μm) occur at about 6 km above the local tropopause. These values drop off quickly at altitudes between 25 to 30 km.

The exact composition of the aerosol particles is uncertain but most evidence now supports an aerosol composed of a 75% (by weight) solution of sulfuric acid. The primary source of the stratospheric sulfur that establishes this background layer is thought to be tropospheric OCS and possibly CS_2. OCS, for example, is thought to diffuse upwards from the troposphere, photodissociate to produce free sulfur, and then SO_2 by oxidation. Similarly, SO_2 and OCS are products of the photo-oxidation of CS_2. Photochemical processes subsequently generate the gas phase H_2SO_4 which then participates in the microphysical processes of nucleation and condensation, leading to particle formation and growth. The aerosol particles formed also coagulate with each other, undergo gravitational sedimentation, and are vertically mixed by eddy diffusion. The exact nucleation mechanism is uncertain. Many have been proposed: homogeneous, ion, and heterogeneous heteromolecular nucleation of H_2SO_4-H_2O systems; the clustering of sulfate radicals; and heterogeneous nucleation onto stable neutral ion-ion recombination complexes. They all appear, however, to lead to the same general properties of the aerosol layer (Hamill et al. 1982). Although the exact nucleation scheme is necessary to determine the small ($r<0.1$ μm) particle contribution to the size distribution, it is not important for predicting aerosol spatial distributions or mass loadings.

Volcanic Disturbances

Violent volcanic eruptions can enhance the stratospheric aerosol layer greatly by orders of magnitude. The eruptions that penetrate the tropopause are thought to contain high amounts of sulfur gases, and in particular SO_2. Some sulfuric acid may even be formed in the volcanic magma and/or eruption cloud. Whereas the tropospheric SO_2 is too reactive to be a significant source for the stratospheric sulfuric acid background layer, this direct injection of SO_2 by volcanoes is probably the necessary source for these enhanced periods. Figure 1 from Swissler et al. (1982) shows the peak aerosol mixing ratio and peak lidar backscatter mixing ratio for the last 7 years, indicating background levels and levels of enhancement due to

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volcanic eruptions. The eruption of Fuego in Guatemala caused the order of magnitude enhancement in late 1974; the eruption of Mount St. Helens is responsible for the enhancement in May 1980. This figure shows that the measured decay after the Fuego enhancement depended somewhat on the particle size measured, or on the particle size that lidar at a wavelength of 0.6943 μm is most sensitive to. The 1/e decay rates are 10.4 months for particles greater than 0.15 μm radius, 7.9 months for particles greater than 0.25 μm radius, and 7.4 months for lidar, which was influenced by particles larger than 0.3 μm.

Table I presents a summary, taken from various sources, of the various properties of stratospheric aerosols thought to be representative of background and enhanced (volcanic) conditions.

Observational Techniques

Experimental observations of the stratospheric aerosol layer have been made for nearly 20 years with a variety of different sensors. Junge et al. (1961) employed particle impactors flown on balloons to measure and describe the stratospheric sulfate aerosol layer. Balloonborne light scattering particle counters called dustsondes have been used extensively by researchers at the University of Wyoming for a number of years (Rosen, 1964; Rosen, 1971; Hofmann et al. 1975; Pinnick et al. 1976; Hofmann and Rosen, 1981). Wire impactors (Farlow et al., 1981) and filters (Gandrud and Lazrus, 1981) have been flown for a number of years on high-altitude aircraft. Ground-based lidar systems to measure the stratospheric aerosol have been developed by a number of different groups (Fiocco and Grams, 1964; Collis and Ligda, 1966; Russell et al., 1976; Russell and Hake, 1977; McCormick et al., 1978). Airborne lidar systems have also been developed to study the stratospheric aerosol (Fox et al., 1973; Fernald and Schuster, 1977; Russell et al., 1981b). More recently, satellite-borne radiometers (SAM II-Stratospheric Aerosol Measurement, and SAGE-Stratospheric Aerosol and Gas Experiment) have been developed (McCormick et al., 1979) to monitor and map out the global distribution of stratospheric aerosols.

Global Observations

Figure 1 gave the long-term characteristics of aerosol size from the University of Wyoming's dustsonde instrument and aerosol backscatter from NASA Langley's lidar system. Both data sets are for fixed ground-based sites. The SAM II and SAGE data are just now being published and represent a large increase in global aerosol information.

Figure 2(a) gives an example of a typical SAGE background profile, and 2(b) shows a SAGE profile with a volcanic enhancement. Figure 3 shows a plot of average zonal aerosol extinction versus altitude and latitude. The tropopause is marked with a dashed line. Below this, tropospheric clouds are evident. Typical background stratospheric aerosol extinction is shown to be about 10^{-4} km^{-1} at 1.0 μm wavelength.

