Volcanism-Climate Interactions

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NASA

MAJOR ELEMENTS OF

VOLCANISM - CLIMATE RESEARCH

RADIATIVE TRANSFER

CLIMATE MODELS

△ TEMP

△ PRECIP

△ uv B

OCEAN HEAT STORAGE AND TRANSPORT

ICE CORES
TREE RINGS

ASH

VOLCANIC DEPOSITS

O_3

ATMOSPHERIC CHEMISTRY

$SO_2 + H_2O \rightarrow H_2SO_4$

ASH , SO_2

PLUME DYNAMICS/ CHEMISTRY

Cl_2
HCl
H_2O

PETROLOGY

MAGMA

CHEMICAL REACTIONS

TECTONICS
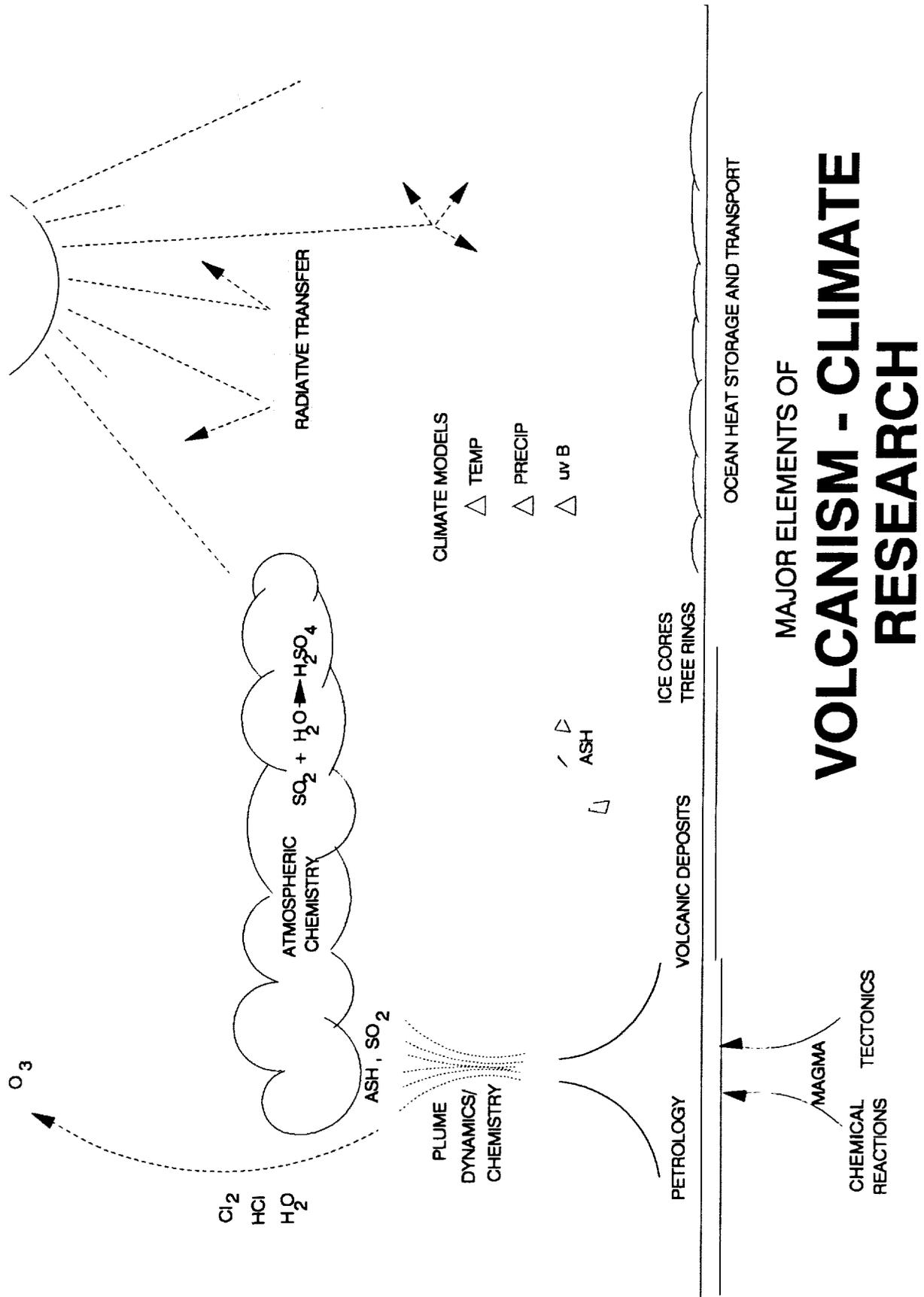
Diagrammatic representation illustrating the major components and processes involved in Volcanism-Climate Interactions.

Report of the
Workshop on

VOLCANISM-CLIMATE INTERACTIONS

held at the University of Maryland
June 18 and 19, 1990

EXECUTIVE SUMMARY

BACKGROUND

Recent recognition of the sensitivity of climate to external (particularly anthropogenic) forcings has resulted in an intensive search for causal connections in the climate system to allow reliable, objective decisions on environmental policy to be made. These connections are formulated through climate models which, in turn, are developed and tested through study of climate records ranging from the present back to past centuries. Understanding natural climate variability over this extended time period is required in order to be able to spot small, early effects of climate change and verify or adjust the models.

Historic eruptions such as Laki (1783), Tambora (1815), and Krakatoa (1883) have yielded considerable anecdotal and scientific evidence of the effects which volcanic eruptions can have on climate. While these effects are not generally themselves significant on the scale of decades, understanding their role in long-term climate trends is a major factor in the development and of climate models and the analyses needed to assess the effects of global change as quickly as possible.

For this reason, NASA, through its Geology Program and with the participation of several other federal agencies, sponsored a Workshop on Volcanism-Climate Interactions. The two-day Workshop was attended by over 30 scientists and program managers and consisted of five thematic sessions. Each session centered around a keynote presentation and featured extended and lively discussion. The agenda also included two sessions concerning research and programmatic requirements.

THEMATIC ELEMENTS

Volcano Paleoclimate (S. Self)
Investigation of the correlation between volcanism and climate is complicated by several factors. One is the incompleteness of both the climate and volcanic records; another is the difficulty in assessing the explosivity of volcanic eruptions and, finally, there are problems involved in assessing the climate impact (due to modulation of effects by the ocean; regionalization of temperature or rainfall anomalies, etc.).
Models indicate that among the most critical factors determining the effect of volcanoes on climate are the mass and height of the aerosol cloud, its residence time in the atmosphere; the size distribution of the aerosol particles

and SO_2-content of the magma and the latitude of the eruption.

Petrology and Tectonics (H. Sigurdsson)

Petrographic and chemical studies are carried out on volcanic rocks to estimate the amount of SO_2 evolved in ancient eruptions. The amount of SO_2 emitted is higher for less siliceous magmas. In the case of island arc volcanoes, excess amounts of SO_2 may reflect degassing of "unseen" magma deeply buried. An important question concerns the extent to which these observations apply to large, climate-modifying eruptions.

The chemical association of sulfur in the magma is also important in the determining the amount and speed of SO_2 generation. To a large extent, this is a function of the oxidation state and composition of the magma.

Cloud Dissipation (S. Thompson)

The evolution of the volcanic cloud will define how far it will spread and how long it, and its effects, will last. Two aspects in this evolution are important: the height to which the cloud is injected into the stratosphere and the rate of its dissipation after injection.

Convective storm models are used to simulate eruptive clouds with large source regions (e.g., effects of a 90-km-long fissure eruption). The models indicate that, for such an eruption, there would be tremendous updrafts up to 70 m/sec with considerable material injected into the stratosphere.

Aerosol Microphysics and Atmospheric Chemistry (R. Turco)

A model of the life cycle of volcanogenic sulfuric acid aerosol droplets indicates that nucleation of droplets occurs during the first months when gas concentrations are high. For the next several months, particles coagulate rapidly. Therefore, the number and size distribution of droplets constantly varies throughout the cycle. It remains to use such models in assessing the changes in optical depth caused by such aerosols.

It is possible that sulfur- and chlorine-containing volcanic gases injected into the stratosphere would affect the ozone layer either through direct reaction or by changing radiative transfer and thus controlling reaction rates .

Climate and Radiation Modeling (A. Robock)

Superimposed epoch analyses improve the signal-to-noise ratio in the volcano/climate record. Some very large eruptions, notably, that of El Chichón in 1982, have resulted in obvious warming of the stratosphere. The tropospheric signal, however, is weak or lost. By removing the ENSO (El Niño-Southern Oscillation) effect, however, the volcanic signal is better resolved and shows tropospheric cooling of 0.2-0.3°C.

FUTURE RESEARCH

The main questions to be addressed in the Volcanism-Climate Effects Research Program should be:

o   What is the effect of volcanic emissions on the global stratospheric ozone layer?

o   What are the dynamics of eruptive columns, especially those involved in historic, climate-perturbing events? To what heights can these columns

reach, especially in the cases of fissure and ignimbrite eruptions?

o   How is the magnitude of eruptions best assessed in terms of ability to evaluate climate impact?

o   What is the reason for the difference between SO_2 degassing measurements made by petrologic analyses, ice cores, COSPEC measurements and TOMS observations?

The Workshop recommends:

o   Investigation of effects of _all_ major volcanic gases;
o   Consideration of these effects at various spatial and temporal scales;
o   Development of models depicting:
    -    Pre-eruptive processes relative to volcanic gases;
    -    Mechanics and chemistry of explosive volcanism;
    -    Global dispersion of volcanic aerosols
    -    Chemical interactions in volcanic clouds;
    -    Effects of volcanic clouds on radiation balance, and,
    -    Effect of these changes on the climate process.
o   Data sets should be acquired to provide a comprehensive record of major volcanic emissions including:
    -    Eruption history (magnitudes, frequencies);
    -    Petrographic, chemical and isotopic analyses;
    -    Field studies of volcanic "plumbing" systems;
    -    Chemical and isotopic data on ice cores;
    -    Studies of tree rings, lacustrian sediments, etc.;
    -    Satellite, balloon and rocket data on atmospheric and climate variables.
o   A system should be set up for intensive, coordinated monitoring of contemporary eruptions including:
    -    A five-station network for stratospheric monitoring;
    -    Agreements for rapid aircraft deployment;
    -    Development of drone aircraft and sensors;
    -    Ground measurements of gaseous constituents;
    -    Appropriate satellite-based observations including AVHRR and improved TOMS.
o   Consideration should be given to a space mission dedicated to observation of volcanoes and their eruptions.

Many of the objectives of the Volcanism-Climate Interactions Research Program will be furthered by the implementation of several of the instruments proposed for EOS, notably SAGE, MODIS, MISER and LAWS.

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BACKGROUND

The anecdotal descriptions of the relationship between volcanic eruptions and climate change are quite descriptive. There have been many references to the "year without a summer" which was correlated with the eruption of Tambora in 1815 (Stommel and Stommel, 1983); Benjamin Franklin's observations of a "dry fog" in Europe following the 1783 Laki eruption (Sigurdsson, 1982), and the atmospheric phenomena following the eruption of Krakatau in 1883 (Self and Rampino, 1988).

The eruption of Mt. Agung in 1963 inspired a modern-day resurgence of interest in the connection between volcanism and climate. Lamb (1970) summarized the basis for this connection: the absorption of solar radiation by H_2SO_4 droplets formed from SO_2 emplaced into the upper atmosphere through explosive volcanic eruptions.

The decade following Lamb's work saw an increasing effort to numerically model global climate and to determine its sensitivity to external forcings. The more advanced models, e.g., by Pollack et al. (1976) and Hansen et al. (1978), found it necessary to include volcanic effects in order to successfully model historic climate trends.

The past decade has seen increasing awareness of the Earth as one system and an appreciation of the interacting relationships among the components of the lithosphere, hydrosphere, biosphere and atmosphere. Thus it is understood that in order to comprehend climate change it is necessary to obtain a deeper understanding of the significant forcings derived from the various Earth System components. In the case of volcanic eruptions, there is, as has been noted, the real possibility that very large events may have pronounced effects upon global-scale climate. These, added to possible regional-scale effects caused by smaller eruptions, will at least result in a significant perturbation in climate

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variations caused by other forcings. Consequently, if one is going to model the climate system, it is necessary to understand the effect of volcanic eruptions. For these reasons, "Volcanism-Climate Interactions" has been selected as an important component of the Global Change Research Program. In a large-scale program formulation meeting (called the "Coolfont" meeting) that defined options for NASA's future program in Solid Earth Sciences, the study of volcanism-climate interactions was designated as the second highest priority for the future program.

In the past decade, two other important interdisciplinary meetings were held on the subject of volcanism-climate interactions. NASA organized a Symposium and Workshop on "The Mount St. Helens Eruption: Its Atmospheric Effects and Potential Climatic Impact" (Newell and Deepak, 1982). This meeting followed upon and was largely based on a campaign of remote sensing and direct sampling carried out during the celebrated eruption of 1980. More recently, the University of Rhode Island Graduate School of Oceanography devoted a Watkins Symposium to the "Environmental Effects of Volcanism" (Palais, ed. 1986). More recently, a workshop was convened under the auspices of the International Association of Volcanology and Composition of the Earth's Interior (IAVCEI). The meeting, which took place in June 1989 in Santa Fe, NM, consisted of an international group of volcanologists and atmospheric scientists who discussed the possible climatic effects of volcanism.

With the importance accorded volcanism-climate research by the Coolfont meeting and the Global Change Research Program, NASA perceived the requirement for, and organized this Workshop on, Volcanism-Climate Interactions.

OBJECTIVES

The main objective of the Workshop was to provide an interdisciplinary forum for researchers who are actively engaged in these studies. The purpose of the forum was to enable an assessment of the current status of knowledge in the broad field and to identify gaps in this knowledge between the standard disciplines, which had to be bridged in order to make significant advances in the field. It was hoped that this assessment would permit scientists, as well as the federal agencies which fund this research, to make better decisions as to the direction of these efforts.

SCOPE

The broad, interdisciplinary nature of the investigation of Volcanism-Climate Interactions is the result of the synthesis of various phenomena. These phenomena, their interactions, and aspects of the modeling and data required to study them are depicted in Figure 1, which presents a generalized diagram showing the relationships among the pertinent disciplines, studies and research activities. Indeed, the refinement of this "roadmap" and quantification of the linkages in it may be considered a major objective of this research program.

ORGANIZATION

The Workshop was funded by NASA's Solid Earth Science Branch. It was organized by a committee consisting of Arlin Krueger (NASA/GSFC), Haraldur Sigurdsson (University of Rhode Island), Richard Turco (UCLA) and Louis Walter (NASA/GSFC). There were 34 participants (see Appendix H) from university and government laboratories as well as representatives of other Federal agencies with interests in the subject.

The meeting took place on June 17th and 18th, 1990 at the Adult Education Center of the University of Maryland. The agenda (see Appendix I) consisted of five major topical sessions covering the breadth of subjects involved in volcanism-climate interactions. Each 2-hour topical session began with a (nominal) half-hour keynote address, but this time often stretched to over an hour due to the lively discussion. (Papers written for the Workshop by the speakers are included in the Appendices.) Following the keynote talks, shorter contributed papers were offered by the participants. At the end of each day's sessions, there was a discussion of the programmatic aspects of the subject: where additional data or models are needed and the means by which these might be obtained.

The sessions were tape recorded and detailed notes of the discussion were taken by the Rapporteur, Shanaka de Silva, a volcanologist with the Lunar and Planetary Science Institute. A draft report of the meeting prepared by Dr. de Silva was redrafted by the organizing committee and reviewed and approved by all participants. The emphasis in this report is on the contributed presentations and discussions. The information imparted in the keynote addresses is conveyed in the papers included in the appendices.
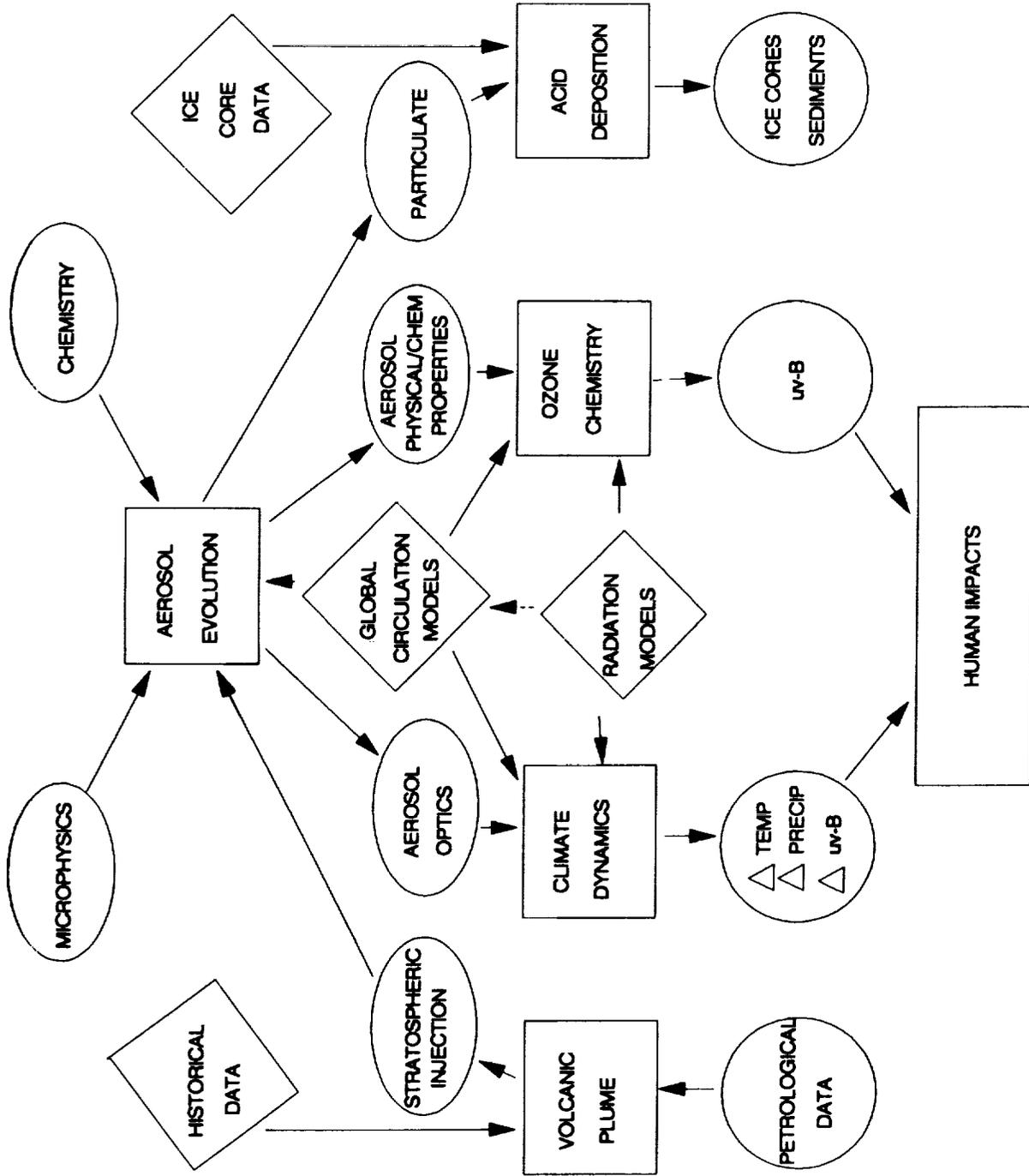
Figure 1. Generalized diagram showing the relationships among the pertinent disciplines, studies and research activities.

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VOLCANO PALEOCLIMATE

The first keynote address was presented by Stephen Self. His remarks and the ensuing discussion are abstracted in this section. Background papers prepared for the Workshop by Michael Rampino appear in Appendices A and B.

There appears to be general agreement that, unlike the greenhouse effect, which results in global warming due to an increase of CO_2 in the atmosphere in geologically short time scales, volcanic eruptions appear to be associated with global cooling on these time scales. However, the true climatic effects of volcanism are poorly understood.

The observed climatic effects of large eruptions such as Krakatau, 1883 (decrease in radiation, cooler temperatures, etc.) demonstrated a definite link between eruptions and climate and had even led to the suggestion that eruptions may contribute to the onset of ice ages (e.g., Wexler, 1951a,b). However, Rampino, Self and Fairbridge (1979) noted that it was possible to interpret the available data to infer that many major eruptions were, in fact, preceded by deteriorating or decreasing climatic trends and provocatively posed the question of whether rapid changes in climate could cause volcanic eruptions.

In general, it is difficult to quantify the effect of volcanic eruptions on climate, a major drawback being the incomplete record of both climate and volcanic eruptions. As an example, the historical records in Europe (Procopeus) and China (when 70 percent of the population died of starvation) indicate a major climate event in 536 A.D. which may have been of volcanic origin but for which the eruption is unknown. The imperfections of the volcanic record are highlighted even further when the record in ice cores of Greenland and Antarctica are considered (e.g., the unknown 1259 A.D. event), revealing volcanic acidity layers for which the sources are unknown.