TABLE I.- STRATOSPHERIC AEROSOLS

AEROSOL PROPERTY	BACKGROUND VALUE (1978-1979)	VOLCANIC VALUE (1980)	REFERENCE
Composition	H_2SO_4 (60-85%)/ H_2O (40-15%) $NH_4(SO_4)_2$?	Same as background after approx. 1-4 mo	Russell et al. 1981a (see Appendix)
Refractive Index	1.40-0i 1.52-0i 1.42-0i 1.52-0.005i 1.43-0i 1.33-0i	"	"
Size Distribution	Combination of 9 model aerosol size distributions	"	"
Optical Depth,* δ (1.0 μm)	0.001	0.01	McCormick et al. 1981a,b
Extinction,* β	1.2×10^{-4} km^{-1}	1.0×10^{-3} km^{-1}	"
Lidar Backscatter Ratio (0.6943 μm)	1.07	≤100	Swissler et al. 1982
Mass Loading†	0.5×10^6 tonnes	$+0.3 \times 10^6$ tonnes	McCormick et al. 1981b
Number Density, N (r>0.15 μm)	0.5 particles/cm^3	≤100 part/cm^3	Hofmann & Rosen 1981
$N_{r>0.15}/N_{r>0.25}$	5.5	2.0	"
Ext. Peak Height Minus Trop. Height (mean, mid-latitude)	~6 km	Depends on time after injection	Hofmann & Rosen 1981 (& SAM II/ SAGE unpublished)
Ext. Ratio Peak Height Minus Trop. Height (mean, mid-latitude)	~10 km	"	"
Single Scattering albedo, ω	0.96-1.0	0.98-0.995	Ogren et al. 1981a,b

*Extreme volcanic values for short periods (<1 yr) may be as high at 0.1 for δ and 10^{-2} km^{-1} for β.

†For Mount St. Helens the estimate is 0.3×10^6 tonnes; for Soufriere it is 2.3×10^3 tonnes; for Agung, 3×10^7 tonnes; Fuego ≈6×10^6 tonnes. Volcanic injections may perturb composition for about 1 month to 4 months.

An example of the effects due to the 18 May 1980 eruption of Mount St. Helens is shown in Figure 4. The background optical depth at 1.0 μm is represented by the diamonds. The squares connected by the long dashes show the initial effects of the eruption, and the solid curve through the circles shows the spread in volcanic material preferentially northward. The enhancement over the initial insertion probably manifests the gas-to-particle conversion process. Figure 5 shows the zonal mean extinction for this period as a function of altitude and latitude.

Status

Although the satellite data set, complemented by increased activities in lidar, dustsonde, and progams specifically designed to delineate aerosol properties, for example, NASA's ACE (Aerosol Climatic Effects) program are providing a much better understanding of stratospheric aerosols globally, there yet remain many unanswered questions regarding their properties, formation, and effects. The exact nucleation process (or processes), for example, is not known, nor are the small and large particle size regimes. The latter is very important if one is to better understand their possible effects on climate. The exact composition, although thought to be a sulfuric acid solution, is not totally understood. Trace material studies would help determine the amount of tropospheric and meteoric material in the aerosol. More information is needed on the vertical profiles of trace gases like OH, CS_2, and OCS that go into aerosol formation. Little is known about their potential for surface or heterogeneous chemistry or if they have any effect on ozone photochemistry via a radiative coupling. Their sources and sinks, vertical and horizontal movements, and dispersion are not well known. Whether there are seasonal variations over various parts of the globe is unknown also. What is the relationship between sulfur emitted from a violent volcanic eruption and the eventual stratospheric aerosol? Can aerosols act as traces for delineating atmospheric motions? These and many more questions remain unanswered.

APPENDIX

EMPIRICAL MODEL OF STRATOSPHERIC AEROSOL OPTICAL MICROPROPERTIES

(SIZE DISTRIBUTION AND REFRACTIVE INDICES)

from

P. B. Russell et al. (1981a)

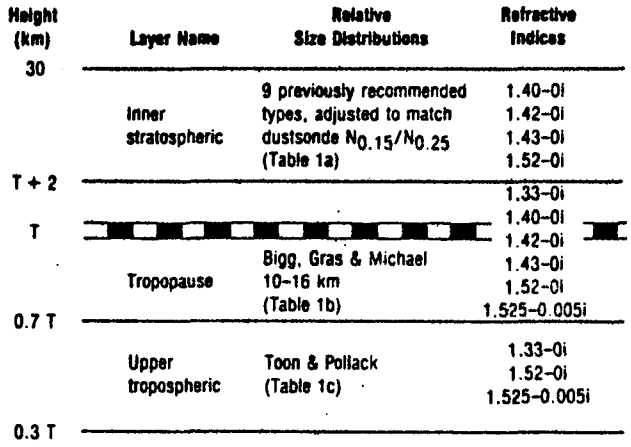

FIG. A-1. Vertical structure of model optical microproperties. T is tropopause height.

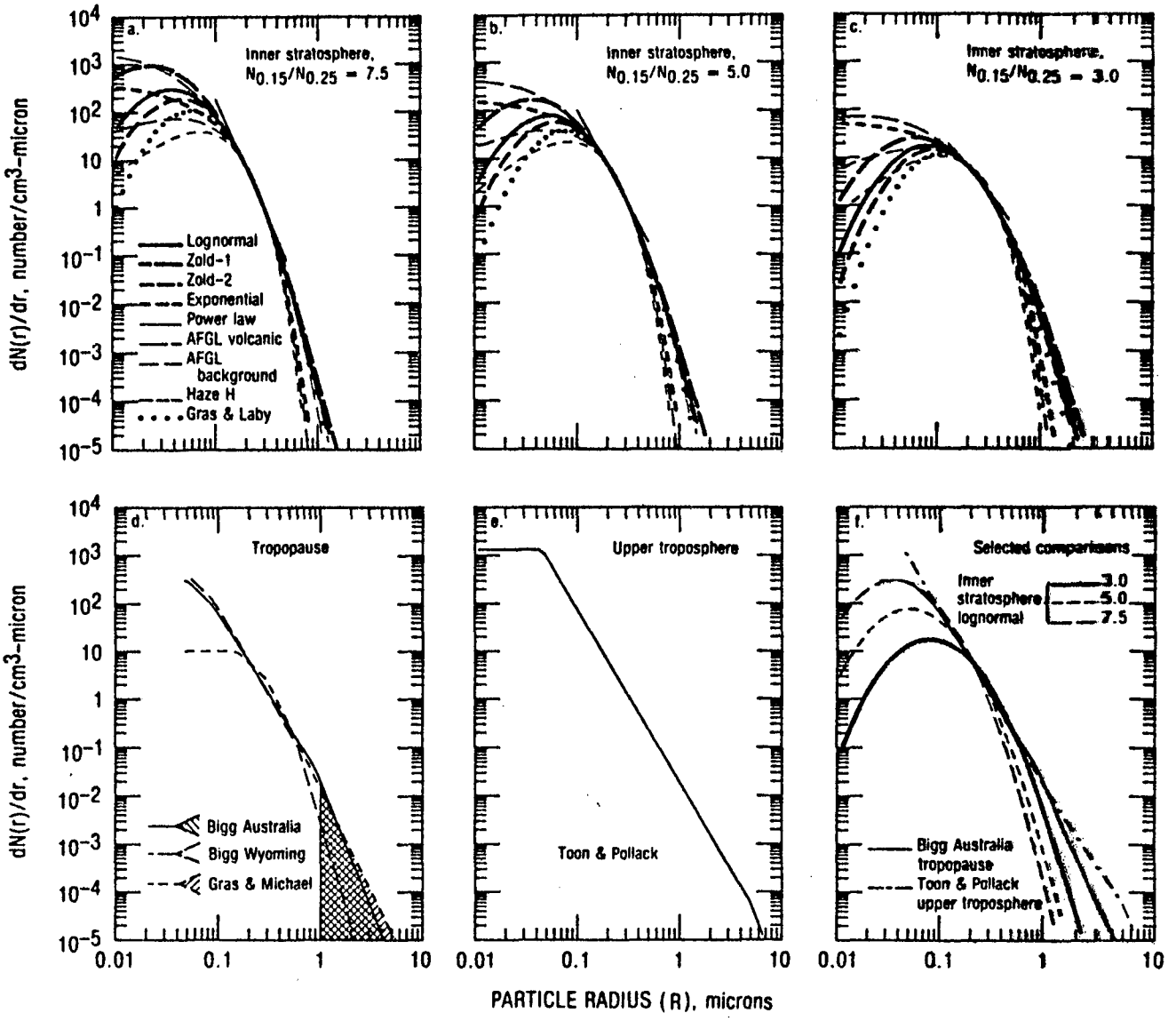

FIG. A-2. Model size distributions: (a) examples of the nine "inner stratospheric" types, each constrained to $N_{0.15} = 1\ cm^{-3}$, $N_{0.15}/N_{0.25} = 7.5$; (b) as in (a), but $N_{0.15}/N_{0.25} = 5.0$; (c) as in (a), but $N_{0.15}/N_{0.25} = 3.0$; (d) the size distributions used in the tropopause layer, each constrained to $N_{0.15} = 1\ cm^{-3}$; (f) Comparison of selected distributions from (a) through (e).

TABLE A-I. Model size-distribution types.

a. Inner stratospheric layer.

Type	Formula	Recommending reference	Recommended parameters	Parameter varied in this paper, with range
Lognormal (nonvolcanic)	$\frac{A}{r \ln \sigma_g} \exp\left[\frac{-\ln^2(r/r_g)}{2 \ln^2 \sigma_g}\right]$	Pinnick et al. (1976)	$r_g = 0.0725$ μm $\sigma_g = 1.86$ ($N_{12} = 4.9$)	$r_g = 0.412$ to 0.028 μm ($N_{12} = 1.2$ to 16)
Exponential (nonvolcanic)	$A \exp -\left[\frac{r}{r_o}\right]$	Pinnick et al. (1976)	$r_o = 0.075$ μm ($N_{12} = 3.8$)	$r_o = 0.548$ to 0.0361 μm ($N_{12} = 1.2$ to 16)
Truncated power law (nonvolcanic)	0, $r < 0.1$ μm Ar^{-p}, 0.1 μm $\leq r \leq 0.5$ μm 0, $r > 0.5$ μm	Pinnick et al. (1976)	$p = 4.0$ ($N_{12} = 5.2$)	$p = 0.362$ to 6.383 ($N_{12} = 1.5$ to 16)
Zold-1 (nonvolcanic)	$A \exp\left[\frac{-\ln^2(r/r_M)}{2 \ln^2 \sigma}\right]$	Toon and Pollack (1976)	$r_M = 0.035$ μm $\sigma = 2.0$ ($N_{12} = 5.0$)	$r_M = 0.263$ to 0.011 ($N_{12} = 1.2$ to 16)
Zold-2 (postvolcanic insoluble)	$A \exp\left[\frac{-\ln^2(r/r_M)}{2 \ln^2 \sigma}\right]$	Toon and Pollack (1976)	$r_M = 0.1$-1.0 μm $\sigma = 1.8$ ($N_{12} = 1.0$ to 2.7)	$r_M = 0.288$ to 0.025 ($N_{12} = 1.2$ to 16)

TABLE A-I. Model size-distribution types (continued).

a. Inner stratospheric layer (continued).