There are two main problems with the various indices that have been constructed to quantify effects of eruptions on climate; circular reasoning (the use of temperature decrease to infer a volcanic eruption) involved in the formulation of Lamb's 1970 dust veil index (DVI), and the need for independent information on aerosol composition. It may be, however, that Lamb's data are broadly useful (especially for eruptions occurring in the last century), as only a few eruptions are DVI-characterized by their temperature decreases.

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The most critical factors (not necessarily in priority order) in determining the climatic effect of volcanic eruptions are:

  height of the aerosol cloud;

  mass of the aerosol;

  size distribution of the aerosol particles;

  SO_2 content of the magma;

  conversion of SO_2 gases into H_2SO_4 aerosols;

  residence time and dissipation of the aerosol cloud;

  self-limiting mechanisms for coagulation and fallout (e.g. Pinto, Turco and Toon 1989);

  the season of the eruption; and,

  the location of the eruption.

It should be stated, however, that our present focus on the role of sulfur and other aerosols has narrowed due to perceptions based on the 1982 El Chichón eruption--a very unusual case. This attention may have obscured other factors which play an important role in affecting climate.

Two major problems face volcanologists and climatologists alike; first, isolating--from natural variability--the climatic effects of known volcanic eruptions (identifying the true climatic effects of known volcanic eruptions), and second, interpreting the climatic variations in the absence of a full volcanic eruption record. The reliable volcanic record exists for only a limited period--at best for the past 150-200 years. During this period, we have experienced only moderate-size eruptions, with no large eruptions of 100 km^3 or greater which historically have occurred once every 1000 years or so. Statistical evidence suggests that for VEI-4 eruptions, the record is unrepresentative before 1940 and for VEI-5 eruptions, there is no statistically significant data. The relationship between magnitude or erupted volume of individual explosive volcanic events and their frequency per million years is shown in Figure 2, which represents subaerial volcanism only. It is evident that large eruptions are rare, and that a Tambora-size eruption may occur every 100-200 years. The figure illustrates only the total magma volume erupted, however, which is not a measure of the potential climate, as discussed later.

There is consensus that no index will be capable of telling us everything we want to know about explosive eruptions. VEI is correlated with, but not a
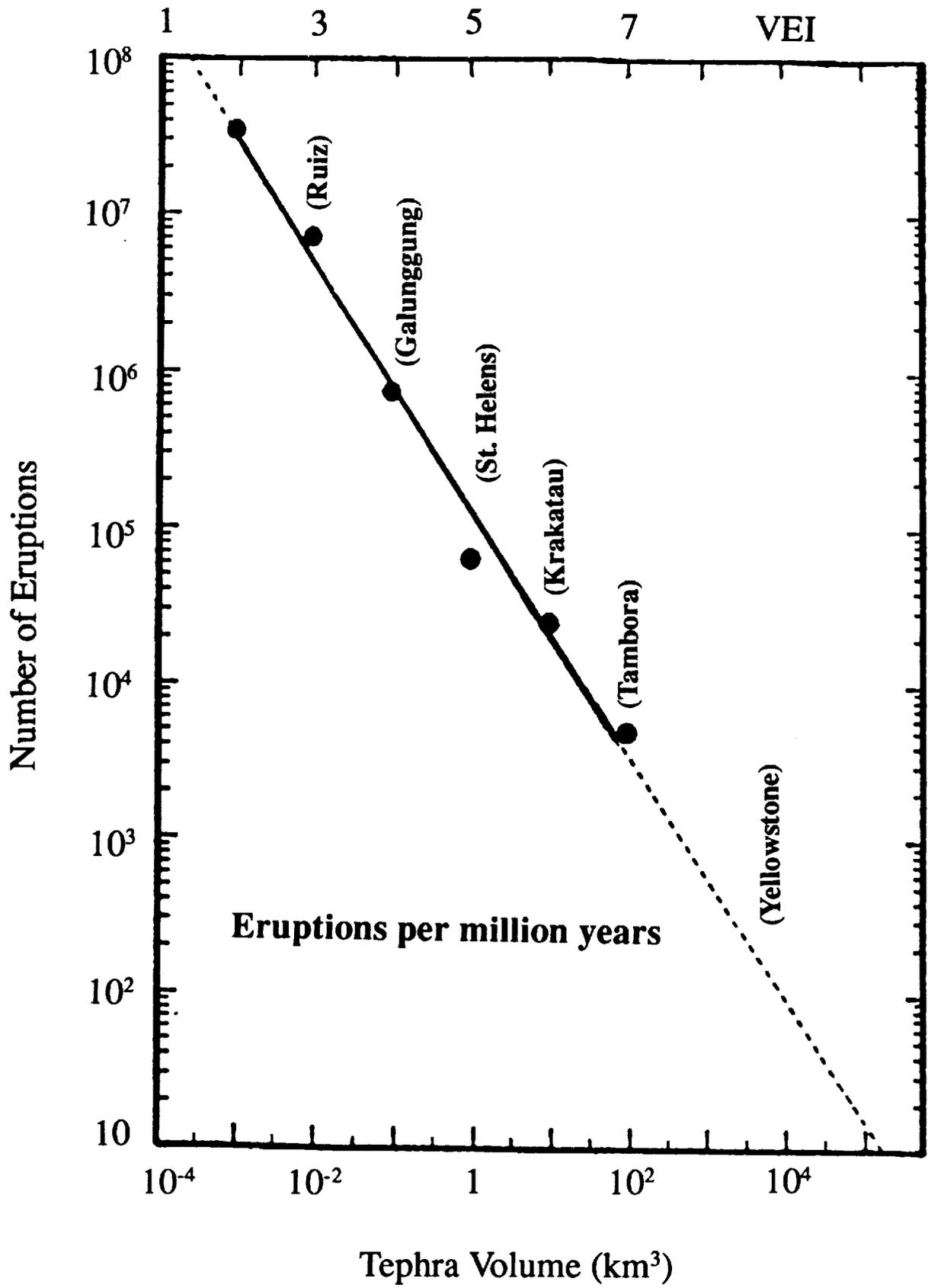
Figure 2. Relationship between magnitude or erupted volume of individual explosive volcanic events and their frequency per million years. The figure represents subaerial volcanism only.

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good measure of, the degree of SO_2 injection into the stratosphere. Measurement of the amount of SO_2 in eruptions also indicates a direct--but weak--correlation with VEI.

The completeness of the ice core record is questionable. If the sources of eruptions are known then the ice core data are useful, but using the ice core data to identify volcanic sources is more problematic, as factors other than volcanism contribute to ice acidity. However, the ice cores do contain evidence of eruptions that deposited volcanic aerosols in both Greenland and Antarctica, indicating that the volcanic aerosol dispersal and the climatic effects may not be restricted to the hemisphere of the source eruption. Work on "chemical fingerprinting" of individual eruptions in the ice core record has met with some success because there appears to be no degradation of the record with time.

However, assessing the exact direct and indirect climatic effects of eruptions is difficult. For example, the 1783 dry fogs in Europe were probably due to the eruption of the Laki fissure in 1783, but at the time, were attributed by Benjamin Franklin to Hekla in Iceland. However, once again, interpretation of the true climatic effect is complicated by the fact that 1782 was also cold due to the development of sea ice, while 1783 (as reported by Gilbert White) was a hot summer in southern England. No tree ring damage has been noted for 1783. The small (1 - 2 km^3) explosive eruption of Asama, Japan in 1783 also may have contributed to these effects.

A perplexing question is if and how a large lava-producing fissure eruption such as Laki could have emplaced the large amounts of volatiles into the stratosphere needed to explain these climate effects. Stothers et al. (1986) showed that fire-fountaining, accompanied by a convective plume, can carry material up to the high-latitude tropopause (Figure 3), implying that during flood basalt eruptions, material may be injected into the stratosphere. Our ability to model such eruptions can be (and will be) improved. In the case of an explosive (e.g., plinian) eruption, material could be injected even to stratospheric heights. Interestingly, it appears that for a dynamic eruption column, the plume seems to be unaffected by the presence of the tropopause but spreads out only when it reaches its level of neutral buoyancy in the stratosphere.

Of all the great eruptions we have known, that of Tambora, 1815, stands out. It was followed, in 1816, by "the year without a summer" (Stommel and Stommel, 1983) and such cultural idiosyncracies as Byron's morbid poems of the
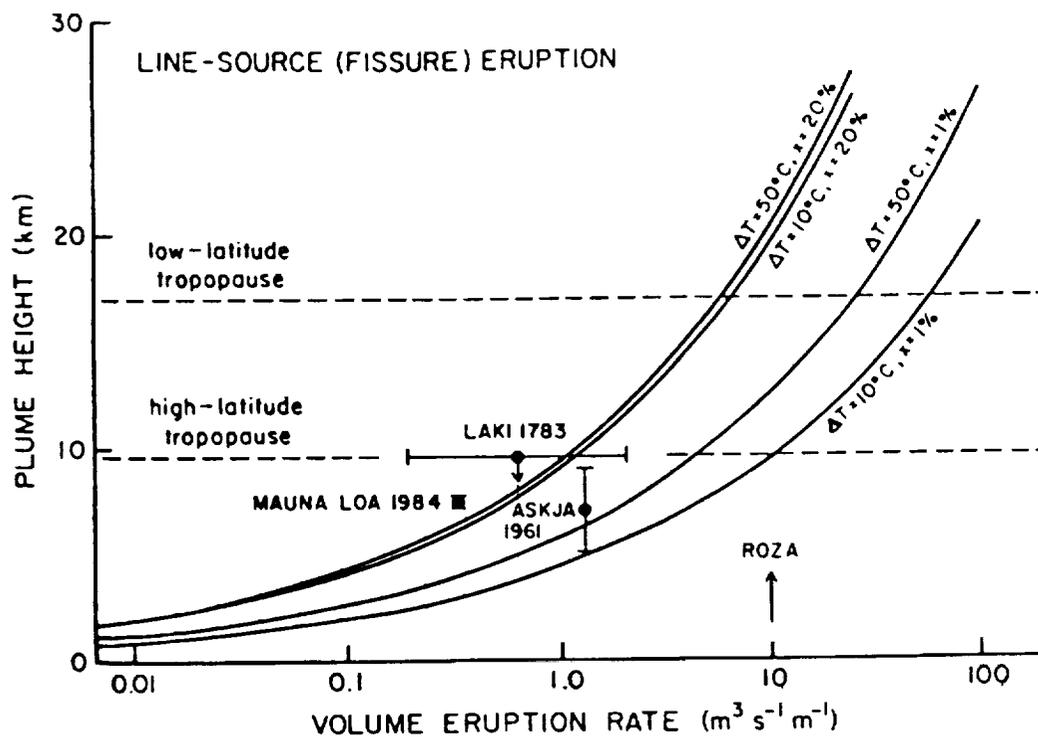
Figure 3. Convective plume height as a function of volume eruption rate per meter length of fissure for a line-source eruption. Theoretical curves are given for two values of the fine-ash content (χ) and for two values of the temperature drop of the fountain clast (ΔT) appropriate for fire-fountaining activity. Predicted plume heights for the Rosa flow eruption can be read off the observationally calibrated theoretical curves. The plotted plume height for Laki is an observational upper limit (after Stothers *et al.*, 1986) (from Rampino *et al.*, 1988)[23].

time and Mary Shelley being driven to begin work on her novel, "Frankenstein." Manley's data following the eruption show a temperature drop of 1.5°C in England. Accounts reveal failed or delayed harvests in 1816. On the other hand, many researchers have examined the climate records of the period, and in this limited data base, there is no clear (or only a small), widespread negative temperature perturbation. Other records of the time indicated higher temperatures in Europe (Landsberg) and catastrophic crop failures in China in 1816 and 1817. This raises the question as to whether volcanic effects can be resolved from other natural climatic variations and natural effects.

Angell (1988) indicated that he had not found any clear signal (i.e, temperature perturbation) of the Tambora eruption at 10 European stations. A simplistic model of the effect of attenuation of solar radiation by volcanic atmospheric aerosols would predict cooling. Interestingly, it is fairly clear that the period after El Chichón saw warming, not cooling, but the season of this eruption (April) and the El Niño-Southern Oscillation phenomenon (ENSO) of the time were likely important contributors to this opposite climatic effect.

Based on studies of annual and superimposed temperature variations, there is some consensus that there is an average, but small cooling after the largest eruptions. This cooling is in the range 0.2 to 0.5 °C, and is only short term (2 to 3 months). Climatic effects in general seem to vary with latitude, but for most eruptions (however large) the effect lasts only 2 to 3 years at most. When the effects of climate system fluctuations such as ENSO are removed, research workers such as Angell (1988) and Mass and Portman (1989) do find a definite--but small--volcanic signal (e.g., Krakatau, 1883; Coseguina, 1835). Resolving the true climate effects of eruptions requires studying the climate records for individual eruptions in greater detail. In this, we must be very careful to exclude natural climatic variations such as those due to air mass shifts and changes in circulation. Global averages may not be of much use.

The problem is that in the past, researchers have been looking at only one aspect of the problem. We must also consider factors such as the effect of the silicate particles which may persist in the atmosphere for some time after eruptions. Differences in sky polarization between Agung (1963) and Krakatau (1883) may have been a function of the Agung eruption being sulfur-rich while the Krakatau eruption plume contained a lot of mineral dust. Even large amounts of volcanic water vapor transported to the atmosphere to form long-lived clouds may

be a factor. In addition, there is evidence for high Cl levels in eruptions that might have an effect on (atmospheric) ozone. The volatile emissions from the Tambora eruption were richer in both Cl and F than S, but no evidence of enrichment in these elements was found in the ice core record. The different thermodynamic and physico-chemical properties of SO_2 and CO_2 when compared to the halogens may be the key. The halogens are more soluble and hence, may get washed out early in the evolution of the eruption column and so explain some of the halogen loss. Furthermore, they are more soluble in magmas and thus are often retained or exsolved only at the late stage of an eruption.

One wonders about the societal impact of the effect of a modern-day eruption such as that of Laki (9 months of continuous aerosol output) on airline transport. Smaller eruptions of Galungung (1982) and El Chichón (1982) had very serious effects on air traffic. A 1 to 1.5 °C temperature decrease could have a very serious effect on crops but this is a strong function of latitude. These are potentially fruitful areas of inquiry requiring interdisciplinary collaboration in order to find new approaches in the assessment of climatic effects of eruptions. For example, the records for replacements of Japan Air Lines aircraft windshields after the 1982 El Chichón eruption turns out to be highly correlated with lidar measurements of stratospheric aerosol concentrations.

Rampino et al. (1988) have compiled estimates of stratospheric aerosol amounts, optical depths, and observed hemispherical temperature decreases for past explosive eruptions (Figure 4). Two very large prehistoric eruptions were also considered as examples of different styles of eruption: the ignimbrite-forming Toba eruption and the Roza lava flow of the Columbia River basalts with aerosol loadings of 8.5 x 10^{15} g and 6 x 10^{15} g, respectively. The aerosol loadings estimated (in a simple way) for these two eruptions could have reduced sunlight to levels below which estimated photosynthesis could have occurred. The time scale of emplacement (and therefore, release of aerosol) would be an important parameter in considering effects of such eruptions. Eruption rates of Columbia River flood basalts were estimated (in previous studies) to be as high as 700 km^3/week.

It is concluded that:

Very large eruptions are capable of releasing large amounts of sulfur-rich volatiles which may affect climate. Other volatiles and aerosols are also probably important;
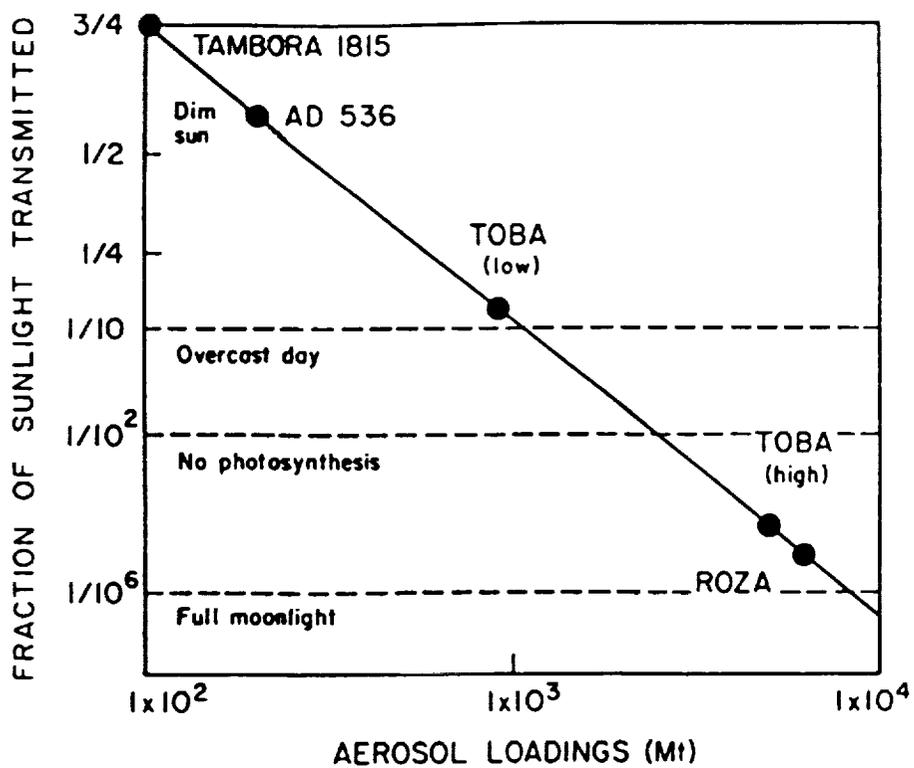
Figure 4. Fraction of sunlight transmitted through stratospheric aerosol and/or fine-ash dust clouds of different masses. Points refer to historic and prehistoric eruptions (from Rampino et al., 1988).

Explosive silicic eruptions are capable of lofting these volatiles into the upper troposphere and stratosphere (the question as to whether this can be accomplished by large flood basalt eruptions is being studied); and

Self-limiting factors may be very important. However, if even some proportion of these volatiles are transformed into aerosols, this could lead to periods of darkness and cooling which might be severe.

A question remains about the source of Cl in volcanic plumes and ice cores. Work done on Augustine has shown that many island arc magmas are very rich in Cl but isotopic evidence indicates that most of the Cl is derived from sea water. It is therefore important to look for excess Cl to distinguish it from sea salt chloride. High Cl without accompanying SO_4 is petrologically viable becuase during evolution, dacite-rhyolite magma systems will become more Cl rich while becoming S poor.

Much of the material on this subject is contained in the paper, "Sources of volcanic aerosols: petrologic and volcanological constraints" by H. Sigurdsson, which appears in Appendix C.

There is a general consensus that the potential climatic impact of a volcanic eruption is in large measure, determined by the mass of the sulfuric acid aerosol emitted to the atmosphere. The lack of control in estimating the volcanic aerosol mass and compositions for early historic and older eruptions requires that techniques need to be developed to address this issue in the absence of direct atmospheric sampling and analysis. One possible technique outlined is known as the "petrologic method" which involves estimating the sulfur loss to the atmosphere based on the difference in S content of glass inclusions in phenocrysts (which may retain the pre-eruption sulfur content of magma) and the matrix glass (which will be degassed upon eruption). The technique gives an independent estimate of the release of sulfur during an eruption but ambiguities arise for reasons such as the decomposition of sulphide minerals or the presence of a separate gas phase so that this method estimates only minimum values. One check on the method is to compare results with those derived from ice core studies but both techniques have their own (though separate) sources of error.

Conceivably, the petrologic method can also overestimate the volatile content as the magma may have largely degassed prior to eruption while the glass inclusions preserve the volatile concentrations at the time they formed. On the other hand, glass inclusions could be used as little time capsules; volatile content of inclusions coupled with an index of crystallization (such as U content) can give a useful estimate of the gas loading (not dissolved gas) of particular magmatic systems prior to eruption.

It appears that many island arc volcanoes emit excess amounts of sulfur-- they release SO_2 at rates in excess of the dissolved S content and volume of magma extruded or resident at shallow depths. Direct determinations of plume SO_2 from two Chilean volcanoes made with a COSPEC (correlation spectrometer) suggested SO_2 release rates of > 2000 tons/day. Assuming that SO_2 is lost through eruptive degassing, conversion of the observed release rates to a percentage of S lost reveals sulfur losses 50 to 100 times higher than typical S loss estimated by the "petrologic method." This requires magma extrusion rates 60 to 150 times

those observed, indicating that the excess SO_2 detected may reflect subsurface degassing of "unseen" magma deeply buried. The observation that sulfur release is an order of magnitude higher than can be accounted for by degassing of the erupted magma is certainly valid for several small, recent eruptions. In these instances, only the upper part of the reservoir is tapped of its magma, whereas gases may be emitted from the reservoir as a whole. It is not clear to what extent this relationship holds for very large and climate-modifying eruptions, when most of the reservoir magma may be erupted.

The speciation of sulfur in the magma is also important. At 100 bars in most magma chamber gases, S would be in the form of H_2S. The O_2 fugacity of most magmas at pressures greater than 100 bars are in the stability field for H_2S (not SO_2) and are unlikely to change rapidly. As pressures drop below about 50 bars, however, sulfur gas speciation favors SO_2. Rapid eruption from magma chambers containing gases at pressures over 100 bars may, therefore, release appreciable sulfur to the atmosphere as H_2S. In these cases, it is important to establish the rate of H_2S oxidation to sulfate in the atmosphere to assess potential climatic impacts.

Modeling the dynamics of large, silicic explosive volcanic eruptions (e.g., Tambora; Krakatau) is also very valuable. More work is needed on modeling the transport of the co-ignimbrite ash clouds which rise from the pyroclastic flows generated in these "ignimbrite" eruptions. Such eruptions may be able to loft material high into the stratosphere (e.g., the giant umbrella cloud of Mount St. Helens). Furthermore, although Tambora contained abundant petrologic evidence for high Cl and F in the magma, there is no such signature in the ice core record, supporting points made above that halogens may be stripped out of the eruption column at an early stage due to their higher solubility in the water vapor after it condenses. There is some indication that climate began to deteriorate even before the Tambora eruption, but the Antarctic ice cores strongly suggest that an eruption about equal in sulfuric acid aerosol content to Tambora occurred around 1809.

In order to address the question of chemical fractionation of volcanic volatiles when they are deposited on ice, studies have been performed on the material deposited through acid rain at the end of the plinian phase of the 1980 Hekla eruption. It has been found that the distribution of HCl and HF between rock and plume favors the chloride in gases and there is an excess of Si (above

that required to neutralize fluoride assuming all F as a fluorosilicate). Thus, the Si/F or Si/Cl ratios are important parameters in differentiating ice core samples stemming from the solid (particulate) or gas phase in volcanic plumes. There is a need for experimental work on gas solubilities in magma, water and ice. Additional experimental studies on reaction rates of gases in plumes are also needed.

Three types of degassing were identified at Kilauea. From summit vents noneruptive degassing emits 95% of the total CO_2 present in the magma which enters the summit magma chamber from the mantle; small amounts of SO_2 are also released. Eruptive degassing at Kilauea is dominated by H_2O (80%), SO_2 (15%), halogens and minor CO_2 (4%) (by volume). The third type of degassing occurs when lava interacts with seawater to produce HCl--the result of seawater being evaporated to dryness producing $MgCl_2$ which then reacts with water vapor (hydrolysis) to produce HCl.

CLOUD DISSIPATION

Traditional meteorological numerical modeling contributes to modeling of eruption cloud growth and evolution. The two main areas of contribution are in convective storm modeling and general circulation models (GCMs).

Convective storm models were developed to simulate thunderstorms and operate in a relatively large domain (10 - 100 km^2) using 3-D hydrocodes. They can be forced by heat sources and are therefore useful for modeling eruption clouds with large source areas such as co-ignimbrite ash clouds and fissure eruptions. These models can be used to calculate plume growth, evolution and stabilization heights and include a variety of effects such as wind shear, rotational effects, rainout, and fallout. They are particularly useful for modeling sources with arbitrary shapes and topographies. The drawbacks are that such models are extremely expensive (in terms of high-speed computer time), they cannot handle microphysics or supersonic flow and, as they are mesh-specific, they cannot be used to model sub-grid-scale phenomena such as point sources (e.g., a plinian eruption).

A specific example of this approach is a simulation of the eruption of the Roza flow of the Columbia River basalts (Tripoli and Thompson, 1988). The model utilized a regional scale domain 100 x 100 km in the horizontal and 22 km high, with a horizontal grid spacing of 2 km and a vertical spacing of 0.75 km. The fissure was assumed to be 90 km long and oriented in a zig-zag pattern and was treated as a heat and water vapor source for the lowest 0.75 km of the model. (Heat input was assumed constant though it is likely that heat input would be pulsing.) Key observations were that, as the heat source starts up, tremendous updrafts of up to 70 m/sec (at the 3-hour point) combined with the atmospheric circulation. Then, as the surface winds (65 m/sec) settled into a large cyclonic circulation the updrafts dropped to 40 m/sec (6 hr point). In this model 14% of the material was above the tropopause (11 km level) after 3 hours and 5% of the material was above it after 6 hours. The observed mesoscale circulation pattern was typical for steady heat source models. This type of circulation results in organizing the plume activity and preventing maximum potential heights being reached. Soluble tracers used in this model did not make it past the tropopause.

GCMs are basically long term climate models that are the outgrowth of

models used to predict daily and weekly weather. The latter model weather, and therefore can be used to model atmospheric chemistry, but have a fundamental predictability limit of a few weeks. In the climate mode, they typically have run times covering years to decades and they generate statistics. The models typically operate on a much coarser grid (5° x 5° x 10 levels) than convective cloud models. Several additions such as tracer transport, improved radiative transfer codes, comprehensive coupled chemistry, and coupled dynamical ocean models can be made to adapt these models for volcanism-climate interactions. Coupled chemistry will be computer-intensive, but because of global change research (for example the ozone hole problem) there is a lot more interest in this area. Atmosphere-ocean models are also computer-intensive and do not work particularly well, but will be necessary if volcanism-climate is to be linked to the occurrence of ENSOs.

Two classes of methods are used to track materials in GCMs: Lagrangian tracer release simulation models (LTRSM) and Eulerian tracer simulations (ETS). LTRSMs basically follow parcels of air around and are particularly good for small sinks or point sources. They are therefore useful for studying volcanic plumes in their initial state. Problems include cost and the fact that they need to be renormalized to counter artifacts of small-scale diffusive processes. ETSs on the other hand, have things defined all over a large grid all the time, and are therefore cheaper to run. They are best suited for looking at large sinks or source areas such as spread-out volcanic plumes. Perhaps the optimum for volcanic simulations would be a hybrid model where initially an LTRSM and then an ETM are utilized in sequence. With these models the behavior of volcanic aerosols can be studied and with modifications, highly interactive plumes, the ozone hole, and even more esoteric longer term effects such as a dust pall from an asteroid impact (e.g. Thompson and Crutzen, 1988) could also be looked at.

The inputs for these models required from volcanologists would include a database with eruptive parameters including information on plume geometry, location, volatile content, and the distribution and transport of the smallest particles.

It is suggested that in order for GCMs to usefully contribute to the issue of volcanism-climate interactions, test cases (known eruptions such as El Chichón) need to be rigorously addressed to validate the models and to examine and identify model deficiencies. Parameter space should be explored, and scaled-

up simulations of large, poorly observed eruptions should be run. Some first-order estimate of how eruption clouds may affect the ozone layer should also be run but this would require improvement in the stratospheric resolution of GCMs.

AEROSOL MICROPHYSICS AND ATMOSPHERIC CHEMISTRY

Most of the information presented on this subject is contained in the paper, "Volcanic aerosols: chemistry, microphysics, evolution and effects" by R. Turco, which is included in Appendix D.

Measurements made since 1950 during periods of volcanic quiescence show that background levels of stratospheric sulfates are increasing at the rate of 5 to 8 percent per year. The reasons for this increase are unclear but if this trend continues, it is possible that the ozone layer will be affected.

It also appears that, as time progresses, the mass of aerosol particles (generally sulfate derived from carbonyl sulfide) increases. This increase is primarily due to an increase in larger particles - those greater than about 1.5 microns in diameter.

One-dimensional models are used to characterize the behavior of volcanic SO_2 (e.g., El Chichón) and the formation of stratospheric aerosol particles. The diffusion of sulfates to levels above 30 km may not be realistic, however, because diffusion constants are actually keyed to other tracer gases. Furthermore, the simple, one-dimensional models do not take into account variations due to differences in latitude. Three-dimensional models, however, cannot consider the resolution and level of detail required.

The buildup of aerosols first depends on the achievement of a critical concentration level above which nucleation can occur. This process continues for about a month until there is enough surface area on the aerosols. This turns off the nucleation process and the particles coagulate rapidly. After about a year, particles evolve by condensation and coagulation to about 0.4-micron size. Thus, the number and size distribution of aerosol particles constantly changes with time. Removal of the particles then occurs by sedimentation.

The model agrees well with actual measurements made on the El Chichón aerosol cloud. The next step is to use these models to assess the optical depth changes caused by the presence of the aerosols resulting from larger eruptions.

The formation of the ozone hole depends, in part, on heterogeneous chemical reactions on ice particles in polar regions. This raises the question as to whether sulfuric acid droplets may cause similar reactions in mid-latitudes. Normally there is not enough acid, but during volcanic events, the higher aerosol levels could affect the ozone layer. However, better data on the sticking

coefficients are needed to make this assessment. For example, it is possible that warming associated with stratospheric injection of volcanic aerosols will decrease water concentrations and hence, prevent pervasive destruction of ozone at mid-latitudes but, in polar regions, ozone would be completely destroyed.

There is the possibility that volcanic chlorine might also contribute to stratospheric ozone depletion, but it is also likely that much of this volcanic chlorine is washed out quickly after an eruption. On the other hand, although volcanoes evolve a large amount of hydrogen, (a 100-km^3 eruption may produce 10^4 tons of H_2), it is unlikely that this will have any substantial effect in the stratosphere because of the normally relatively high abundances of hydrogen there.

The effect of the volcanic aerosols on the total radiative transfer in the atmosphere can, in turn, produce an effect on the abundance of ozone by controlling the rates of various chemical reactions. A radiative transfer model showed that the total radiation in the El Chichón aerosol cloud increased by as much as 8%. (Michelangeli et al., 1989). Combining this model with a 1-D photochemical model and comparing the differences in species concentration with and without aerosols, it was found that the chemical changes could be quite large. The model predicts depletions of ozone in agreement with the observations (3.5% with radiative effects only and up to 7% if a temperature effect is also included), but it cannot explain the observed increase of HCl (40%), and decrease on NO_x (50%). Injections of HCl, Cl_2, or H_2O could not reproduce the observed changes. Investigation of heterogeneous chemical effects has revealed that while none of the well-known heterogeneous reactions are adequate, a poorly understood system where $ClNO_3$ produces HCl (+ NOx) heterogeneously could explain most of the variations. This requires that nitric acid be dissolved in the aerosols. To study this further, more information on aerosol composition is needed. It is apparent, however, that there was a definite chemical effect caused by the El Chichón eruption, that ozone was affected by changes in radiation and temperature, and that the observed changes in volatile species required a combination of heterogeneous reactions.

Large eruptions may involve more efficient transfer of volatile components into the stratosphere as there may be insufficient time available to strip them out. It is unlikely that this would result in the formation of large, stable clouds, however, as any excess water above the vapor pressure would condense out

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onto ash particles. When these particles disperse into the ambient air, the relative humidity would be so low that the water would evaporate. However, the increase in water vapor would alter the photochemical effects of the aerosol, as well as increase its reactivity. A critical question is the injection efficiency into the global stratosphere of a major eruption (probably very high). The most important aerosol components are H_2O, sulfur gases and HCl.

While the aerosols can persist in the stratosphere for a year or longer, it appears from ice core records that deposition can occur over a relatively short period. This is attributed to seasonal effects and rainout during specific weather events. However, the speciation of the fallout is largely unknown. Other questions surround the physical and chemical nature of the deposition and differences in the deposition in the Arctic and Antarctic.

Satellite measurements of aerosols have been made following volcanic eruptions using SAM 2 and SAGE 2, which provide measurements of aerosol optical depths. The former operates at a single wavelength of 1 μm and there is now a 12-year record of polar stratospheric optical depth available. These data show a factor of 1.5 increase in 1 μm aerosol optical depth between 1978 and 1990. SAGE 2 operates at a variety of aerosol wavelengths and maps global aerosol optical depths once every 3 weeks or so and has been used for studies of the El Chichón and Ruiz plume evolutions. These data show that the Ruiz plume remained confined to a tight band around the Equator at heights between 19 and 27 km for about a year, and that fallout occurred only at high latitudes. The implication is that, for fallout to occur, the net flux of aerosol has to be polewards from lower latitudes.

Using chemical analyses of ice core samples from the time of Laki, Hekla, Tambora and Katmai, Laj et al. (1990) have identified an anticorrelation between SO_4 and H_2O_2 (Figure 5). They attribute this depletion to the reaction

$$SO_2 + 2H_2O_2 \quad ---------> \quad SO_4^= + 2H^+ + H_2O$$
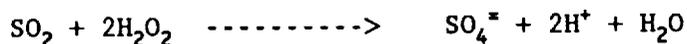
implying that a significant portion of SO_4 in the ice core record is due to oxidation of SO_2 by H_2O_2 during wet deposition. They back up this assertion with calculated SO_2 conversion rates in the stratosphere which were very slow (10^{-4} % h^{-1}), implying that only a limited amount of H_2SO_4 was present in the plume at any time and that significant SO_2 was left after transport from the eruption site to the ice. One possibility is that the anticorrelation is due to reactions which occurred in the ice after deposition, but this may be unlikely as the solubility is very low in the ice and so would be unlikely to deplete H_2O_2.
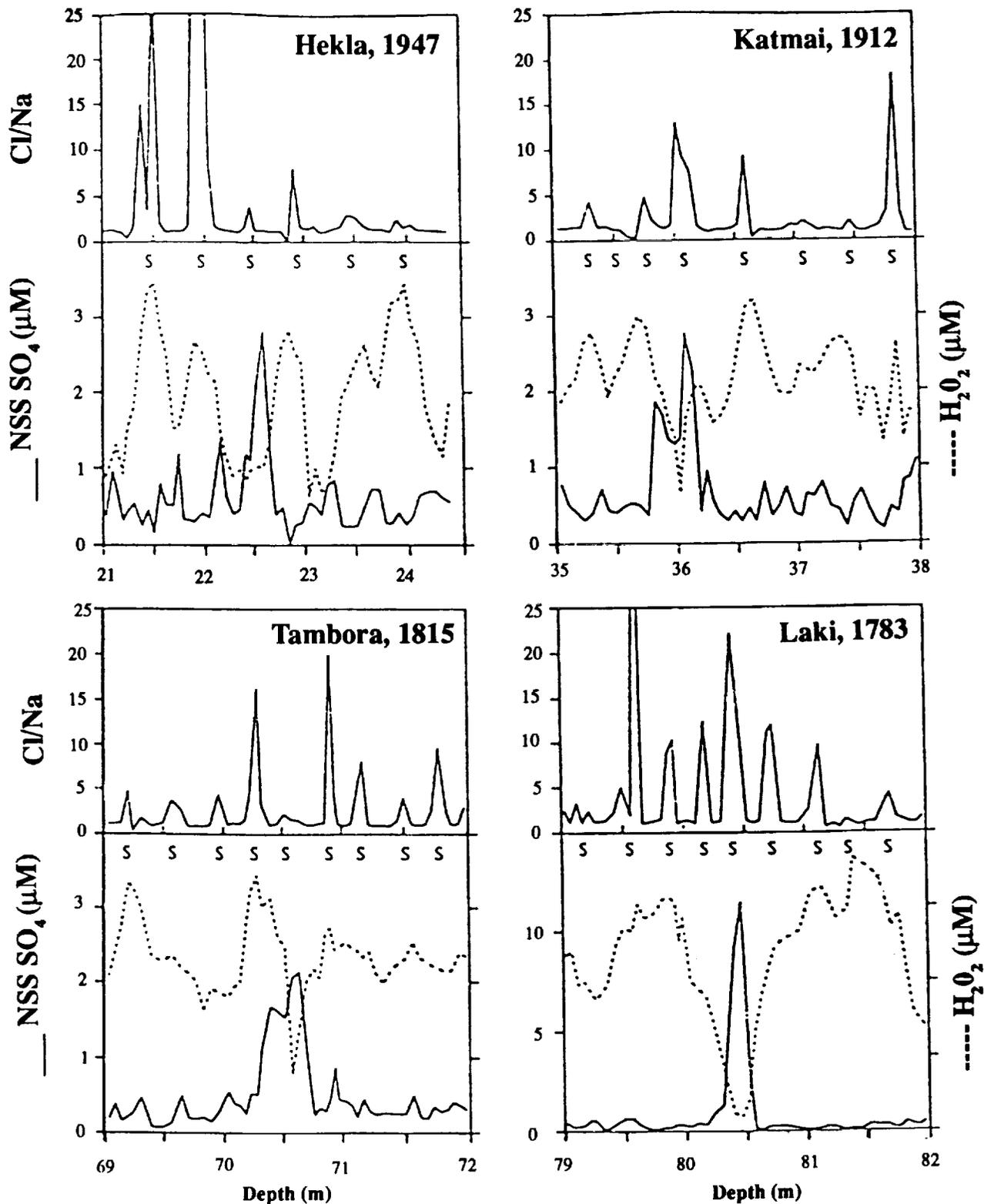
Figure 5. Variation of non-sea-salt concentration (___) SO^{2-}_4 and H_2O_2 (---) concentration as a function of depth for four sections of Greenland ice core. Summer periods, based on variation of Cl:Na ratio, are indicated by S (from Laj *et al.*, 1990).

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RADIATION AND CLIMATE MODELING

The paper in Appendix E, "Volcanic Effects on Climate" by Alan Robock served as the basis for the discussion of this subject. In it, Robock reviews the data, analyses and models linking volcanic eruptions with climate change and their potential effects on delaying and masking the effects of the increase in greenhouse gases. Records of past atmospheric loading of volcanic aerosols are unsatisfactory: methods for estimating the explosive magnitude and/or the gaseous emission of an eruption are far from accurate. Superposed epoch and seasonal cycle analyses are useful, however, in improving the signal-to-noise ratio in the volcano/climate record.

The paper notes that only global climate models are able to determine the interactions of volcanic eruptions and all of the other phenomena associated with the climate system such as the ENSO, monsoons, etc.

Stratospheric warming induced by El Chichón aerosols is apparent in rocketsonde data up to at least 31 km. The stratospheric temperature effects of other eruptions such as Agung and Fuego are not as impressive but can nevertheless be seen in the data. However, the volcanic signals in tropospheric temperatures are weak or lost. There is tropospheric cooling before Agung occurred, and 1983 (the year after El Chichón) was one of the warmest years on record. However, things improve if one removes the ENSO effect as manifested by the sea surface temperature (SST) of the eastern equatorial Pacific (Angell, 1990). The SST is strongly correlated with the tropospheric tropical, and global, air temperatures. (The SST signal precedes the global air temperature by about two seasons). By removing the SST influence on tropospheric temperature, the volcanic signals are better resolved and indicate a 0.2 to 0.3°C cooling (Figure 6). The removal of ENSO signals is of paramount importance if the true volcanic signal is to be resolved.