Type	Formula	Recommending reference	Recommended parameters	Parameter varied in this paper, with range
Zold-3 (southern hemisphere)	$A \exp\left[\frac{-\ln^2(r/r_M)}{2 \ln^2 \sigma}\right]$	Gras and Laby (1979)	r_M = 0.035-0.04 μm σ = 1.72 (N_{12} = 13-16)	r_M = 0.296 to 0.035 (N_{12} = 1.2 to 16)
Modified gamma-1 (haze H)	$Ar^{\alpha} \exp(-br^{\gamma})$	Deirmendjian (1969)	b = 20 μm^{-1} α = 2 γ = 1 (N_{12} = 3.4)	b = 4.079 to 36.527 μm^{-1} (N_{12} = 1.2 to 16)
Modified gamma-2 (AFGL background)	$Ar^{\alpha} \exp(-br^{\gamma})$	Shettle and Fenn (1975)	b = 18 μm^{-1} α = 1 γ = 1 (N_{12} = 4.1)	b = 4.079 to 32.119 μm^{-1} (N_{12} = 1.2 to 16)
Modified gamma-3 (AFGL volcanic)	$Ar^{\alpha} \exp(-br^{\gamma})$	Shettle and Fenn (1975)	b = 8 and 16 $\mu m^{-\frac{1}{2}}$ α = 1 γ = $\frac{1}{2}$ (N_{12} = 1.4 and 3.2)	b = 5,689 to 30,867 $\mu m^{-\frac{1}{2}}$ (N_{12} = 1.2 to 16)

TABLE A-I. Model size-distribution types (continued).

b. Tropopause layer

Type	Formula	Data source	Remarks
Segmented power law	Ar^{-p}, $p = \begin{cases} 0 & r < 0.05\ \mu m \\ 2.1 & 0.05\ \mu m \le r \le 0.09\ \mu m \\ 3.4 & 0.09\ \mu m \le r \le 0.09\ \mu m \\ 5.3 & 0.9\ \mu m \le r \le 1.0\ \mu m \\ 5.3\text{-}100 & r > 1.0\ \mu m \text{ (extrapolation)} \end{cases}$	Bigg (1976)	Our fit to averaged data from 35 flights, 10 to 16 km, Australia, 1969-1974
Segmented power law	Ar^{-p}, $p = \begin{cases} 0 & r < 0.05\ \mu m \\ 2.1 & 0.05\ \mu m \le r \le 0.09\ \mu m \\ 3.7 & 0.09\ \mu m \le r \le 0.6\ \mu m \\ 7.1 & 0.6\ \mu m \le r \le 1.0\ \mu m \\ 7.1\text{-}100 & r > 1.0\ \mu m \text{ (extrapolation)} \end{cases}$	Bigg (1975, 1976)	Our fit to averaged data from 6 flights, 10 to 16 km, Wyoming, 1972
Segmented power law	Ar^{-p}, $p = \begin{cases} 0 & r < 0.15\ \mu m \\ 2.0 & 0.15\ \mu m \le r \le 0.3\ \mu m \\ 4.3 & 0.3\ \mu m \le r \le 1.0\ \mu m \\ 4.3\text{-}100 & r > 1.0\ \mu m \text{ (extrapolation)} \end{cases}$	Gras and Michael (1979)	Our fit to data from four flights, 10 to 16 km, Australia, 1976-1978

c. Upper tropospheric layer

Type	Formula	Recommending reference	Remarks
Segmented power law	Ar^{-p}, $p = \begin{cases} 0 & r < 0.045\ \mu m \\ 3.6 & 0.045\ \mu m \le r \le 5\ \mu m \\ 5.6 & 5\ \mu m \le r \le 30\ \mu m \\ \infty & r > 30\ \mu m \end{cases}$	Toon and Pollack (1976)	Their fit to six published data sets

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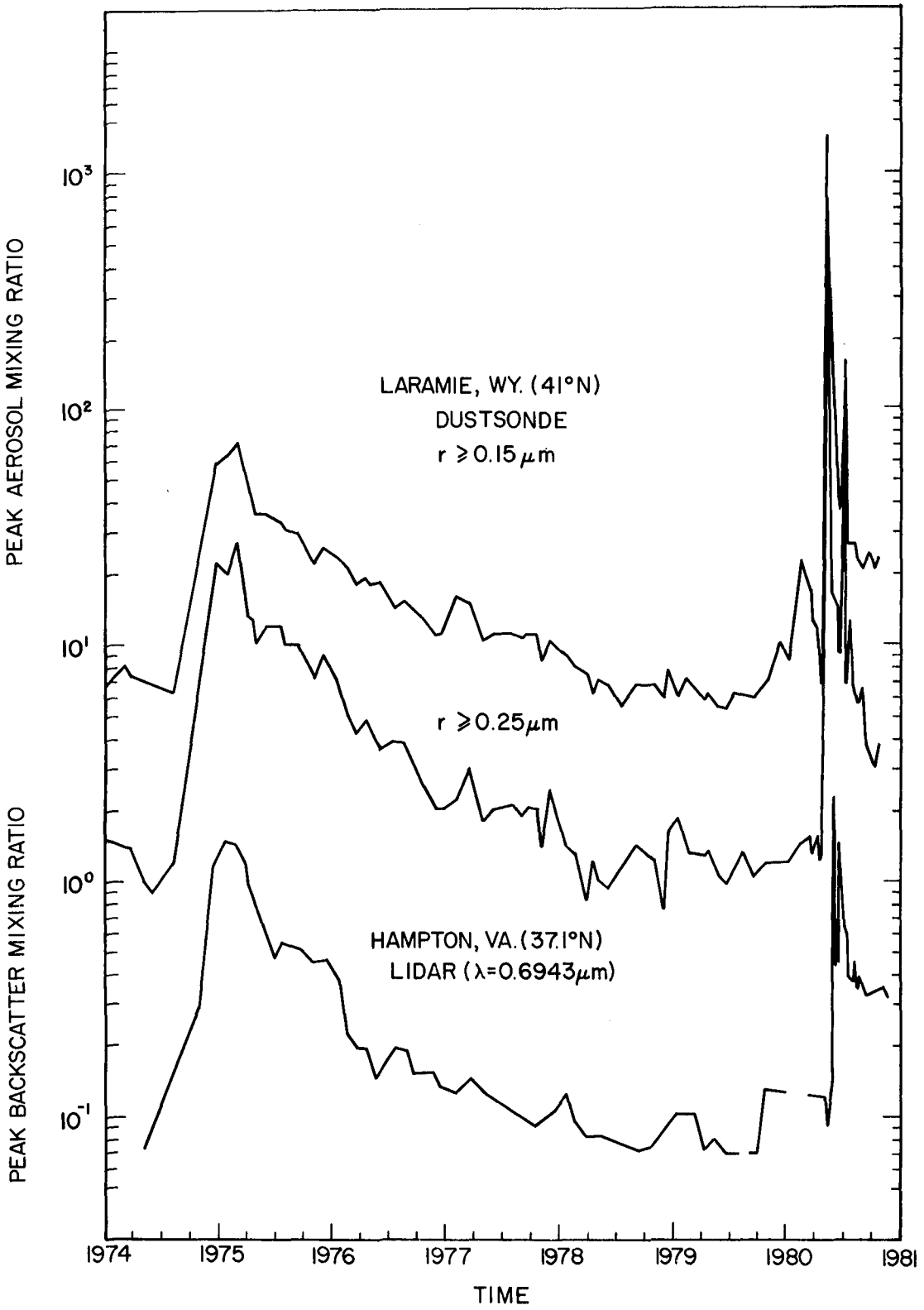

Figure 1. Peak aerosol mixing ratio from dustsonde measurements and peak backscatter mixing ratio from lidar measurements versus time from 1974 to 1981. The top curve and middle curve are the dustsonde data for particles of radius greater than or equal to 0.15 μm and 0.25 μm, respectively. The bottom curve is for lidar backscatter at the ruby laser wavelength of 0.6943 μm (from Swissler et al., 1982).