As noted, the largest part of the interannual climate variability is a result of the ENSO phenomena. One model attributes these events to forcing of the climatic system by decreased solar radiation at the Earth's surface due to volcanic aerosols. There is some evidence that the last 11 "strong" ENSO events have coincided with periods of decreased solar radiation and that 22 of the last 23 ENSO events had corresponding significant acid peaks in the Greenland ice core. The relationship between eruptions and ENSOs is based on an hypothesis in

GLOBE, 850-300 MB
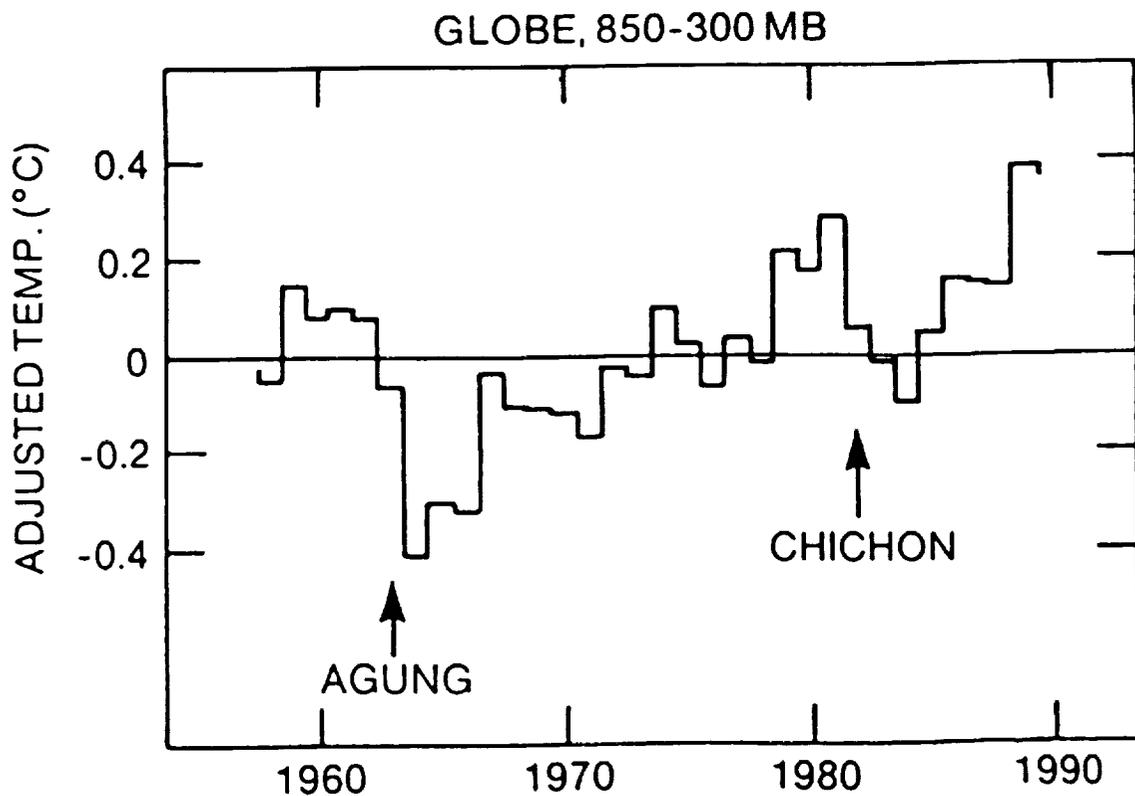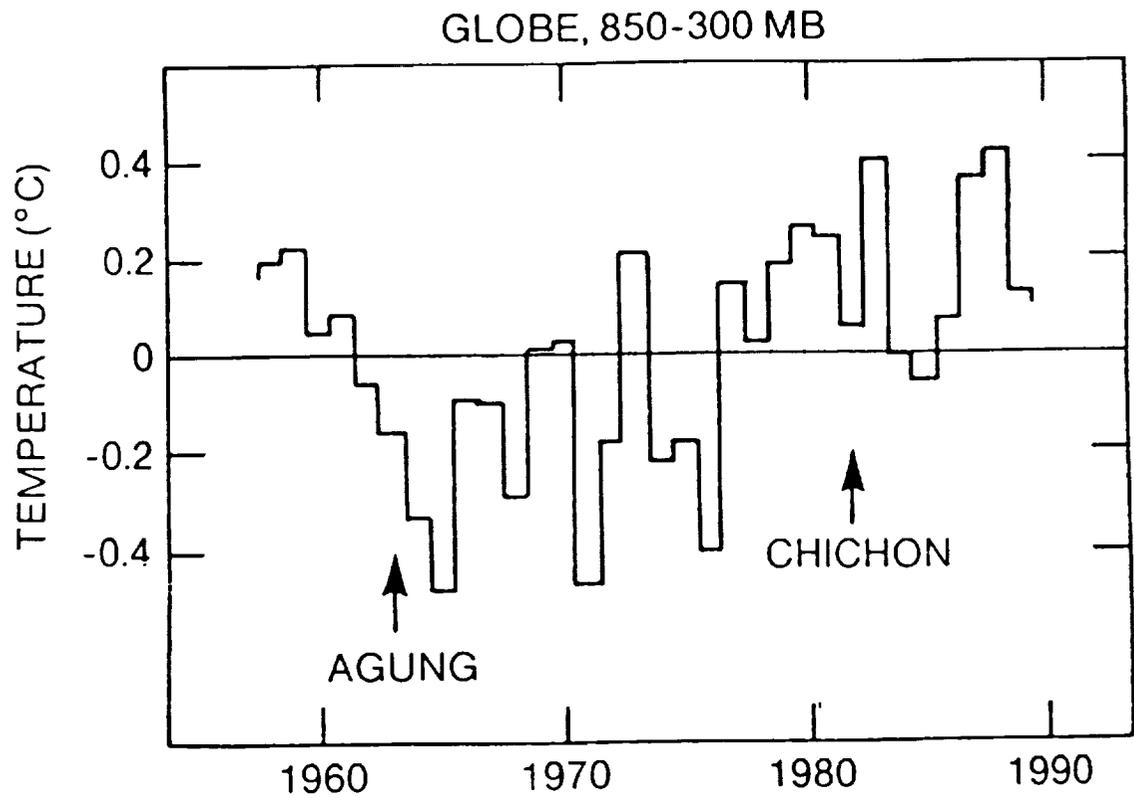
GLOBE, 850-300 MB

Figure 6. Variations in global atmospheric temperature in the region from 850-300 mb from 1958 to 1989: a.) temperatures normalized to 1958-1988 mean; b.) normalized temperatures compensated for by deviations in annual sea surface temperatures (from Angell, 1990)[24].

24. Angell, J.K., 1990, Variation in global tropospheric temperature after adjustment for the El Nino influence, 1958-89, *Geophys Res. Lett.* **8**, 1983-1096.

which the events are caused by the transfer of air mass from land to ocean modulated by changes in the solar flux brought about by absorption and reflection by volcanic aerosols. Data from DeLuisi et al. (1983) indicate a 7.7% decrease in insolation after the El Chichón 1982 eruption. However, these data were for the 1st of June after the April eruption. Averaging over a time period of 5 months is difficult, but a value of 2% might be appropriate for a 5-month period.

Various GCMs also indicate that a decrease in solar radiation is accompanied by decreased monsoonal rainfall, suggesting a correlation between decreased monsoonal rainfall and volcanic eruptions (Figure 7). The models also suggest that high-latitude aerosols will cause anti-ENSO events, while low-latitude aerosols would result in ENSO events. However, there is a large body of evidence indicating ENSO events to be quasi-periodic, the result of a natural internal oscillation and coupling of the ocean-atmosphere system. It is possible that volcanic aerosols may have a contributory effect, but are not causative. The questions surrounding these subjects are provocative and were strongly debated in the Workshop.

On the other hand, recent analyses by Jaeger and Wegley (1990) show that the Quasi-Biennial Oscillation is a major contributor to the stratospheric "noise" when attempting to isolate the volcanic signals in the climate record. When this effect is removed, climate effects due to eruptions such as 1974 Fuego and 1982 El Chichón are easily recognized (Figure 6).

Climate modeling using a multilayer energy balance model, taking into account the properties of the volcanic aerosols, predicted a surface or tropospheric cooling of some 0.6-0.7 °C after El Chichón. These results were not noticeable because of the ENSO effect discussed above. Removing this effect, one sees a decrease of only 0.2 to 0.3°C. so the model predicts a cooling which is slightly too large, probably because ocean coupling in the model is insufficient. However, it appears also that water molecules in the stratosphere are 1000 times more effective in producing a greenhouse effect than they are at lower altitudes. If the water remained at lower altitudes, this could explain the discrepancy between the model and the observations.
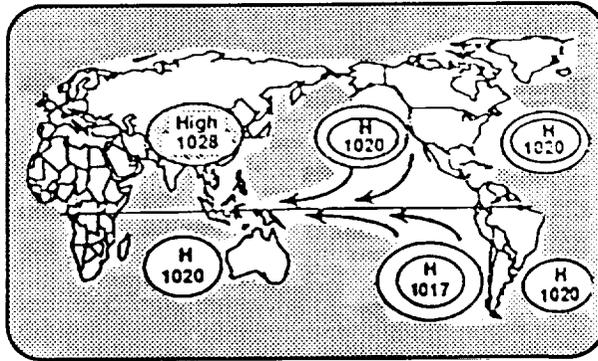
a. Northern Hemisphere Winter Conditions
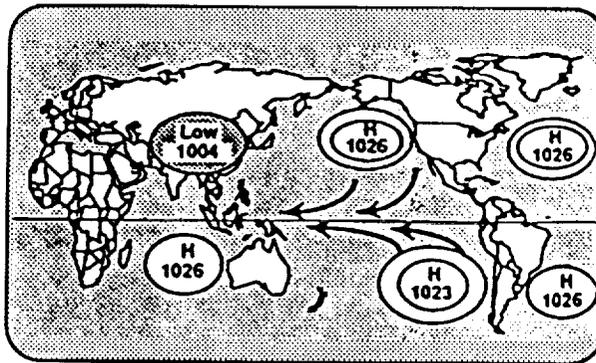
b. Northern Hemisphere Summer Conditions
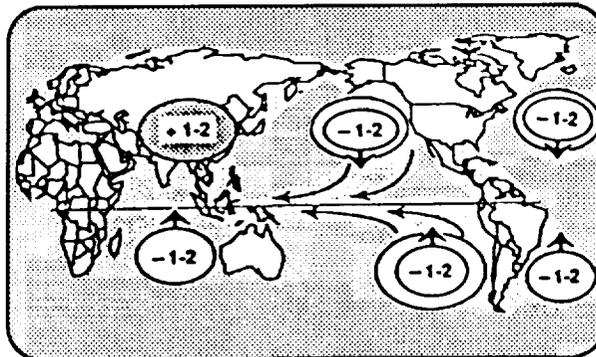
c. Approximate Deviation in mb of Central Pressure
for Conditions of Decreased Incoming Solar Radiation

Figure 7. Deviation from normal of Indian monsoon precipitation in standardized units from 1942 to 1984. Sources of low-latitude aerosol are shown on the left while sources of high-latitude aerosol are shown on the right (Handler, 1986)[25].

25. Handler, P., 1986, Stratospheric aerosols and the Indian monsoon, *J. Geophys. Res.* **91**, 14475-14490.

FUTURE RESEARCH: QUESTIONS AND DATA NEEDS

It is clear from the previous sections that the Workshop provided an excellent opportunity for assessing the state of knowledge regarding volcanism-climate interactions. It met its primary objective which was to bring together experts from diverse disciplines in an environment which would stimulate discussion and interaction. It was equally clear that the ultimate question--the degree and manner in which volcanic eruptions can affect regional and global climate--was still to be answered. Several points in this regard became clear.

With the increased importance of climate change research as a national commitment, it is now even more important to understand the volcanism-climate connection. It is just possible that the once-a-decade-size eruption (such as Agung or Fuego) might result in a sensible climatic perturbation but existing models and data are not yet sufficient to tell. However, even if the effects are not pronounced, they are nevertheless likely to be sufficiently strong and sufficiently frequent that they would create a spurious "noise" on any greenhouse warming "signal," thus making it difficult to assess the validity of greenhouse warming. For these reasons, an understanding of volcanism-climate interactions must be a component of the Global Change Research Program. More importantly, it is critical to evaluate and understand the potential major climate effects and other atmospheric impact of truly large eruptions such as Tambora 1815 and Laki 1783, which may occur on the order of once a century. Because of their rarity, we are largely ignorant of their effects, but such eruptions could potentially lead to great societal and economic disruption over large areas of the Earth.

As with the case of other Global Change studies, that of Volcanism-Climate Interactions brings together elements from diverse scientific disciplines. While this Workshop had representation from climate modelers, atmospheric chemists and volcanologists, references during the meeting to other highly relevant disciplines (such as dendrochronology, glaciology, isotope geochemistry to name a few examples) underscored their under-representation. Furthermore, because of the diversity of interests, it is virtually impossible for scientists working in all of the parts of this interdisciplinary field to come together at the regular meetings of the various societies (although major scientific organizations such as the American Meteorological Society have sponsored meetings on the subject). Thus, despite considerable hesitance to proliferate meetings, the Workshop noted

a need and strong interest to convene a meeting on the subject of Volcanism-Climate Interactions in about a year and to include representatives from those disciplines represented at this meeting as well as others such as ice studies, limnology, astronomy and dendrochronology. Such a meeting should concentrate on (though not be exclusively limited to) a central issue (e.g., the El Chichón 1982 eruption and its climate effects).

Ultimately, the key question to be answered in clarifying the volcano/climate connection is how, and to what degree, gaseous and aerosol constituents of major eruptions can modify climate. Until now, this question focused primarily on the global or hemispherical effects caused by sulfate aerosols and their role in decreasing radiative transfer through the atmosphere. The Workshop recommends:

o   Increased attention should be paid to the potential climate effects of all major volcanic gases (e.g., SO_2, H_2O, HCl, HF, H_2S, H_2, NO_x) as well as particulate aerosols.

o   Effects should be considered over a range of spatial (local, regional, hemispherical, global) and temporal (monthly, annual, multi-annual) scales, also considering the large spectrum of volcanic eruption magnitudes and types.

o   Development of models with improved capability to depict:

-    Sources of radiatively active volcanic gases and their concentration during pre-eruptive magmatic processes;

-    Mechanisms, thermodynamics and fluid mechanics of explosive injections of volcanic gases and particulates into the atmosphere;

-    Global dispersion of volcanic aerosols;

-    Chemical interactions in the volcanic cloud and its reactions with normal stratospheric constituents;

-      Effect of volcanically induced changes in the atmosphere on radiation balance;

-      Effect of these changes in radiation balance on the interlinked factors of the climate process, particularly those such as the El Niño.

The following questions should be addressed:

o   What is the effect of volcanic emissions on the global stratospheric ozone layer?

o   What are the dynamics of eruptive columns, especially those involved in historic, climate-perturbing events? To what heights can these columns reach, especially in the cases of fissure and ignimbrite eruptions?

o   How is the magnitude of eruptions best assessed in terms of ability to evaluate climate impact?

o   What is the reason for the difference between SO_2 degassing measurements made by petrologic analyses, ice cores, COSPEC measurements and TOMS observations?

Data sets should be acquired to provide a comprehensive record of major volcanic emissions of the historic past, the present, and the future by:

o   Development of a detailed database of historic eruptions through:

-      Compilation of data on eruption occurrence, magnitude, intensity, and frequency;

-      Petrographic, chemical, and isotopic analyses of volcanic ash and pristine magma samples;

-      Field studies of active and potentially active volcanoes, their

"plumbing" systems and ejecta blankets;

- Chemical and isotopic studies of ice cores representing a broad geographic and temporal distribution with increased emphasis on more recent eruptions for which historical chemical data may be available;

- Studies of tree rings, lacustrian sediments, and other proxy data sources, for evaluation of possible correlation between volcanic and climatic events;

- Acquisition and compilation of these data and available satellite, balloon, rocket and other data on atmospheric aerosol concentrations together with relative climate data focused on the decade of the '80's for intensive study of climate effects during that period.

o We recommend the intensive coordinated monitoring of contemporary eruptions, especially a major volcanic event including:

- A five-station network for stratospheric change detection (already being implemented);

- Standing equipment and agreements for aircraft monitoring (see paper entitled, "The study of atmospheric volcanic emissions" by Friend included as Appendix F);

- Development of drone aircraft and sensors;

- Ground in situ measurements and ground truth determinations of all major gaseous constituents and including, as necessary, development of ground-based, remote sensing instrumentation for measurement of volcanic gases;

- Appropriate satellite-based remote sensing including AVHRR, TOMS (designed specifically for SO_2 measurements), geosynchronous

observations and EOS instruments (see section on EOS observational requirements). Consideration should be given to the development of concepts for a mission dedicated to the observation of volcanoes and their eruptions.

Earth Observing System (EOS) Instruments

The complement of potential EOS instruments includes several of interest to Volcanism-Climate Interaction studies. Some of these pertain to the need for information on the global distribution of volcanic aerosols. A synopsis of this material, prepared by Tom Ackerman, appears in Appendix G.

SAGE is a limb-viewing instrument with a long history of previous space observations. It observes the Sun through the Earth's limb, scanning vertically through the atmosphere. It is perhaps the ideal way to obtain measurements of aerosols, particularly in thin layers. Measurements made as a function of wavelength may be used to model the size distributions of the aerosol particles and can directly determine the vertical structure of the atmospheric aerosols. However, it averages over the whole limb scan path - about 200 km - and has limited tropospheric resolution especially when clouds are present.

The MODIS instrument will present data similar to the current AVHRR in that these are moderate-resolution, nadir-looking imaging instruments. Aerosol distribution is inferred from temporal changes in apparent reflectivity or apparent haze over water bodies. Limitations are due to the spatial and temporal variability of ground albedo.

The measurement of aerosol properties and concentrations has been achieved on Venus using the Venus Polarimeter. A similar instrument is proposed for the EOS mission and should give valuable data. However, polarimetry of terrestrial aerosols is subject to larger errors because of the thinner aerosol density.

MISER will use eight cameras to observe the same place on the Earth's surface from different angles. Assuming or knowing the angular reflectivity, it is then possible, to determine the atmospheric turbidity because of the variable

path length. This instrument should provide much valuable aerosol information though the vertical definition will be limited.

The LAWS laser sounder will be able to measure the atmospheric (aerosol) backscatter coefficient. However, as scattering in directions other than 180° is important for much of the information concerning aerosols, the input from these data will have some limitations.

Fortunately, the underlying precept of the EOS mission is that instruments such as these will provide simultaneous observations and, thus, it will be possible to use all of them for optimum measurements. The SAGE instrument, for example, will be most useful for observing stratospheric aerosols at high vertical resolution while MISER and LAWS will be able to provide detailed observations and the spectral/ size distribution information. An important task remains: to coordinate all these observations with the observations of volcanic eruptions and to use the data to track the resultant aerosols and relate them to climate effects.

REFERENCES

Angell, J.K., Variation in global tropospheric temperature after adjustment for the El Niño influence, 1958-89, Geophys. Res/ Lett., 8, 1093-1096, 1990.

Angell, J.K., Impact of El Nino on the delineation of tropospheric cooling due to volcanic eruptions, J. Geophys. Res., 93 3697 - 3704, 1988.

Angell, J., Variation in global tropospheric temperature after adjustment for El Nino influence, 1958 - 1989, Geophys. Res. Lett., 17,1093-1096, 1990.

DeLuisi, J.J., E.G. Dutton, K.L. Coulson, T.E. DeFoor, and B.G. Mendonca, On some radiative features of the El Chichón volcanic stratospheric dust cloud and a cloud of unknown origin observed at Mauna Loa, J. Geophys. Res., 88, 6769-6772, 1983.

Handler, P., Stratospheric aerosols and the Indian monsoon, J. Geophys. Res., 91, 14475-14490, 1986.

Hansen, James, Wei-Chung Wang and A.A. Lacis, Mount Agung provides a test of global climate perturbation, Science, 199, 1065-1068, 1978.

Jaeger, H., and K. Wege, Stratospheric ozone depletion at northern mid-latitudes after major volcanic eruptions, J. Atm. Chem., 10, No. 3, 273-287, 1990.

Laj, Paolo, Scott M. Drummey, Mary Jo Spencer, Julie M. Palais, and Haraldur Sigurdsson, Depletion of H_2O_2 in a Greenland Ice Core: Implication for Oxidation of Volcanic SO_2, Nature, 346, 45-48.

Lamb, H.H., Volcanic dust in the atmosphere; with a chronology and assessment of its meteorological significance. Phil. Trans. Roy. Soc. London, A266, 424-533, 1970.

Mass, Clifford F., and Daniel A. Portman, Major volcanic eruptions and climate,:

A critical evaluation, J. Climate, 2, 566-593, 1989.

Michelangeli, D.V., M. Allen, and Y.L. Yung, El Chichón volcanic aerosols: impact of radiative, thermal, and chemical perturbations, J. Geophys. Res., 94, 18429-18443, 1989.

Newall, R.E., and A. Deepak, Atmospheric Effects and Potential Climatic Impact of the 1980 Eruptions of Mt. St. Helens, Langley Research Center, 1982.

Palais, Julie, editor, Environmental Effects of Volcanism, Abstract volume, Graduate School of Oceanography, University of Rhode Island, Kingston RI 02881, 1986.

Pinto. Joseph P., Richard P. Turco, and Owen B. Toon, Self-limiting physical and chemcial effects in volcanic eruption clouds, J. Geophys. Res., 94, 11165-11174, 1989.

Pollack, J.B., O.W. Toon, C. Sagan, A. Summers, B. Baldwin, and W.V. Camp, Volcanic explosions and climate change: A theoretical assessment, J. Geophys. Res., 81, 1071-1083, 1976.

Rampino, Michael R., Stephen Self,Stephen, and Rhodes W. Fairbridge, Can rapid climate change cause volcanic eruptions? Science, 206, 826-829, 1979.

Rampino, M.R., S. Self, and R.B. Stothers, Volcanic winters, Ann. Rev. Earth Planet. Sci., 16, 73-99, 1988.

Self, Stephen, and Michael R. Rampino, The relationship between volcanic eruptions and climate change: Still a conundrum?, EOS, 69, 74-83, 1988.

Sigurdsson, Haraldur, Volcanic pollution and climate: The 1783 Laki eruption, EOS, 63, 601-602, 1982.

Stommel, Henry and Elizabeth Stommel, Volcano Weather. Seven Seas Press, Newport, RI, 177 pp, 1983.

Stothers, Richard B., John A. Wolff, Stephen Self,Stephen, and Michael R. Rampino, Basaltic fissure eruptions, plume heights, and atmospheric aerosols, Geophys. Res. Lett., 13, 725-728, 1986.

Thompson, S., and P. Crutzen, Acute effects of a large bolide impact simulated by a global atmospheric circulation model. (Abst.), Global Catastrophes in Earth History; An Interdisciplinary Conference on Impacts, Volcanism and Mass Mortality, Snowbird, UT, Oct. 20-23, 1988, LPI, Contribution 673, 195, 1988.

Tripoli, P., and S. Thompson, S., A three -dimensional numerical simulation of the atmospheric injection of aerosols by a hypothetical basaltic fissure eruption. (Abst.), Global Catastrophes in earth History; An Interdisciplinary Conference on Impacts, Volcanism and Mass Mortality, Snowbird, UT, Oct. 20-23, 1988, LPI, Contribution 673, 200-201, 1988.