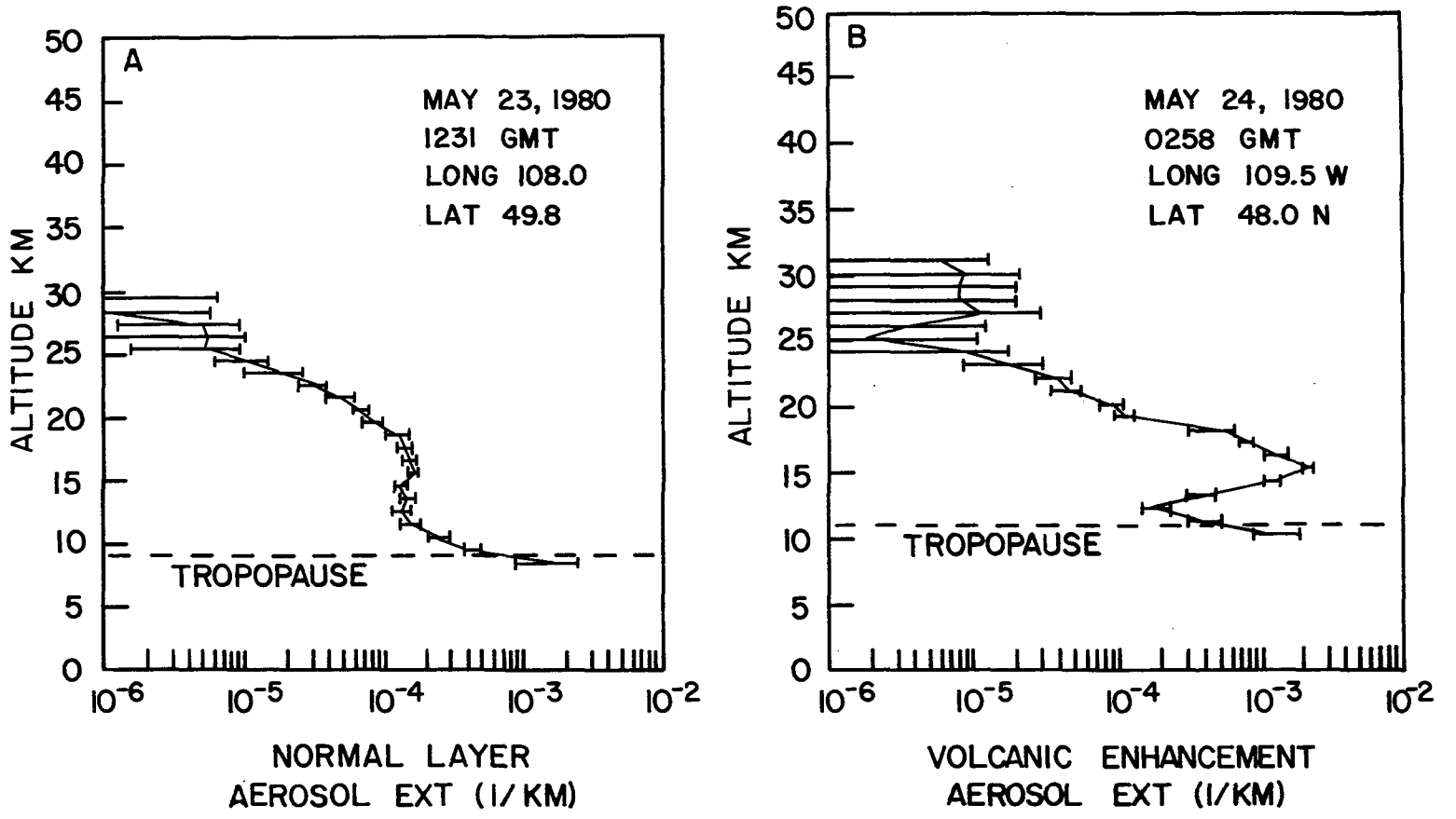

Figure 2. SAGE 1.0 μm wavelength data of aerosol extinction versus altitude for (A) a background region, and (B) a region enhanced by the volcanic effluents from the eruption of Mount St. Helens, Washington, USA.