Wexler, Harry, On the effects of volcani dust on insolation and weather, Bull. Am. Meteorol. Soc., 32, 10-15, 1951A.

Wexler, Harry, Spread of the Krakatoa volcanic dust cloud as related to the high-level circulation, Bull. Am. Meteorol. Soc., 32, 48-51, 1951B.

Historical Evidence for a Connection Between Volcanic Eruptions and Climate Change

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Considerable evidence for a connection between volcanic eruptions and climate change exists in the form of historical and proxy records of climatic change such as instrumental temperatures, tree rings, and agricultural records, and observations of volcanic disturbances of the atmosphere such as "dry fogs" and "dim sun" conditions, and proxy evidence from acidity anomalies in ice cores. A number of studies have compared the times of historical volcanic aerosol clouds with changes in atmospheric temperatures on regional, hemispheric and global scales. These involve either a direct comparison of individual significant eruption years with temperature records, or a comparison of eruption years with composited temperature records for several years before and after a chosen sets of eruptions . The various studies give similar results--the composites show a northern hemisphere cooling of 0.2 to 0.3°C for 1 to 3 years after eruptions for a number of eruptions grouped together, and individual volcanic events that produced significant aerosol clouds such as Krakatau, 1883 or Tambora, 1816 are followed by a hemispheric cooling between 0.3 to 0.7°C for 1 to 3 years after the eruptions.

Some studies have challenged the connection between individual eruptions and climate change . Mass and Portman (1989) recently suggested that the volcanic signal was present, but smaller than previously thought. It was limited to those eruptions that created the densest aerosol clouds, and was enhanced by subtracting out other sources of interannual variability, e.g. the El Nino/Southern Oscillation. These authors stress that the volcanic "signal" of a few 10ths of a degree C is of the same order as "background" temperature variations in non-volcanic years. Moreover, it may be that stratospheric aerosol clouds have some effect on the ENSO phenomena, either triggering them, or intensifying already existing ENSO patterns. P. Handler has also suggested a connection between stratospheric aerosols and the strength of the yearly Indian monsoonal precipitation, so that climatic perturbations by volcanic aerosols may be more pervasive than commonly thought.

In a study designed to test the idea that eruptions could cause small changes in climate, Hansen and others (1978) chose one of the best monitored eruptions at the time, the 1963 eruption of Agung volcano on the island of Bali. Their observational temperature data showed a 0.5°C cooling at the surface in the tropics and a stratospheric warming (from the presence of the aerosols) of more than 6°C in the two years after the eruption. Using a simple radiation-balance model, in which they simulated an aerosol cloud in the tropics, they were able to reproduce this basic pattern of temperature change in the tropics and subtropics.

There may be natural limits to the atmospheric effects of any volcanic eruption . Pinto and others (1989) have recently proposed self-limiting physical and chemical effects in eruption clouds. Model results suggest that aerosol microphysical processes of condensation and coagulation produce larger aerosols as the SO_2 injection rate is increased (rather than a larger number of particles of the same small size). Larger particles have a smaller optical depth per unit mass than do smaller ones; they also settle out of the stratosphere at a faster rate, restricting the total number of particles in the stratospheric cloud. These processes may act to moderate the impact of volcanic aerosol clouds on the earth's radiation budget and climate, and suggest that volcanic aerosol effects might be self

limiting -- eruptions of the magnitude of Tambora, 1815 or the AD 536 eruption may be reaching that size where self-limiting effects become increasingly important.

The key to discovering the greatest effects of volcanoes on short-term climate may be to concentrate on regional temperatures where the effects of volcanic aerosol clouds can be amplified by perturbed atmospheric circulation patterns, especially changes in mid-latitudes where meridional circulation patterns may develop. Such climatic perturbations can be detected in proxy evidence such as decreases in tree-ring widths and frost damage rings in climatically sensitive parts of the world, changes in treelines, weather anomalies such as unusually cold summers, severity of sea-ice in polar and sub-polar regions, and poor grain yields and crop failures. For some eruptions, such as Tambora, 1815, these kinds of proxy and anecdotal information have been summarized in great detail in a number of papers and books. These studies lead to the general conclusion that regional effects on climate, sometimes quite severe, and unevenly distributed across the globe, may be the major impact of large historical volcanic aerosol clouds.

Climatic Impact of Volcanic Eruptions

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Volcanoes and Climate

Connections between volcanic eruptions and climatic change have been suggested on timescales from days to 10^8 years. On short timescales, one approach to the volcano/climate problem has been to compare the times of historic volcanic eruptions with changes in yearly, monthly or seasonal surface temperatures on regional, hemispheric and global scales. These studies involve either a direct comparison of the times of significant eruption years with temperature records (e.g., Rampino and Self, 1982, 1984; Stothers, 1984; Angell and Korshover, 1984; Mass and Portman, 1989) (Table 1) or a study of composited temperature records for a number of years (months, seasons) before and after a chosen set of eruptions (e.g., Mass and Schneider, 1977; Self et al., 1981, Taylor et al., 1980). Eruptions are usually chosen using various measures of volcanic intensity including the VEI (Volcanic Explosivity Index) of Newhall and Self, 1982, or the DVI (Dust Veil Index) after Lamb, 1970, or on the basis of the stratospheric aerosol loading determined directly by observations or indirectly from the acidity record of ice cores. The various studies give similar results—the composites show a Northern Hemisphere cooling of 0.2 to 0.3°C for 1 to 3 years after eruptions for a number of eruptions grouped together (Fig. 2), and individual volcanic events that produced significant aerosol clouds such as Krakatau, 1883 or Tambora, 1816 are followed by Northern Hemisphere coolings of 0.3 to 0.7°C for 1 to 3 years after the eruption (Table 1) (Baldwin et al., 1976; Rampino and Self, 1984; Stothers, 1984; Angell and Korshover, 1985). Zonally, the cooling is amplified at high latitudes. Regional records show more variability , especially meridionally.

Bradley (1988) has used monthly and seasonal temperature data, and finds that several of the larger eruptions of the past 100 years are followed by significant negative anomalies in summer and fall temperatures. Temperature decreases after major eruptions are found to be abrupt and short lived (1 to 3 months), with a recurrence of the cooling about 12 and 24 months after the eruptions. The maximum effect of about 0.4°C occurs in the summer and fall months immediately following the eruptions, and falls off in the same seasons over the next 2 years.

Mass and Portman (1989) recently suggested that the volcanic signal was present in temperature records, but smaller than previously thought. It was limited to those eruptions that created the densest aerosol clouds in the last 100 years, and was enhanced by subtracting out other sources of interannual variability, e.g. the El Nino/Southern Oscillation. Mass and Portman stressed that the volcanic "signal" of 0.1 to 0.2°C is of the same order as "background" temperature variations in non-volcanic years, and found no evidence of large coolings in the first few months after the eruptions. It may be, however, that stratospheric aerosol clouds have some effect on the ENSO phenomena, either triggering them, or intensifying already existing ENSO patterns (Handler, 1984). Handler (1986) has also suggested a connection between stratospheric aerosols and the strength of the yearly Indian monsoonal precipitation .

These studies have attempted to "isolate" the volcanic signal in noisy temperature data. Thyis assumes that it is possible to isolate a distinct volcanic signal in a record that may have a combination of forcings (ENSO, solar variability, random fluctuations, volcanism) that all interact. The key to discovering the greatest effects of volcanoes on short-term climate may be to concentrate on temperatures in regions where the effects of volcanic aerosol clouds may be amplified by perturbed atmospheric circulation patterns. This is especially true in sub-polar and mid-latitude areas affected by changes in the position of the polar front. Such climatic perturbations can be detected in surface temperatures and in proxy evidence such as decreases in tree-ring widths and frost rings, changes in the treeline,weather anomalies such as unusually cold summers, severity of sea-ice in polar and sub-polar regions, and poor grain yields and crop failures (for a review see Rampino et al., 1988). In low latitudes, sudden temperature drops have been correlated with the passage overhead of the volcanic dust cloud (Stothers, 1984). For some eruptions, such as Tambora, 1815, these kinds of proxy and anectdotal information have been summarized in great detail in a number of papers and books (e.g., Post, 1978; Stothers, 1984; Stommel and Stommel, 1986; C.R. Harrington, in press). These studies lead to the general conclusion that regional effects on climate, sometimes quite severe, may be the major impact of large historical volcanic aerosol clouds.

Tambora and 1816: The Test Case?

Instead of searching for the small climatic effects of small historic eruptions in climate data sets, it may be instructive to look in detail at the strongest volcanic perturbation in recent history, the Tambora eruption of April, 1815 and the events that followed it. Perhaps

smaller eruptions (in terms of aerosols released) might have similar, but less extreme, effects on climate that can be detected if one knows what to look for. Stothers (1984) estimates an optical depth of about 1.0 following the Tambora eruption, and the presence of large acidity peaks in ice cores from Greenland and Antarctica argues for a global disperesal of the aerosol cloud of some 10^{14} g. In most studies of weather records or proxy data, the year 1816, following the Tambora eruption, displays the strongest signal of possible "volcano weather" in historical times. For example, in a key area in the Eastern Hudson Bay region in midsummer 1816, the reduction in mean daily temperature from the long-term average was about 5 to 6°. It has been suggested that a reduction in mean daily temperature in this range could lead to the formation of perennial snowcover in northern Canada (Wilson, 1985). The negative anomaly in July 1816 brought the absolute mean temperature to below $5^{\circ}C$, more than 2° lower than the modern record in 1965. The median height of the freezing level above eastern Hudson Bay in July 1965 (two years after the Agung eruption) was about 600 to 700 m lower than the modern 10 year value. This suggests that the $-6^{\circ}C$ anomaly in 1816 might have produced a 1000 m drop, to bring the freezing level to within 1500 m of the surface. Such conditions were maintained for only two years, 1816 and 1817, but one may wonder how many years of consecutive extreme seasonal weather would be required to establish an effective snowcover, and bring ice/albedo feedback into play. Frequent and extended snowfall also took place in the region in 1816-1817— in 1816, there were only five weeks without measurable snowfall (mid-July to mid-August). Furthermore, the atmospheric circulation pattern was dominated by a high pressure area over the surface of Hudson Bay, which remained ice covered through the summer of 1816 (Wilson, 1985).

Catchpole and Faurer (1983) show sea ice patterns in 1816 that are also consistent with a highly meridional atmospheric circulation pattern over eastern North America, showing strong north to northwest winds in July and August of 1816 (Catchpole and Faurer, 1983). Northwesterly winds brought cold air southward into the northeastern United States, bringing the "Year without a Summer". The meridional circulation pattern also brought cold weather to western Europe, and there is some evidence that, between these two waves, warmer weather dominated, with an opening of the usually ice-covered Greenland Sea between 74° and $80^{\circ}N$ (Wilson, 1985).

It should be pointed out however, that the cold weather in the Hudson Bay area began before April 1815, with unusually cold years beginning in 1811/12, so that the Tambora eruption cannot be the only cause of the climate shift. Perhaps the coincidence of the

Tambora eruption with a time of low sunspot numbers (indicating lower solar activity) led to the unusually cold weather of the 1811-1817 period.

Frost rings in trees in the Western U.S. also show a correspondence with years of volcanic eruptions (LaMarche and Hirschboeck, 1984). These may indicate outbreaks of exceptionally cold weather during the summer months, again possibly the result of increased meridional circulation. Light rings in subarctic trees from northern Quebec also show a correspondence with volcanic eruptions; for example, damage is most widespread in 1816-1817 (Filion et al., 1986). Tree rings have also been used to create synoptic summer temperature patterns for Europe, which show significantly cooler summers from 1812 to 1816 (Briffa et al., 1988). The same kinds of data can be assembled for the other major volcanic aerosol clouds of the last 200 years, and similarities noted. If one can establish patterns that appear to be characteristic of "volcanic" perturbation, then it may be possible to detect such patterns even for the smaller eruptions, and to see how well climate models simulate such perturbations. It may be necessary not only to model the volcanic perturbation, but also to include the effects of ENSO events and possible solar variations in the model runs.

VOLCANISM AND LONG-TERM CLIMATE CHANGE

One problem with studying historical eruptions is that they are quite small compared to those in the geologic record. The largest explosive eruption in historic times was probably the Tambora, 1815 explosion, with about 50 km^3 of erupted magma. This can be compared with large ignimbrite forming eruptions such as the Toba event of 75,000 yr BP, which erupted more than 2,800 km^3 of magma (Rose and Chesner, 1987). For effusive eruptions, the largest historic event was Laki, 1783, which produced 12 km^3 of basalt, compared with great flood basalt eruptions like the Roza flow in the Columbia River Group (14 Myr BP), composed of 700 km^3 of basaltic lava in a single flow (Devine et al., 1984; Rampino et al., 1988). Both of these eruptions may have released about 10^{16} g of sulfurous gases. The effects of these and other pre-historic eruptions on climate are not known. Both the Roza and Toba events appear to coincide with climatic coolings, but no cause and effect relationship has been established.

If historic eruptions can cause small changes in climate, then perhaps larger eruptions or groups of eruptions can cause major climate change. A number of authors have claimed correlation between volcanic eruptions and glacial fluctuations on 10^3 to 10^5 year time

scales. For example, Bradley and England (1978) and Porter (1981,1986) proposed that second-order "Little Ice Age" glacial advances during the Holocene could have been driven by eruptions, since the glacial advances correlate with peaks of acidity in polar ice cores. Porter (1981) estimates that a global cooling of $1^{\circ}K$ could lead to a snowline depression sufficient to cause glacier advances equivlent to those of the last several centuries, but this seems to be greater than the measured volcanic perturbations. Bray (1977, 1979a, b) proposed that large eruptions preceded glacial periods, with the glaciations triggered by atmospheric aerosol clouds. However, in this case, because of the inaccuracies in dating, it is often difficult to determine which came first, the eruptions or the glaciation.

On even longer timescales, studies by Kennett et al (1977) have delineated pulses of explosive volcanic activity in the Circum-Pacific region for the last 30 million years from studies of age determinations (mostly K/Ar dates) on igneous rocks, and counts of ash layers in deep-sea sediment cores. They find significant pulses of volcanism in the Plio-Pleistocene (about 2 Myr BP), latest Miocene to Early Pliocene (6 to 3 Myr BP), Late Miocene (11 to 8 Myr BP) and Middle Miocene (16-14 Myr BP). Combined with other data (e.g., Hein et al., 1978), it seems that pulses of widespread (perhaps global) explosive volcanism took place near 0.5, 2.5, 5, 10, 15, 20 and 40 Myr BP (Kennett et al., 1985). Some of these spurts in volcanism correlate with times of apparent pulses of plate motion and sea level changes (Masuda, 1986; Rampino and Stothers, 1987), so the three may be linked. These volcanic pulses can be also be correlated with times of global cooling and ice formation as seen in oxygen-isotope data for the Cenozoic. Flood basalt episodes at 65, 35, and 17 Myr BP also correlate with global coolings (Devine et al., 1984). This apparent correlation of volcanic episodes with times of global cooling may be coincidental, however, and there is no direct cause and effect mechanism to suggest beyond possible ice/albedo feedback.

Recommendations

1. Look at the strongest aerosol perturbations, e.g. Tambora, Krakatau, Agung, El Chichon, etc. Don't average together the effects of small and large eruptions.

2. Use regional and seasonal climate data sets where the effects of volcanism on climate may be amplified.

3. Study the spread of aerosol clouds for volcanic eruptions in different parts of the world at different times of the year. Compare with tracer models.

4. Look for changes in atmospheric circulation patterns that could be studied with GCMs. Observed temperature series and tree-ring studies could provide synoptic weather patterns.

5. Establish a new volcanic index based on VEI and data from ice cores, petrologic studies, optical depth measurements, etc.

6. Look at climate records that have both climate and volcanic signals, e.g. ice cores.

REFERENCES

ANGELL, J.K., AND KORSHOVER, J., 1985, Surface temperature changes following th six major volcanic episodes between 1780-1980: Journal of Climate and Applied Meteorology, v. 24, p. 937-951.

BALDWIN, B., POLLACK, J.B., SUMMERS, A., TOON, O.B., SAGAN, C., AND VAN CAMP,W., 1976, Stratospheric aerosols and climatic change: Nature, v. 263, p. 551-555

BRADLEY, R.S., AND ENGLAND, J., 1978, Influence of volcanic dust on glacier mass balance at high latitudes: Nature, v. 271, p. 736-738.

BRAY, J.R., 1974, Volcanism and glaciation during the past 40 millenia: Nature, v. 252, p. 679-680.

_____ , 1977, Pleistocene volcanism and glacial initiation: Science, v. 197, p. 251-254.

BRIFFA, K.R. et al., 1988, Summer temperature patterns over Europe: A reconstruction from 1750 AD based on maximum latewood density indices of conifiers: Quat. Res., v. 30, p. 36-52.

BRYSON, R.A., 1989, Late Quaternary volcanic modulation of Milankovitch climate forcing: Theoretical and Applied Climatology, v. 39, p. 115-125.

CATCHPOLE, A.J.W., AND FAURER, M-A., 1983, Summer sea ice severity in Hudson Strait, 1751-1870: Climatic Change, v. 5, p. 115-139.

DEIRMENDJIAN, D., 1973, On volcanic and other particulate turbidity anomalies: Advances in Geophysics, v. 16, p. 267-296.

DEVINE, J.D., SIGURDSSON, H., DAVIS, A.N., AND SELF, S., 1984, Estimates of sulfur and chlorine yield to the atmosphere from volcanic eruptions and potential climatic effects: Journal of Geophysical Research, v. 89, p. 6309-6325.

ELLSAESSER, H.W., 1986, Comments on "Surface temperature changes following the six major volcanic episodes between 1780 and 1980": Journal of Climate and Applied Meteorology, v. 25, p. 1184-1185.

FILION, L. ET AL., 1986, Light rings in subarctic conifers as a dendrochronological tool: Quat. Res., v. 26, p. 272-279.

HANDLER, P., 1984, Possible associations of stratospheric aerosols and El Nino type events: Geophysical Research Letters, v. 11, p. 1121-1124.

RAMPINO, M.R. AND STOTHERS, R.B., 1987, Episodic nature of the Cenozoic marine record: Paleoceanography, v. 2, p. 255-258.

_____ , 1988, Flood basalt volcanism during the past 250 million years: Science

ROSE, W.I., JR., AND CHESNER, C.A., 1987, Dispersal of ash in the great Toba eruption, 75 ka: Geology, v. 15, p. 913-917.

SELF, S., RAMPINO, M.R., AND BARBERA, J.J., 1981, The possible effects of large 19th and 20th century volcanic eruptions on zonal and hemispheric surface temperatures: Journal of Volcanology and Geothermal Research, v. 11, p. 41-60.

STOMMEL, H., AND STOMMEL, E., 1983, Volcano Weather: Newport, R.I., Seven Seas Press.

STOTHERS, R.B., 1984, The great eruption of Tambora and its aftermath: Science, v. 224, p. 1191-

_____ , AND RAMPINO, M.R., 1983, Volcanic eruptions in the Mediterranean before A.D. 630 from written and archaeological sources: Journal of Geophysical Research, v. 88, p. 6357-6371.

TAYLOR, B.L., GAL-CHEN, T., AND SCHNEIDER, S.H., 1980, Volcanic eruptions and long-term temperature records: An empirical search for cause and effect: Quarterly Journal of the Royal Meteorological Society, v. 196, p. 175-199.

TOON, O.B., AND POLLACK, J.B., 1980, Atmospheric aerosols and climate: American Scientist, v. 68, p. 268-278.

TURCO, R.P., TOON, O.B., WHITTEN, R.C., HAMILL, P., AND KEESEE, R.G., 1983, The 1980 eruptions of Mount St. Helens: Physical and chemical processes in the stratospheric clouds: Journal of Geophysical Research, v. 88, p. 5299-5319.

WILSON, C., 1985, The Little Ice Age on Eastern Hudson/James Bay: the Summer weather and climate at Great Whale, Fort George and Eastmain, 1814-1821, as derived from Hudson's Bay Company records: Syllogeus, No. 55, p. 147-190.

_____ , 1986, Stratospheric aerosols and the Indian monsoon, Journal of Geophysical Research, v. 91, p. 14,475 -14,490.

HANSEN, J.E., WANG, W.C. AND LACIS, A.A., 1978, Mount Agung eruption provides test of a global climatic perturbation: Science, v. 199, p. 1065-1068.

HEIN, J.R., SCHOLL, D.W., AND MILLER, J., 1978, Episodes of Aleutian ridge explosive volcanism: Science, v. 199, p. 137-141.

KENNETT, J.P., MCBIRNEY, A.R., AND THUNELL, R.C., 1977, Episodes of Cenozoic volcanism in the circum-Pacific region: Journal of Volcanology and Geothermal Research, v. 2, p. 145-163.

LAMARCHE, V.C., AND HIRSCHBOECK, K.K., 1984, Frost rings in trees as records of major volcanic eruptions: Nature, v. 307, p. 121-126.

LAMB, H.H., 1970, Volcanic dust in the atmosphere; with its chronology and assessment of its meteorological significance: Philosophical Transactions of the Royal Society of London A, v.266, p. 425-533.

LANDSBERG, H.E., AND ALBERT J.M., 1974, The summer of 1816 and volcanism: Weatherwise, v. 27, p. 63-66.

MASS, C.F. AND PORTMAN, D.A., 1989, Major volcanic eruptions and climate: A critical evaluation: Journal of Climate, v. 2, p. 566-593.

_____ , AND SCHNEIDER, S.H., 1978, Statistical evidence on the influence of sunspots and volcanic dust on long-term temperature records: Journal of the Atmospheric Sciences, v. 34, p. 1995-2008.

MASUDA, F., 1986, "Vail sea-level curve" records the ages of plate motion changes: Annual Report of the Institute of Geoscience, the University of Tsukuba, No. 12, p. 64-67.

NEWHALL, C.G., AND SELF, S., 1982, The Volcanic Explosivity Index (VEI): An estimate of explosive magnitude of historic eruptions: Journal of Geophysical Research, v. 87, p. 1231-1238.

PORTER, S.C., 1981, Recent glacier variations and volcanic eruptions: Nature, v. 291, p. 139-141.

PORTER, S.C., 1986, Pattern and forcing of northern hemisphere glacier variations during the last millennium: Quaternary Research, v. 26, p. 27-48.

RAMPINO, M.R., AND SELF, S., 1984, Sulphur-rich volcanic eruptions and stratospheric aerosols: Nature, v. 310, p. 677-679.

RAMPINO, M.R., SELF, S. AND STOTHERS, R.B., 1988, Volcanic winters: Ann. Rev. Earth and Planet. Science: v. 16, p. 73-99.

SOURCES OF VOLCANIC AEROSOLS:
PETROLOGIC AND VOLCANOLOGICAL CONSTRAINTS

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Contents:

Summary

Global climatic effects brought about by volcanism are related to the impact of volcanic gases and their derivative aerosols on the atmosphere, rather than the effects of volcanic ash. Evidence from both historic eruptions and polar ice cores indicates that volcanic sulfur gases are the dominant aerosol-forming component, resulting in production of a sulfuric acid-rich stratospheric aerosol that can have profound effects on the Earth's radiation budget over periods of a few years. Due to highly variable sulfur content of different magma types, the climatic effects do not relate simply to total erupted mass. There is a close relationship between volcanic sulfur yield to the atmosphere and hemispheric surface temperature decrease following an eruption, with up to 1 $^{\circ}$C surface temperature decrease indicated following a major volcanic event such as the 1815 Tambora eruption. While the erupted mass of HCl and HF is equal to or greater than that of sulfur gases in some

volcanic events, the halogens do not form known aerosols nor are they abundant in ice core acidity layers. The early removal of halogens from eruption columns occurs by rain flushing and adsorption onto tephra particles, but the fate of halogens in the atmosphere following very large explosive eruptions is unknown. CO_2 flux to the atmosphere from volcanic eruptions is volumetrically one of the most important of the gas species, but owing to the huge size of the atmospheric reservoir of this gas, the volcanic contribution is likely to have negligible effects.

Introduction

Due to the effects of volcanic aerosols on the Earth's radiation budget, volcanic activity is frequently cited as one of the principal forcing functions that lead to short-term climate change. Two principal types of atmospherically transported particles of volcanic origin are recognized: (a) silicate dust particles, derived from the quenching of magma to glass and its fragmentation to volcanic ash, and (b) sulfuric acid aerosols derived from the conversion of volcanic sulfur dioxide gas to sulfuric acid liquid particles. The earliest documented recognition of the effects of volcanic eruptions on climate was made in 1784 by Benjamin Franklin, who observed the cooling effects of a "dry fog" on the atmosphere over Europe following the great Laki fissure eruption in Iceland in 1783.

Initial assessment of the effects of volcanism were based on the assumption that the silicate dust particles or volcanic ash played a major role in climate change by backscattering incident solar radiation and increasing the planetary albedo, leading to a net cooling of the Earth. Direct sampling of the stratospheric volcanic aerosol following the 1963 Agung eruption showed, however, that it was composed principally of sulfuric acid. The minor role of the silicate component of the volcanic aerosol in climate modification is in large measure due to its short atmospheric half-life, because of rapid silicate ash aggregation that leads to fallout of clumps of even the finest tephra within a few hundred to one thousand km from the source.

Volcanic aerosols from explosive eruptions can have at least six important effects on the Earth's atmosphere, some of which with opposite surface temperature signal. During fallout of tephra and condensation of volcanic gases in the troposphere, volcanic activity leads to increased condensation nucleii, cloud formation and thus increased tropospheric albedo, causing local surface cooling. Secondly, a stratospheric sulfuric acid aerosol formed by SO_2 gas-to-particle conversion leads to increased backscatter of solar radiation and surface cooling. This is likely to be the most significant volcanic effect on global climate, and recent studies show that the magnitude of cooling of the Earth following major volcanic eruptions is controlled by the sulfuric acid aerosol mass. Third, enhanced stratospheric absorption of solar radiation by the volcanic aerosol can also contribute to surface cooling. Fourth, a volcanic aerosol containing large ($\geq 1\mu m$) silicate particles, which cause infrared heating, can lead to surface warming which may balance the cooling effect of a sulfuric acid aerosol, but this warming is likely to be short-lived or negligible due to very rapid fallout of such coarse silicate ash.

Fifth, increase in greenhouse gases, such as CO_2 from a volcanic source can contribute to surface warming. As shown below, this factor is likely to be negligible due to the small mass of volcanically derived CO_2 emissions, compared to the large anthropogenic source of this gas. Finally, volcanic components, such as HCl and other halogen gases and magmatic water vapor, may contribute to stratospheric ozone depletion, due to reactions of volcanic odd-chlorine and hydroxyl molecules with ozone.

Volcanic eruptions of sufficient magnitude to have significant climatological effects occur about once a century. Recent and frequent minor events such as the 1963 Agung eruption in Indonesia, whose atmospheric effects have been well monitored, are two orders of magnitude smaller than the great eruptions, such as Tambora in 1815. Such large-magnitude events are capable of producing short-term climate change and may thus severely impact the biosphere and influence the future of mankind. The study of past events of this type has been hampered by paucity of observations, but the recent development of techniques to extract data on past eruptions of this magnitude, both from their volcanic deposits and from ice cores, has resulted in the development of a data base on source parameters of major explosive volcanic eruptions. Results of these studies show that the magnitude of observed surface cooling after large volcanic events correlates with the mass of sulfur degassing during the eruption.

Source Parameters

A volcanic eruption represents rapid transfer of matter and energy from the Earth's interior to the surface and to the atmosphere. The critical source parameters for evaluation of the atmospheric and global effects of an eruption include the mass eruption rate (intensity), total erupted mass (magnitude), eruption column height, and the type and mass of volcanic aerosols (principally sulfuric acid and water vapor) and other but non-condensible volatile components released to the atmosphere, principally CO_2, HCl and HF.

The first three source parameters can be determined from characteristics of the proximal volcanic deposit from an eruption. Figure 1 shows that a typical range of column height of fifty recent explosive eruptions is 10 to 50 km, corresponding to magma discharge rate or intensity of 10^6 to 10^9 kg s^{-1}. The range in magnitude or total erupted mass may reach values as high as 10^{15} kg, or of the order 400 km^3 of magma. Rare high-magnitude events with 10^{16} kg total erupted mass (~4000 km^3) are present in the geologic record when one considers volcanic activity during the past one hundred thousand years, such as the great Toba eruption in Indonesia about 75,000 years before present. At present, magma eruption rate estimates are only available for peak rates of plinian explosive eruptions; rates during ignimbrite phases of explosive eruptions are unknown but significantly higher.

Ash Fallout

Fallout from major explosive eruptions can blanket vast areas of the Earth's surface with a layer of volcanic ash. Very large explosive eruptions have produced ash fallout blankets that cover over 10^4 km^2 with 1 m of ash, and up to 10^5 km^2 with 10 cm thick ash. The environmental effects of volcanic ash blankets of this magnitude include increase in the Earth's albedo and possibly consequent surface cooling. Effects of ash fall on agriculture can also be very severe, both in terms of crop damage and also because of the harmful effects of ash ingested by grazing livestock. Fallout from some volcanoes is often high in fluorine, due to adsorption of fluorine from the eruption plume on ash particles, thus leading to poisoning of livestock by fluorosis.

Volcanic eruption plumes which reach levels of the upper troposphere and lower stratosphere down-wind from active volcanoes can be very hazardous to aircraft. Serious incidents occured in Indonesia in 1982, when two large commercial jet aircraft flew through the eruptive plume of Galunggung volcano at 11 km altitude and suffered severe engine damage that resulted in emergency landings. Volcanic ash and sulfur-rich volcanic aerosol clouds in the stratosphere are also known to cause severe windshield damage to commercial aircraft, such as after the 1982 El Chichon eruption in Mexico. The eruption columns reached heights of 21 to 25 km, but the stratospheric plumes gradually subsided due to sedimentation and conversion of SO_2 gas to sulfuric acid aerosol particles, until the widespread aerosol plume enveloped the flight paths of intercontinental jets at 8 to 12 km altitude, about nine months after the eruption. The windshields of commercial aircraft flying at this altitude suffered deterioration due to corrosion and abrasion by the volcanic aerosol, leading to over fifty times higher rate of windshield replacement during 1983 and 1984 than in non-volcanic periods.

Sulfur Sources

Because of the predominant role of volcanic sulfur in atmospheric impact, it is essential to evaluate the origin of this element and the factors that control its output from volcanic sources. The behaviour of sulfur in magmas is complex, as it occurs in four oxidation states and in different physical states, as solid, liquid or in gaseous form. The four oxidation states of sulfur in geological environments are S^{2-} (in sulfides and H_2S volcanic gas), as S (elemental sulfur), as S^{4+} (in SO_2 volcanic gases) and as S^{6+} (in SO_3 volcanic gases and sulfates). While sulfur is relatively abundant in our planet, or about 2.7 wt.% of the bulk Earth composition, its distribution is rather uneven due to its complex chemical and physical behaviour. Thus sulfur is highly concentrated in the deep Earth as dense sulfides, while it is also enriched in the oceans as the sulfate anion (0.265 wt.% of sea water).

The yield of sulfur to the atmosphere during volcanic eruptions depends on a number of factors, such as the total erupted mass of magma, its pre-eruption sulfur solubility, the efficiency of sulfur exsolution before quenching of the erupted magma, the presence of sulfur-rich mineral phases that may break down

and release a sulfur-dominated gas during eruption, and the presence of a sulfur-dominated volatile phase in the magma reservoir.

The evolution of sulfur in magmas is intimately linked with the fO_2, fSO_2 and activity of iron in the melt. Thus basaltic magmas, characterized by high temperature (1200°C) and low fugacity of oxygen (10^{-10}) are generally saturated with a sulfide phase, which occurs as imiscible droplets in equilibrium with the silicate melt, which may crystallize upon cooling (pyrrhotite; FeS). An understanding of the behaviour of sulfur in imiscible systems is important not only for sulfur volatile emissions from major volcanic eruptions, but also because of the partitioning of so-called chalcophile elements into the imiscible sulfide phase, leading to the concentration of such metals in the Earth as Fe, Ni, Pd, Ir, Pt and other platinum group elements, and formation of economically important magmatic sulfide mineral deposits. As sulfide minerals are present in a wide variety of volcanic rocks, it is likely that the majority of magmas that have evolved at low fO_2 are sulfide saturated. The sulfides in the erupted magmas are, however, in extremely low abundance, probably largely due to their high density and tendency to sink and accumulate at the base of magma bodies. Sulfur may also occur as a free fluid phase in magmas, but direct evidence for such discrete vapor (fluid) sulfur phase is as yet lacking.

In a melt with a separate sulfide phase, the sulfur dissolved in the silicate melt decreases with increasing O content of the melt, due to the fact that sulfur dissolves by displacing oxygen bonded to Fe^{++} and the increasing oxygen results in an increase in Fe^{+++} at the expense of Fe^{++} in the melt. In natural magmas which contain a free imiscible sulfide phase, there is a strong positive correlation between the sulfur dissolved in the silicate melt and the FeO and to lesser extent TiO_2 content of the melt, and the sulfur content increases with decreasing fO_2 at constant FeO. A typical range of sulfur dissolved in natural basaltic magmas is from about 1000 ppm at 9 wt. % FeO, to 2000 ppm at 15 wt% FeO, but sulfur content of silicic and other Fe-poor magmas is significantly lower, as shown in Figure 3.

Sulfur solubility is also sensitive to temperature at constant fO_2 and FeO content; thus the dissolved sulfur increases by factor of three to eight per 100°C temperature increase in the range 1000 to 1400°C. Sulfur solubility in magmas is also sensitive to pressure. In the pressure range from 15 to 30 kb, which is approximately equivalent to depths of 45 to 100 km in the Earth's mantle, the solubility increases with decreasing pressure at a rate of about 50 ppm/kb. On the other hand, at pressures lower than 15 kb, i.e. in the Earth's crust, the sulfur solubility decreases with decreasing pressure, at a rate of about 70 ppm/kb. Because of these solubility relations, magmas ascending in the crust are likely to reach sulfide saturation and experience some sulfur loss due to precipitation of sulfides and their removal by settling to deeper levels of the magmatic system.

Solubility of sulfur in melt with 5.72% FeO coexisting with immiscible sulfide melt (1 to 5% pyrrhotite) at 10 kbar at 1200 to 1400

^{o}C ranges from 285 ppm S at 1200 to 753 ppm S at 1400 ^{o}C for 1 % pyrrhotite runs. The general equation for sulfur solubility at 10 kbar and 1 % pyrrhotite is:

$$S (ppm) = 1.34(^{o}C) - 1209 \qquad (1)$$

In magmas characterized by high oxygen fugacity (log fO_2 > -10), the stable sulfur phase in the melt is the sulfate mineral anhydrite ($CaSO_4$), as shown in Figure 2. Although anhydrite is not a common mineral in magmas, it has been observed e.g. in the erupted products of El Chichon in Mexico, Nevado del Ruiz in Colombia and Mt. Lamington, Papua New Guinea. The high sulfur content of these oxidizing and anhydrite-bearing silicic magmas is of great interest to the study of volcanic sulfur emission, and stands in contrast to the relatively low sulfur content typical of most silicic magmas. Breakdown of anhydrite to yield sulfur-bearing gases during eruption of oxidized silicic magmas may contribute significantly to the volcanic aerosol.

Magmas rising in the Earth's crust generally reach saturation in one or more volatile species before eruption, because of the decreasing solubility of H_2O and CO_2 with decreasing pressure. In basaltic magmas, carbon dioxide is the dominant volatile component, whereas in silicic magmas, such as rhyolites and dacites, water is dominant. Nucleation and growth of vapor bubbles will occur at some depth in the magma conduit and sulfur is likely to be partitioned from the silicate melt into this volatile phase, either as H_2S or SO_2, depending on fO_2. Volatile exsolution of these main volatile phases in magmas and bubble expansion play a crucial role in the loss of sulfur during volcanic eruptions, as the process increases the magma surface area and facilitates sulfur degassing.

Sulfur Output

The emission of sulfur and other volatiles to the atmosphere through volcanic activity occurs primarily in two ways. On one hand there is the steady-state or rather continuous volatile emission to the troposphere from fumarolic and low-level eruptive activity, which for sulfur is estimated 12×10^9 kg yr^{-1} globally. This volcanic sulfur emission is dominantly from weak eruptions or fumaroles, and therefore little or none of this output is injected into the stratosphere. It is therefore removed rapidly by normal precipitation processes in the troposphere, where it has no global environmental impact and is minor in comparison to the 65×10^9 kg yr^{-1} global emission rate of anthropogenic sulfur. Annual sulfur output to the troposphere from individual and frequently active volcanoes such as Kilauea and Etna is in the range 0.17 to 1.1×10^9 kg yr^{-1}. Locally, the emission of sulfurous gases combines with atmospheric moisture and produces acid rain that severely affects vegetation around the volcano, such as during emission of sulfur from the hot crater lake of Poas volcano in Costa Rica.

Of much greater significance than these low-level tropospheric emissions is

the highly episodic but nearly instantaneous emission of sulfur during large volcanic events, which generate high (20 to 50 km) eruption columns and stratospheric injection. Thus events such as the 1815 Tambora eruption and the Laki 1783 eruption injected 24 to 26×10^9 kg sulfur to the stratosphere, or factor of three to four larger than the annual steady-state tropospheric volcanic background value. Owing to the great height attained by large explosive eruption columns, their sulfur output may have a long residence time in the stratosphere and consequently significantly impact the Earth's radiation budget.

Measurements of the stratospheric volcanic aerosol mass have been made following some recent eruptions, by use of the SO_2-sensitive Total Ozone Mapping Spectrometer on the Nimbus-7 satellite due to absorbtion by volcanic gases at the wavelengths of 312.5 and 317.5 nanometers, by airborne and ground-based Lidar and by balloon-borne sensors. Thus a notable stratospheric input of a volcanic aerosol was observed following the 1985 eruption of Nevado del Ruiz volcano in Colombia, appearing first as a plume over the Pacific at 24 to 27 km elevation. The stratospheric sulfur dioxide plume was detected and tracked for about seven days, indicating mass output of 6.6×10^8 kg SO_2. This is a relatively large mass, considering the small magnitude of the eruption.

The 1982 eruption of El Chichon volcano in Mexico led to the greatest enhancement of stratospheric aerosols since the 1963 Agung eruption in Indonesia and produced 3×10^{12} kg of magma. The El Chichon plume consisted of two major layers: at about 25 km height and a lower layer which extended from the tropopause to about 21 km height. The longitudinal spread of the cloud was quite fast and circled the globe in 21 days, with a mean speed of 22 m/s, or comparable with the longitudinal propagation of the 1883 Krakatau cloud at about 30 m/s. The latitudinal or poleward velocity of the El Chichon plume was between 0.13 and 0.19 m/s. Ballon-borne measurements showed that the El Chichon aerosol was dominated by sulfuric acid droplets, but the stratospheric plume also contained solid silicate particles between 2 and 40 microns in size. Over 80% of the solid particles were angular glass shards and the remainder were crystals of plagioclase, pyroxene, anhydrite, amphibole and magnetite. The silicate particles are generally coated by spherical droplets of sulfate gel that are less than 1 micron in diameter, and even smaller droplets of relatively unstable sulfuric acid. Fallout of silicate glass from the El Chichon eruption affected all of the northern hemisphere and glass shards from the eruption a few micron in diameter are even found in snow that accumulated in central Greenland in 1982 and early 1983. Tracking of the volcanic cloud by satellite shows that a total of 3.3×10^9 kg SO_2 was injected to the atmosphere, corresponding to about 6.7×10^9 kg sulfuric acid aerosol. In contrast, airborne lidar surveys showed an aerosol mass of about 12×10^9 kg remaining in October 1982, and total aerosol mass from the event is estimated as 20×10^9 kg. The El Chichon stratospheric volcanic aerosol led to warming of the stratosphere by up

to $6^{\circ}C$ in the months following the eruption, due to absorption of solar radiation by the aerosol. As this represented loss of heat, that would normally be transmitted to the Earth's surface, it was predicted on theoretical grounds that the eruption would lead to surface cooling. However, the effect was too small to cause significant climate perturbation.

Eruptions of sufficient magnitude to lead to climate modification occur at intervals of several decades or even hundreds of years. Because of their low frequency, their aerosols have not yet been observed or studied directly, and the study of the aerosol output of such large-magnitude eruptions must therefore rely on a study of their deposits. The yield of volcanic volatiles to the atmosphere from past volcanic eruptions can be evaluated from acidity layers in ice cores, and from petrologic studies of the tephra deposit. In the latter method, pre-eruption volatile concentration of the magma can be determined by electron microprobe analysis of glass inclusions in phenocrysts from the tephra, and the post-eruption concentration is determined from the degassed matrix glasses. These studies show, that typically 0.5 to 0.9 of the total sulfur mass fraction is degassed during eruption (Figure 4), whereas the fraction of chlorine and fluorine degassed from the magma is considerably smaller, or typically 0.2 to 0.5 of that dissolved in the melt prior to eruption (Figures 5 and 6). The yield of a given volcanic volatile species, i.e. the mass fraction released to the atmosphere, varies strongly with the composition of the magma, as shown in plot of sulfur yield versus SiO_2 content of the magma in figure 7. In the case of sulfur, where the data is most abundant, the results demonstrate clearly that the yield is highly dependent on the magma composition, and that eruptions of basaltic and trachy-andesite magmas can potentially have greatest impact on the atmosphere.