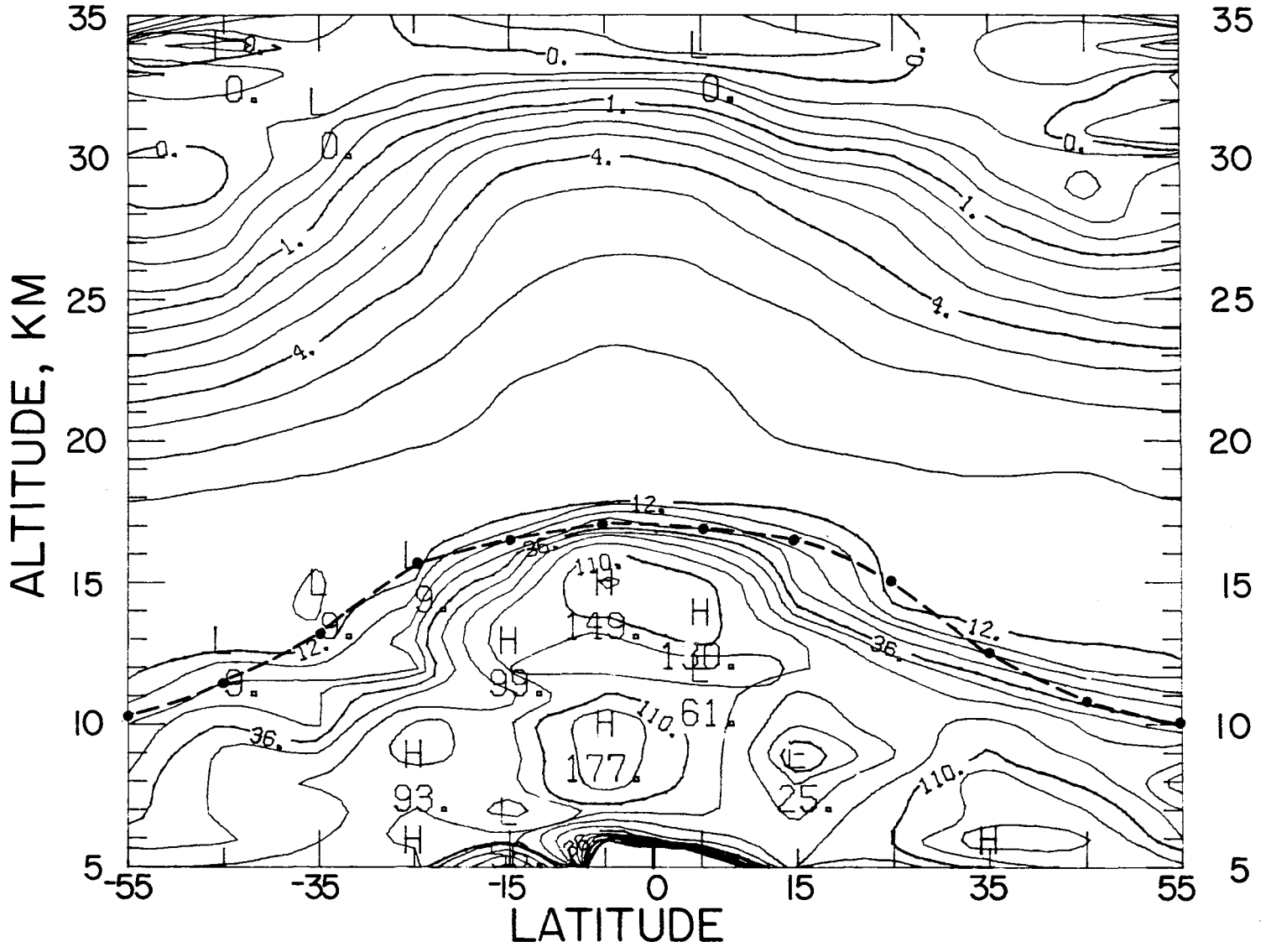

Figure 3. Zonal averages of SAGE-derived aerosol extinction at 1.0 μm for April 1979 versus altitude and latitude. Extinction contours are scaled by 10^{-5} km^{-1}. A number 4, for example, represents 4 x 10^{-5} km^{-1}. The separation between contours is scaled by a factor of 1.32. The tropopause height is marked by a dashed line.

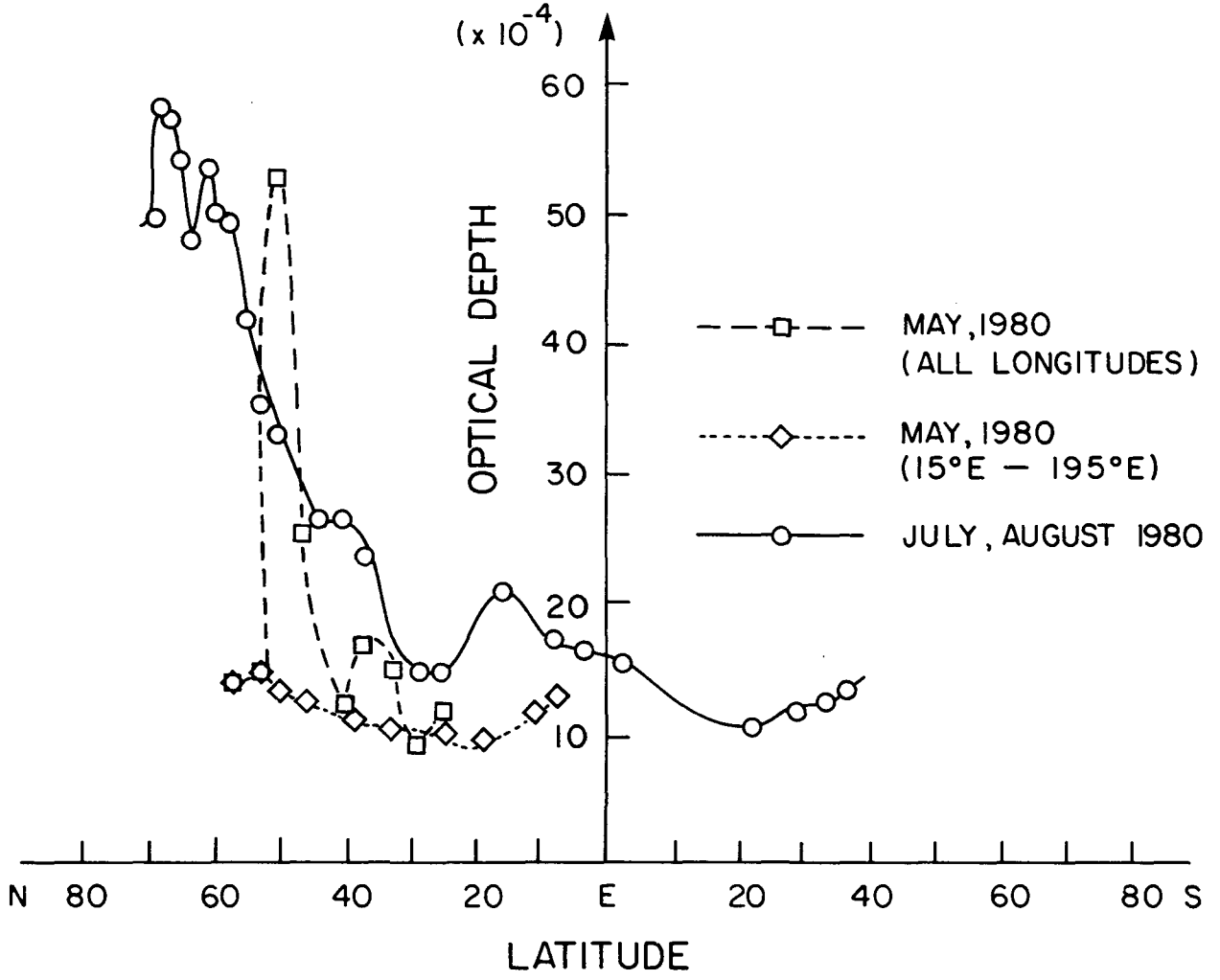

Figure 4. Stratospheric optical depth at 1.0 μm versus latitude measured by SAGE. The values in May 1980, corresponding to regions with and without the effects of the Mount St. Helens eruption, are shown with diamonds and squares, respectively. The effects of transport and gas-to-particle conversion processes are shown in the July-August data circles.