The petrologic estimates of volatile yield are clearly minimum estimates, as they only include the degassing of erupted magma. Studies of some small, recent eruptions show that in addition, a significant portion of volatile output to the atmosphere may occur from non-erupted magma at depth in the volcanic system. In the small eruptions of Fuego in 1974, El Chichon in 1982 and Nevado del Ruiz in 1985, degassing of the erupted magma accounts for only 10 per cent of the total sulfur emission. In case of El Chichon, the additional sulfur yield, over and above the amount from degassing of erupted magma, is probably related to sulfur released by break-down of erupted anhydrite phenocrysts. In case of the Ruiz eruption, the excess sulfur could be attributed to degassing of non-erupted magma at depth.

During the 1985 Ruiz eruption a total of 7×10^{10} kg magma was erupted. In addition to the observed atmospheric sulfur output of 6.6×10^8 kg SO_2, about 3900 ppm or 1.4×10^8 kg of sulfur had been adsorbed onto tephra particles and deposited with fallout from the plinian eruption. Thus the total yield from the eruption is estimated as 4.7×10^8 kg sulfur, corresponding to 0.67 wt.% of the erupted magma.

The concentration of sulfur dissolved in Ruiz magma shows a range from

about 100 to 700 ppm (Figure 3). Even the highest pre-eruption dissolved sulfur concentration in the magma is an order of magnitude lower than the 0.67 wt.% sulfur concentration observed in the total erupted products, and the source of the excess sulfur emission from Ruiz thus presents an intriguing enigma. The mass of sulfur degassed from the erupted magma is about 650 ppm, which represents an atmospheric yield of about 4.6×10^7 kg sulfur, or an order of magnitude lower than the total erupted observed sulfur mass. Thus a major part of the sulfur emission during the Ruiz eruption is from a source other than degassing of the erupted magma, either the degassing of non-erupted magma in the Ruiz magma reservoir, from break-down of anhydrite crystals present in the magma, or the excess sulfur may have been present as a free volatile phase in the magma prior to eruption. The free sulfur volatile phase would have represented up to 4 to 7 vol.% of the magma volume in the reservoir system.

It is unclear how such degassing of non-erupted magmas may affect sulfur yield during very large eruptions, such as those that have climatological impact. Judging from the good correspondence between petrologic estimates of volcanic aerosol yield and ice core aerosol mass estimates for the same eruptions, it seems unlikely that the volatile yield of very large eruptions contains a significant contribution from non-erupted magmas.

Oxidation Reactions and Aerosol Formation

The dominant sulfur gases emitted to the atmosphere during volcanic eruptions are H_2S and SO_2. The relative abundance of these sulfur species in magmas depends on oxygen fugacity and sulfur fugacity, but the H_2S/SO_2 ratio is typically quite low. A number of reactions in the atmosphere lead to oxidation of the sulfur dioxide, both in the gas phase, in the aqueous phase and by catalysis on aerosol surfaces. It is generally recognized that a reaction with the hydroxyl radical (OH^-) is most important in oxidation of sulfur dioxide in the gas phase:

$$OH + SO_2 + M \longrightarrow HOSO_2 + M$$
$$HOSO_2 + O_2 \longrightarrow HO_2 SO_3$$
$$SO_3 + H_2O \longrightarrow H_2SO_4$$
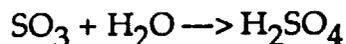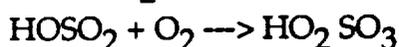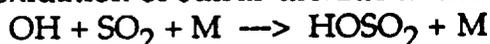
In the aqueous phase, SO_2 oxidation to H_2SO_4 occurs by reactions with O_3:

$$HSO_3^- + OH + O_3 \longrightarrow SO_4^- + H_2O + O_2$$
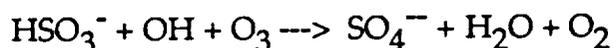
and by reactions with H_2O_2:

$$H_2O_2 + HSO_3 \longrightarrow A^- + H_2O \longrightarrow A^- + H^+ \longrightarrow H_2SO_4$$
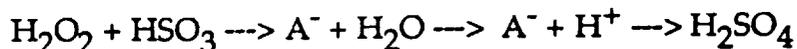
These reactions in the aqueous phase occur at a much higher rate than the gas phase oxidation, but they may be limited by the relatively low atmospheric abundances of ozone and hydrogen peroxide. However, because of the low H_2O content of air, homogeneous gas phase reaction is likely to be the main process of SO_2 oxidation in the stratosphere, where most of the volcanic SO_2 of

climatological significance resides following major volcanic eruptions. Recent discovery of nearly complete peroxide depletion in association with volcanic sulfate aerosol deposition events in Greenland ice cores following the Laki (1783) and Tambora (1815) eruptions has dramatically shown the importance of the peroxide reaction in sulfate formation (Figures 8 and 9). Finally, SO_2 conversion to sulfuric acid is aided by adsorption of SO_2 on aerosol surfaces and catalysis by metallic species such as Mn and V which are present in the volcanic dust.

Climate Impact

The petrologic estimate of sulfur yield to the atmosphere during a volcanic eruption correlates well with the observed decrease in northern hemisphere surface temperature, as shown in Figure 10. The observed temperature decrease (ΔT) is related to the volcanic sulfur output by a power function, with the sulfur mass raised to a power close to a cube root law:

$$\Delta T = 5.9 \times 10^{-5} \, S^{0.31} \tag{2}$$

where S is the volcanic sulfur mass yield to the atmosphere, in grams. It has been argued that a linear change in the mean surface temperature of the Earth occurs in response to increase of the optical depth perturbation due to volcanic sulfuric acid aerosols. With constant relative humidity profile and stratospheric temperature, the expected surface temperature change (ΔT) versus optical depth perturbation (Δt) due to sulfuric acid aerosol loading is according to the equation:

$$\Delta T \, ^{\circ}K = 0.35 - 6.8 \, (\Delta t) \tag{3}$$

where Δt is the enhancement of the stratospheric aerosol optical depth at a reference wavelength of 0.55 μm. These results indicate a linear variation of optical thickness with the cube root of the change in mass of the sulfuric acid aerosol.

The relationship between volcanic sulfuric acid aerosol mass and surface temperature response (Figure 10) can be combined with the trends of typical sulfur yield from volcanic eruptions (Figure 7) in order to extrapolate to the sulfur loading of the atmosphere from truly climate-modifying events. The derived equation (2) implies that emission of 10^{12} kg sulfur would cause a major climatic event, with surface temperature decrease of the order $-4^{\circ}C$. On the basis of the sulfur yield as a function of magma composition (Figure 7), this would require an eruption of 10^{15} kg or approx. 400 km^3 of basic magma (typical sulfur yield of 1000 ppm), or about 2×10^{15} kg or 800 km^3 of trachytic (Tambora-type) magma, and greater than 10^{16} kg or >4000 km^3 of silicic magma. These results further amplify the importance of magma type when considering the potential impacts of volcanic eruptions.

As is evident from Figure 10, eruptions of sufficient magnitude to cause major climatological effects are rare. The most recent and largest such Quaternary explosive event was the silicic Toba eruption in Indonesia about 75,000 years ago, with   about   2800

km^3 dense-rock equivalent (7×10^{15} kg) and estimated sulfur dioxide emission of 6.5×10^{12} kg. A surface temperature decrease of about $4^{\circ}C$ would be predicted from such an event on the basis of equation (2), and the eruption has been correlated with abrupt lowering of sea water temperatures in the Indian Ocean at the stage 4-5 isotopic boundary.

The March 1963 Agung explosive eruption on the island of Bali in Indonesia represents a very instructive recent case, when a major decrease in mean solar transmittance to the Earth of approximately 1.5 % occurred as a result of the eruption, representing a change which is order of magnitude larger than observed short-term variations in the so-called solar constant. Greatly elevated sulfate concentration levels were measured in the stratosphere following the eruption, reaching a peak value of 40 µg/SCM about one year after the event, but the significant time delay is in part due to the slow rate of conversion of sulfur gases to sulfuric acid aerosols.

Agung produced about 2.5×10^{12} kg of magma during the 1963 eruption. Degassing of volatiles from this magma, on basis of petrologic evidence, yielded a minimum of 5×10^9 kg total acids to the atmosphere: 2.8×10^9 kg H_2SO_4, 1.5×10^9 kg HCl, and 0.8×10^9 kg HF. An estimate of total volcanic acids deposition on basis of Greenland ice core evidence is 2×10^{10} kg, or factor of four higher than the petrologic estimate. An optical depth perturbation of the atmosphere of 0.3 occurred after the Agung eruption.

A stratospheric warming of the order $6^{\circ}C$ was observed for three years following the eruption, mainly due to the absorption of infrared thermal radiation by the volcanic stratospheric aerosol and in part due to direct absorption of solar radiation. A global surface cooling was also observed following the Agung eruption, which may in part be attributed to scattering of sunlight back to space by the volcanic aerosol. Thus a mean annual northern hemisphere surface temperature decrease of $0.3^{\circ}C$ is reported after the eruption.

The 29 March and 4 April 1982 explosive eruptions of El Chichon volcano in southern Mexico was followed by stratospheric warming of the equatorial lower stratosphere, of about $6^{\circ}C$ at 30 mbar (24 km) and about $4^{\circ}C$ at 50 mbar (20 km). The global stratospheric mass of volcanic aerosol from the eruptions was concentrated between 21 and 25 km altitude, and has been estimated in the range 1.2 to 2×10^{10} kg, consisting 99% of sulfuric acid. The eruptions were relatively minor, producing a total of only 1.09 km^3 magma (2.7×10^{12} kg), from eruption columns with column heights ranging from 20 to 24 km. Thus the volcanic aerosol mass represents approximately one per cent of the total erupted magma, which is far in excess of volatile yield that can be accounted for by the degassing of the erupted magma. The source of the excess sulfur from the eruption remains a mystery, but a likely culprit may be breakdown of the anhydrite phenocrysts present in the erupted magma.

Because of the large aerosol mass and observed stratospheric warming after the El Chichon eruption, climate models predicted temperature reduction of 0.5°C at the surface. Any temperature signal associated with the eruption was, however, swamped by the effects of an unusually strong El Nino, resulting both in extreme sea surface warming and tropospheric warming.

The great 1912 Katmai eruption in Alaska was the most voluminous eruption of this century, producing about 15 km^3 magma, with eruption column heights of 20 to 30 km. Petrologic estimates of volcanic volatile yield to the atmosphere are 7.9×10^9 kg H_2SO_4, 3.2×10^9 kg HCl, and 6.6×10^7 kg HF. The petrologic estimate of total volcanic acids is thus 1.1×10^{10} kg, whereas other volcanic aerosol estimates range from 1.34×10^{10} to 3×10^{10} kg, on basis of atmospheric studies and ice cores, respectively. From June to August 1912, up to 20 % change in direct radiation has been reported over a large area in both Europe and America, after the Katmai eruption, fading to background level in mid-1913. The northern hemisphere surface temperature decrease associated with the eruption is estimated as 0.2°C.

The largest explosive volcanic eruption in historical time is the April 1815 explosive eruption of Tambora volcano in Indonesia, which also led to the largest documented volcano-related atmospheric disturbance. The event caused the eruption of over 50 km^3 dense-rock equivalent of magma, in two plinian explosions with eruption column height of 33 and 43 km, followed by a major ignimbrite eruption. It is generally accepted that the remarkable global meteorological and optical phenomena, observed months and years after the Tambora eruption, had a strong connection with activity of the volcano. Most of these phenomena can be attributed to the effect of the stratospheric volcanic aerosol. The optical depth of the Earth's atmosphere increased rapidly after the eruption and reached a maximum of 1.4 in about five months, but did not decay to background levels until three years after the event.

Various features of the Earth's climate exhibited highly abnormal behaviour following the Tambora eruption. The annual deviation of the northern hemisphere mean temperature due to the eruption was -0.7° in 1816. In the eastern United States, the great climate anomaly of the year 1816 is a unique event which also persists in 1817 (figure 11). Summer temperature was about 1.5°C below the two-hundred year average, and the June 1816 temperature about 3°C below average. The climate effect of the Tambora event cannot be considered in isolation, however, as it was coincident with a depression in solar activity between about 1790 to 1830, i.e. the Dalton Minimum in sunspot numbers and aurorae. During these decades the characteristic eleven-year cycle in solar activity persists, but the amplitude is reduced by an order of magnitude or more. Variations in sunspot frequency have been linked to changes in the solar "constant" and in turn related to climate changes. It therefore appears likely that climate was already

deteriorating by the beginning of the nineteenth century, due to reduction in solar activity. This climate trend was then greatly amplified by the impact of the Tambora volcanic aerosol, culminating in the "year without summer" in 1816. Both ΔO^{18} data on ice cores and northern hemisphere decadal temperature trends support the contention that a climate change had set in by the first decade of the nineteenth century. Thus for example evidence from Peru ice cores show that the decade 1810 to 1820 is characterized by the most negative ΔO^{18} values (coldest temperatures) of the entire recor, culminating in the southern hemisphere wet season of 1819-20. The relative contribution of solar variability to the deterioration occuring after 1815 is estimated 10 to 20%.

The Tambora magma was enriched in volatile components, with 2.4 wt % H_2O, 570 ppm sulfur, 2220 ppm chlorine and 570 ppm fluorine. The sulfuric acid aerosol mass injected to the stratosphere from Tambora ranges from 1 to $2x10^{11}$ kg on basis of petrologic data, ice cores and atmospheric phenomena. In addition to the large sulfur yield, petrologic data indicate major emission of halogens to the atmosphere from the eruption, or 10^{11} kg of chlorine, and $7x10^{10}$ kg fluorine. The fate of these volcanic halogens in the atmosphere is unclear. Fluorine and chlorine most likely form HCl and HF gas molecules upon degassing from the magma. The latter is relatively inert in the stratosphere, as HF photolysis is shielded by oxygen and HF is also relatively indifferent to OH abundance.

Chlorine was probably removed in significant amounts from the high-temperature region of the Tambora eruption column by adsorption onto tephra. Although HCl is not known to form stratospheric aerosols, chlorine may conceivably enter other aerosol droplets. Studies of ice cores cited above indicate that acidity layers from some eruptions contain significant chlorine, requiring incorporation of this species into the aerosol by some process. While HCl is relatively inert in the stratosphere, reaction with OH or by photolytic reactions leads to formation of atomic chlorine. As the Tambora eruption cloud was dominantly in the region below 30 km which is photolytically inactive, formation of Cl and ClO by the latter process would have been minor. On the other hand, water vapor was injected in large quantities, involving both magmatic and atmospheric water. Thus OH radicals were abundant in the eruption column and available for reaction with HCl to produce atomic chlorine. Reactions of atomic chlorine with ozone are catalytic, and a single chlorine atom may destroy thousands of ozone molecules before it becomes inert and enters the HCl reservoir. Independent of their role in generation of single chlorine atoms, OH radicals from the eruption cloud would also lead directly to destruction of ozone.

Flood Basalt Eruptions

The potential environmental impact of large-scale basaltic volcanism has received increased attention in connection with the debate of causes of the terminal Cretaceous extinctions. Basaltic volcanism is an attractive mechanism for inducing climate change for two reasons: basalts display the highest solubility of

sulfur of all magmas and therefore have the highest potential sulfur yield to the atmosphere per unit mass of erupted magma. Secondly, basaltic magma is the dominant magma type extruded on Earth, with a combined annual mass eruption rate of 20×10^{12} kg yr^{-1} for hotspots and ocean ridge basaltic volcanism, compared to 5×10^{12} kg yr^{-1} for the dominantly andesitic volcanism of volcanic arcs. As mid-ocean ridges are almost entirely submerged, however, the sulfur associated with submarine eruptions is largely trapped in quenched glassy pillow lavas or dissolved in the oceans. On the other hand, the dominantly subaerial hotspots exhibit a total magma discharge rate comparable to that of volcanic arcs, and their sulfur yield is directly to the atmosphere.

Although basaltic volcanism is clearly a major potential source of sulfur injection to the atmosphere, there are other factors, however, which cast some doubt on the ability of this type of volcanism to form stratospheric sulfate aerosol layers that lead to climate change. Basaltic explosive eruptions are extremely rare in the geologic record, but very large basaltic fissure eruptions have been considered as a potential climate modifiers. Numerical simulation of a very large basaltic fissure eruption has indicated that little or no stratospheric injection would result from such an event. On basis of these results, it is unlikely that flood-basalt volcanism can have major climate effects, except perhaps in the case of high-latitude basaltic eruptions, where the tropopause is at lower altitude, such as in the case of the 1783 Laki eruption in Iceland.

This basaltic fissure eruption is the only one that has received detailed petrologic study for the determination of volatile loss to the atmosphere. The pre-eruption CO_2 and H_2O content dissolved in the Lakagigar magma has been determined by infra-red spectrophotometry analysis of glass inclusions trapped in phenocrysts. The CO_2 content of 430 to 510 ppm is consistent with a reservoir at a depth of about 4 km. On basis of Quadropol mass spectrometry, the bulk CO_2 content of glass inclusion-bearing crystals from the Lakagigar eruption may be as high as 0.85 wt.%, indicating a free CO_2-dominated volatile phase in the magma. The H_2O content of glass inclusions is about 0.47 wt.%, indicating that water exsolution would not have occurred in the magma reservoir at the depth indicated by the CO_2 solubility estimates. The volatile gradient in the reservoir and the observed correlation between vesicularity and chemical composition of the magma (Figure 12A) is therefore attributed to a gradient in the abundance of a CO_2-dominated volatile phase, ranging from vesiculated quartz tholeiite magma (Mg# 38 to 40) in the uppermost part of the reservoir, to volatile-poor tholeiite (Mg# 44 to 47) at deeper levels. We envisage CO_2 nucleation at depths corresponding to lower parts of the reservoir, and bubble rise by buoyancy through the reservoir, leading to a vesiculation gradient and volatile accumulation in the roof zone.

The yield of sulfur to the atmosphere from the Lakagigar eruption is one of the largest volcanic pollution events in historical time, and comparable to the

great eruption of Tambora. Studies of Greenland ice core acidity layers from the eruption indicate a total H_2SO_4 volcanic aerosol mass of 2.8×10^{11} kg, whereas estimate of sulfur degassing, based on the difference of sulfur concentration between glass inclusions and matrix glasses, is equivaent to 1.35×10^{11} kg H_2SO_4 aerosol. The close correspondance of these independent estimates indicates that degassing of sulfur dissolved in the magma was the dominant mechanism, and that other sources of the sulfur aerosol need not be invoked, such as an immiscible sulfide phase.

As shown in Figures 13 and 14, sulfur degassing during the eruption was strongly dependent upon both the vesicularity of the magma and its chemical composition. Thus degassing was most efficient from the evolved quartz tholeiite, which was also most vesicular and presumably highest in volatile content. On the other hand, sulfur loss was relatively minor from the less evolved and poorly vesiculated matrix glasses. These results demonstrate the critical role of the abundance of a major exsolving volatile phase in the degassing of sulfur during eruption.

The Halogens

Diffusion of anthropogenic chlorofluoromethanes (CFC) from the troposphere is currently the principal source of stratospheric chlorine, but volcanic emissions are also a potential source of stratospheric chlorine during major eruptions. The large-scale introduction of odd-chlorine species into the stratosphere during eruptions is important because of the potential of chlorine in catalyzing the removal of O_3 and thus damaging the Earth's ozone layer, which shields the biosphere from the effects of damaging solar ultraviolet radiation, such as effects on DNA and the immune system response, skin cancer and sunburn.

The stratospheric injection rate for volcanic chlorine is normally quite low, or about 2×10^7 kg yr^{-1}. During major eruptions, however, such as the 1815 Tambora event, the HCl emission is of the order 10^{11} kg. By comparison, the annual release of chlorofluorocarbons is about 7×10^8 kg yr^{-1} and the budget of stratospheric chlorine is about 10^9 kg yr^{-1}. HCl is generally the principal chlorine molecule in volcanic gases, but studies of the Mount St. Helens 1980 stratospheric cloud show that concentrations of methyl chloride (CH_3Cl) were as high or higher than concentrations of HCl. As HCl is highly soluble in water, it is possible that large quantities of the emitted HCl are dissolved in eruption cloud water and returned to the surface of the Earth as precipitation during or shortly after eruption.

A very large volcanic chlorine emission to the atmosphere, such as during the Tambora eruption, is of potential concern for the stability of the Earth's stratospheric ozone layer. Stratospheric HCl injection was documented during the small 1982 El Chichon eruption and some workers have previously alerted to the possible consequences of large volcanic chlorine emissions. HCl in the

stratosphere may react with OH radicals from magmatic water, thus forming odd-chlorine atoms which can lead to catalytic decomposition of ozone. Similarly, dissociation of magmatic water in the eruption column can form OH^- radicals, which alone can destroy ozone.