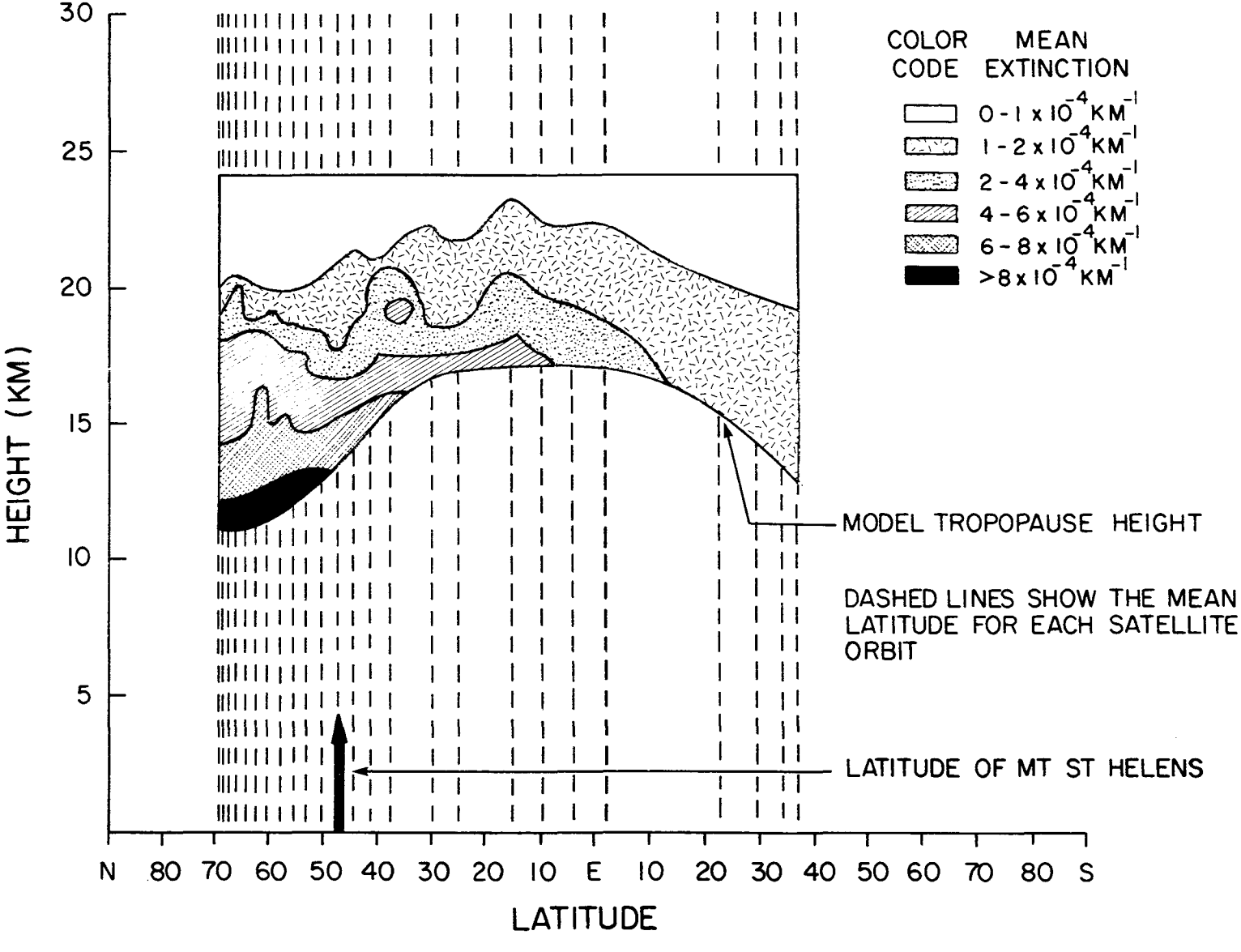

Figure 5. Zonal averages of SAGE-derived extinction at 1.0 μm for July 1 to August 16, 1980, versus altitude and latitude.

1. Report No.	2. Government Accession No.	3. Recipient's Catalog No.
NASA TM-83217		

4. Title and Subtitle	5. Report Date
Stratospheric Aerosols	November 1981
	6. Performing Organization Code 619-12-30-21

7. Author(s)	8. Performing Organization Report No.
M. Patrick McCormick	
	10. Work Unit No.
9. Performing Organization Name and Address	
NASA Langley Research Center Hampton, VA 23665	11. Contract or Grant No.
	13. Type of Report and Period Covered
12. Sponsoring Agency Name and Address	Technical Memorandum
National Aeronautics and Space Administration Washington, DC 20546	14. Sponsoring Agency Code

15. Supplementary Notes

16. Abstract

This paper presents a review of the current state of information on stratospheric aerosols. It presents, in a condensed fashion, aerosol properties such as size, size distribution, composition, refractive index, number density, extinction, optical depth, and single scattering albedo. These are generalized as much as possible to be representative of the global aerosol in times of volcanic and nonvolcanic (background) periods. Data are presented that show the global distribution of stratospheric aerosols as measured by the Stratospheric Aerosol and Gas Experiment (SAGE) satellite system for background and volcanic (post-Mount St. Helens) conditions. In addition, lidar and dustsonde data are presented that show the changes in stratospheric aerosol over an 8-year period.

17. Key Words (Suggested by Author(s))	18. Distribution Statement
Stratosphere Aerosol Volcanoes Aerosol formation and effects	Unclassified - Unlimited Subject Category 46

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price
Unclassified	Unclassified	19	A02

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