Although large quantities of chlorine and fluorine are shown to be emitted by Tambora, it should not be assumed that these gases form aerosols in the stratosphere, as physical and chemical data indicate that HCl and HF gases are unlikely to form liquid aerosols under normal stratospheric conditions. The halogen aerosols may conceivably form at higher temperatures in the eruption column, and the presence of elevated concentrations of HCl and HF in volcanic acidity layers in Greenland ice cores suggests that halogens have indeed become incorporated into some volcanic aerosols. Thus the acidity layer from the 934 AD Eldgja eruption in a Greenland ice core contains 65% HCl and high levels of F. Similarly elevated Cl levels have been noted in the Greenland ice core acidity layer from the Laki 1783 eruption. In general, the ice core data thus suggest that Cl and possibly F may enter the volcanic aerosol. This may not imply the formation of a discrete halogen aerosol, but rather that HCl and HF may be absorbed and dissolved in the sulfuric acid aerosol.

HCl is inert toward ozone, but reaction of HCl with OH leads to formation of atomic chlorine, followed by the catalytic decomposition of the ozone by the Cl. Thus in the stratosphere Cl can be released from HCl by reactions of the type:

$$HCl + OH \rightarrow H_2O + Cl$$
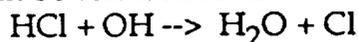
Similarly, methyl chloride can produce atomic chlorine by photolytic decomposition and attack by OH. Several reactions involving gaseous chlorine have the effect of converting odd-oxygen molecules (including ozone) to diatomic oxygen by ClO catalysis. They are reactions of the type:

$$O_3 + Cl \rightarrow O_2 + ClO$$
$$O + ClO \rightarrow O_2 + Cl$$
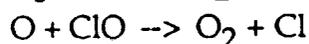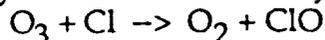
A Krakatau-size emission, involving 3×10^8 kg Cl_x would result in about 7% depletion of the ozone layer. Chlorine output was two orders of magnitude higher than this value during the 1815 Tambora eruption, and major ozone depletion by that event cannot be ruled out. Given the great importance of the ozone layer to the biosphere and climate, the modeling of the potential impact on atmospheric chemistry by a Tambora-size eruption is timely.

During the 1815 Tambora eruption, about 7.4×10^{10} kg HF gas emission occurred to the atmosphere. In general, HF is assumed to be inert in the stratosphere. The photolysis of HF is shielded by oxygen, and the reaction of HF with OH is endothermic, so that it is beleved that F atoms do not play the same role in stratospheric chemistry as chlorine atoms. Furthermore, fluorine and to some extent chlorine, are known to adsorb onto tephra particles and thus may be relatively rapidly removed from the atmosphere in the tephra fallout.

Large fraction of the fluorine is stripped from the high-temperature region

of the eruption column (338 to 700°C) by adsorption onto tephra and thus incorporated in the fallout deposit near source. Majority of the Tambora fluorine emission may have been removed by this process, and led to fluorosis and thus accounting for the observed death of livestock. During the 1970 Hekla eruption in Iceland, fluorine-rich fallout led to poisoning of large numbers of livestock up to 200 km from the volcano. The tephra fall from the eruption was unusually rich in adsorbed fluorine (2000 ppm). The concentration of the adsorbed fluorine in the fallout deposit was directly dependent on surface area of the tephra grains, and thus the concentration increased with decreasing grain size. The total mass of fluorine deposited is estimated as 3×10^7 kg or corresponding to 700 ppm of the total erupted mass from Hekla.

CO$_2$ Output from Volcanoes

In view of the major role that CO_2 plays in the Earth - atmosphere system, it is appropriate to evaluate the contribution of volcanism in general, and that of very large Tambora-size eruptions of CO_2 to the atmosphere. As shown below, however, the output of this gas from even the largest eruptions is trivial in the context of the huge atmospheric CO_2 reservoir. The degassing of CO_2 from the Earth's mantle occurs dominantly during submarine volcanism at the mid-ocean ridges, and consequently most of the gas is dissolved in the oceans and does not enter directly the Earth's atmosphere. Estimates of total mantle CO_2 degassing via mid-ocean ridge volcanism range from 0.3 to 2.4×10^{11} kg yr^{-1}. The output of carbon dioxide from subaerial events such as the Tambora eruption could be determined by infra-red spectroscopy measurement of carbon in glass inclusions trapped in crystals in the tephra, but such analyses have yet to be made. Estimates of CO_2 output can be made, however, on the basis of known solubility of carbon dioxide in magmas. CO_2 solubility of Tambora magma is likely to be of the order of 500 ppm judging from experimental evidence and almost all of the carbon dioxide is likely to have exsolved or degassed, by analogy with the gas budget of the Hawaiian volcano Kilauea. With total magma output of 2.4×10^{14} kg, the CO_2 yield from the Tambora eruption would be 1.2×10^{11} kg, or approximately equal to the annual mantle output. Much more meaningful estimates of CO_2 degassing can be made for the Laki basaltic fissure eruption in Iceland in 1783, based on infra-red spectroscopy measurements of CO_2 content of glass inclusions in crystals and bulk CO_2 content of crystals and trapped glass. These studies show, that while the magma contained 400 to 500 ppm dissolved CO_2 prior to eruption, the magma also contained a free CO_2-dominated vapor phase, with a bulk CO_2 concentration of 0.85 wt.%, representing a total release of about 3×10^{10} kg of the gas during this large

eruption. The high yield from the Laki eruption is surprising, and if the results may be applied to eruptions of other hotspots, they indicate that weight fraction estimates of 0.005 CO_2 for the Deccan basalts may be quite reasonable. It has been proposed, that massive CO_2 degassing during extrusion of the Deccan Traps at time of the Cretaceous-Tertiary transition destabilized atmospheric and marine mixed-layer pCO_2, leading to terminal extinctions. Results of numerical models of Deccan Traps degassing indicate that while a significant "greenhouse" effect is likely, with a global warming less than 2 °C the process was on too small a scale to have produced massive extinctions.

The anthropogenic production rate of CO_2 from the burning of fossil fuels is estimated as 4×10^{14} mole yr^{-1}, or 1.8×10^{13} kg yr^{-1}. Thus the Laki and Tambora volcanic output was only approximately one percent of the current anthropogenic CO_2 output. Consequently these eruptions would have represented a very minor addition to the total CO_2 atmospheric reservoir, which is estimated as 5.5×10^{20} moles in 1985, or 2.4×10^{19} kg. Thus the CO_2 output from a Tambora-size eruption would represent only an addition at the 5 ppb level to the huge atmospheric reservoir of this gas.

Volcanic Water

Observations and theory shows that the solid particle weight fraction in high eruption columns $(1-n_c)$ is only of the order 0.018; the remainder being almost entirely entrained atmospheric air and expanding volcanic gases. Assuming that most of the tephra which generated the fallout deposit (5.8×10^{13} kg) had entered the lower stratosphere, the mass of associated air lofted to the stratosphere would then be about equal, and equivalent to approximately 7×10^{13} m^3 at the surface. The water content of saturated air at 1 atm and 14°C is about 0.01 kg H_2O/kg air. Thus total mass of atmospherically derived water entrained into the stratospheric eruption column could have been as high as 5×10^{11} kg. A portion would condense with rise in the eruption column and cause precipitation, but some portion would enter the stratosphere. Although large, this figure is only one third of the mass of magmatic water introduced into the atmosphere (1.7×10^{12} kg), as discussed in a previous section. Normally the content of water vapor decreases with height due to lowering of both temperature and saturation vapor pressure and condensation. However, water vapor is likely to be introduced to high levels under the conditions of elevated temperatures and turbulence within a buoyantly rising eruption column.

Water vapor introduced to the stratosphere by an eruption column could be a major source of OH radicals by reaction of water vapor with photodissociated oxygen atoms. Evidence from ground-based spectroscopic measurements of OH

during the 1982 El Chichon eruption indicates that water vapor was injected at the level of 20 ppm, or two to four times normal and may have been responsible for the large ozone depletion observed in 1982-1983. Elevated levels of volcanically derived OH from Tambora may have played a major role in generation of H_2SO_4 by reaction with SO_2, in the regeneration of free Cl atoms from HCl and in direct reactions with stratospheric ozone.

Conclusions

The principal effects of volcanism on the Earth's climate are related to the effects of volcanic aerosols on the radiation budget of the Earth, both in terms of incoming sunlight and outgoing terrestrial radiation. Although silicate particles (volcanic ash) form a major part of the erupted component from volcanoes, they are probably not important component of the volcanic aerosol due to the process of particle aggregation and thus a short residence time of volcanic ash in the atmosphere. On the other hand, sulfuric acid aerosols formed by gas-to-particle conversion from SO_2 and H_2S volcanic gases are potentially most important in climate modification due to a long stratospheric residence time of this aerosol. Examples of petrolgic estimates of sulfuric acid aerosol mass for several eruptions are shown in Figure 15.

The potential climate impact of volcanism is thus largely dependent on the mass yield of sulfur gases from an eruption which is capable of penetrating the tropopause and forming a stratospheric aerosol. Petrologic studies indicate that eruptions of low-silica magmas, such as basalts and trachy-andesites, have the highest sulfur yield, whereas high-silica magmas, such as rhyolites and dacites, transport relatively minor quantities of sulfur to the atmosphere. Similar dependence of volcanic mass yield and magma composition is exhibited also for chlorine and fluorine gases. Empirical data on climate effects of several historic eruptions correlate very well with the mass yield of sulfur to the atmosphere during these volcanic events. When this relationship is applied as a basis for climate prediction following volcanic events, it is evident that the eruption of very large volumes (>1000 km^3) of silicic magmas would be required to have major climate effects (>$2^{o}C$). Only one eruption of this magnitude is known in the last one hundred thousand years: the Toba eruption in Indonesia 75,000 years ago.

The largest known sulfuric acid aerosols have been formed by trachyte or trachy-andesite explosive eruptions, such as the Tambora event in 1815, with column height up to 43 km. Both ice core evidence, atmospheric phenomena and petrologic studies indicate that the sulfur output during this event was exceptionally large, and a climatic deterioration followed the eruption, as discussed above. However, some uncertainty exists regarding the magnitude of the climate event that is purely due to the Tambora volcanic aerosol, as there is some indication that the eruption occurred during a decade of cool climate. Tambora and several other volcanoes also emit very large masses of halogen gases during eruption, but the atmospheric impact of volcanic chlorine and fluorine during

such events has not been explored. While fluorine may be largely stripped out of the eruption column by adsorption onto tephra particles, it is possible that chlorine may be injected into the stratosphere during such events, where odd-chlorine may severely deplete the Earth's ozone layer. Similarly, stratospheric depletion of ozone by volcanic hydroxyl radicals derived from magmatic water vapor must be considered during very large explosive eruptions.

Recent moderate-size volcanic eruptions such as Agung 1963 and El Chichon 1982, which produced comparabel stratospheric aerosols and both led to stratospehric warming, yet their effects on the Earth's northern hemisphere surface temperature were quite different, with 6 ^{O}C decrease in the case of Agung, and no observable effect in the case of El Chichon. These differences werve as a reminder of the great buffer of short-term climate change: the heat capacity reservoir of the ocean. The thermal inertia of the ocean-atmosphere system is dramatically underscored when one considers that the heat capacity of the atmosphere corresponds to only 3 m thick layer of sea water. Therefore, while El Chichon-size volcanic events have significant effects on the thermal radiation budget and generally heat the stratosphere, their ability to cool the troposphere and the Earth's surface is tempered by the ocean heat reservoir. Storage and re-distribution of heat from the ocean can thus dominate short-term climate.

Figure Captions

Figure 1: Typical ranges of eruption column height (A), intensity or magma discharge rate (B), and magnitude (total erupted mass) for fifty recent explosive eruptions. The equations describe the curve fit to the relations between column height, discharge rate and magnitude, with correlation of 0.88 and 0.82, respectively.

Figure 2: Relationship between sulfur solubility and oxidation state of magmas.

Figure 3: Sulfur solubility vs. FeO content of magma, from melting experiments on Mt St Helens 1980 magmas (crosses) and for glass inclusions in phenocrysts in Nevado del Ruiz 1985 magmas.

Figure 4: The relation between pre-eruption sulfur concentration in magmas (sulfur content in glass inclusions in phenocrysts) and sulfur content in degassed matrix glasses, in parts per million. The diagonal line shows no sulfur loss. For most eruptions, the fraction of sulfur which is degassed from the magma ranges from 0.5 to 0.9 of the initial concentration.

Figure 5: The realtionship between pre-eruption chlorine concentration in magmas (chlorine in glass inclusions in tephra phenocrysts), and the chlorine concentration in degassed matrix glasses. The diagonal line shows no chlorine loss. Note that the chlorine fraction degassed from the magma is typically less than 0.5 of the inital amount present in the magma.

Figure 6: Relationship between pre-eruption fluorine concentration and matrix glass fluorine content for seven eruptions. Fraction of fluorine degassed during eruption is generally less than 0.5. The diagonal line shows no fluorine loss.

Figure 7: Yield of sulfur to the atmosphere as a function of magma composition (SiO_2). Note the high sulfur yield in the case of eruptions of basaltic (low-silica) magmas, in contrast to the low yield for eruptions of rhyolitic and other high-silica magmas.

Figure 8: The relationship between hydrogen peroxide and volcanic (non-sea salt) sulfate in the Laki 1783 volcanic acidity layer in a Greenland ice core.

Figure 9: The relationship between hydrogen peroxide and volcanic (non-sea salt) sulfate in the Tambora 1815 volcanic acidity layer in a Greenland ice core.

Figure 10: Correlation between sulfur yield to atmosphere from volcanic eruptions and the observed northern hemisphere surface temperarture decrease for several volcanic events. Sulfur yield based on the petrologic estimates.

Figure 11: Observed climate response following the Tambora 1815 eruption. Upper curve is annual summer temperature data for the eastern United States, at the latitude of Philadelphia (Pa), based on several long temperature series. The solid horizontal line shows the 224-year average summer temperature. Lower curve is annual June tempeature data for New Haven (Conn). The horizontal line shows the 145-year New Haven June mean temperature.

Figure 12: Vesicularity and crystal content of Laki 1783 basaltic glasses as a function of magma composition.

Figure 13: Vesicularity and sulfur content of Laki 1783 matrix glasses, demonstrating the importance of a major carrier gas (CO_2) in sulfur degassing.

Figure 14: Relationship between sulfur and iron content in Laki 1783 glass inclusions and matrix glasses. The dotted line is Mathes' solubility curve for basaltic magmas. Note the extreme depletion of sulfur in matrix glasses as a function of degassing, whereas glass inclusions in phenocrysts contain pre-eruption sulfur concentrations.

Figure 15: Petrologic estimates of sulfuric acid aerosol mass for several historic and recent volcanic eruptions. The Minoan age eruption of Thera (Santorini) volcano has a relatively low sulfate yield, according to petrologic data, whereas the ice core acidity layer which has been tentatively correlated with the eruption is order of magnitude higher. The 1259 eruption is observed in ice cores; the source volcano of the huge sulfate input into the atmosphere is unknown, but is probably in the equatorial region.

A

y = - 60.4 + 7.2x   R = 0.88

B

y = 2.59 + 0.43x   R = 0.82
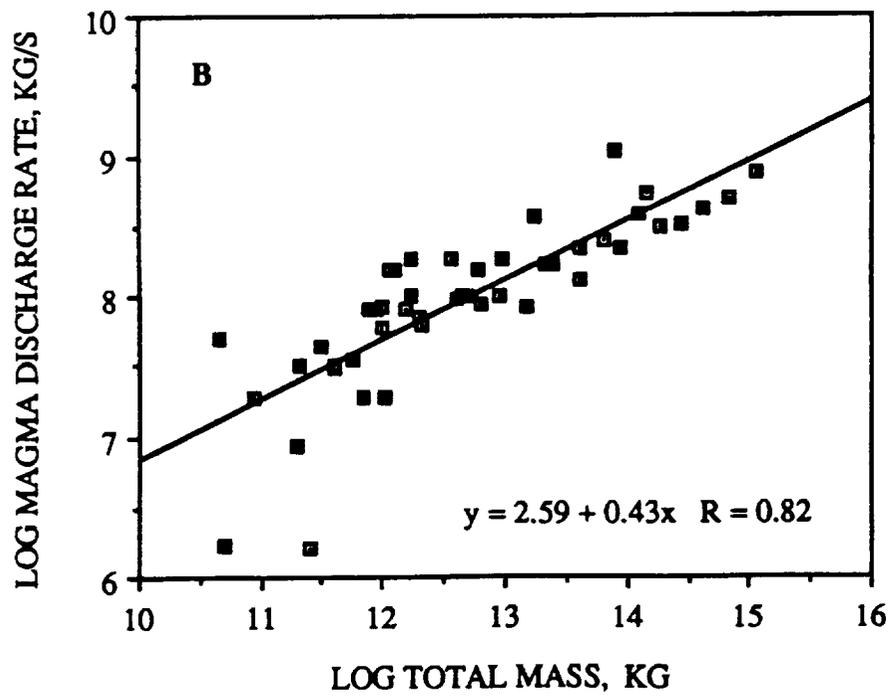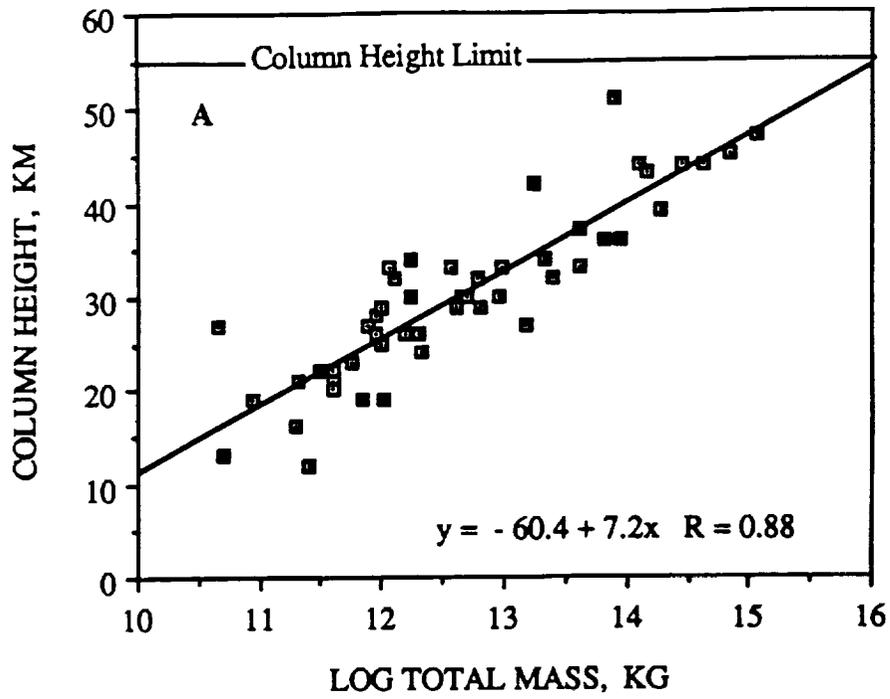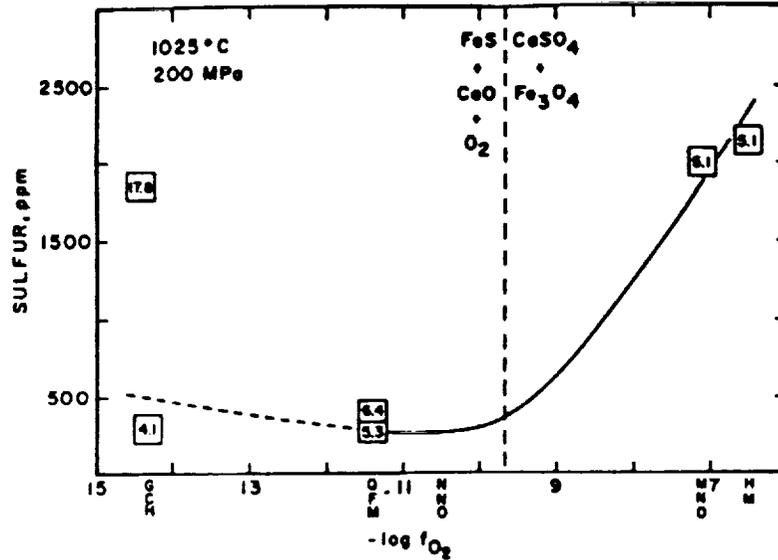
Fig. 4. Sulfur solubility as a function of oxidation state. All experiments done at 1025°C and approximately 200 MPa. Numbers in boxes are silicate melt FeO contents. The curved line represents a saturation surface for hydrous silicate melts with approximately 5 wt % FeO. The dotted vertical line schematically indicates an oxidation reaction whereby FeS breaks down to form $CaSO_4$. This reaction has not been closely bracketed, but at 1025°C it must lie between the QFM and MNO buffer curves. GCH and QFM experiments are sulfide (FeS-rich liquid) saturated. HM and MNO experiments are sulfate ($CaSO_4$) saturated. Diagram is polybaric in fS_2, with log fS_2 ranging (approximately) from -3.0 at GCH to -1.0 at HM (error ±1.0 log units).

C-24

$y=9.18+76.48x \quad R=0.95$

y = 2989 - 57x + 0.23x^2   R = 0.77

SULFUR ppm

SiO2

C-29
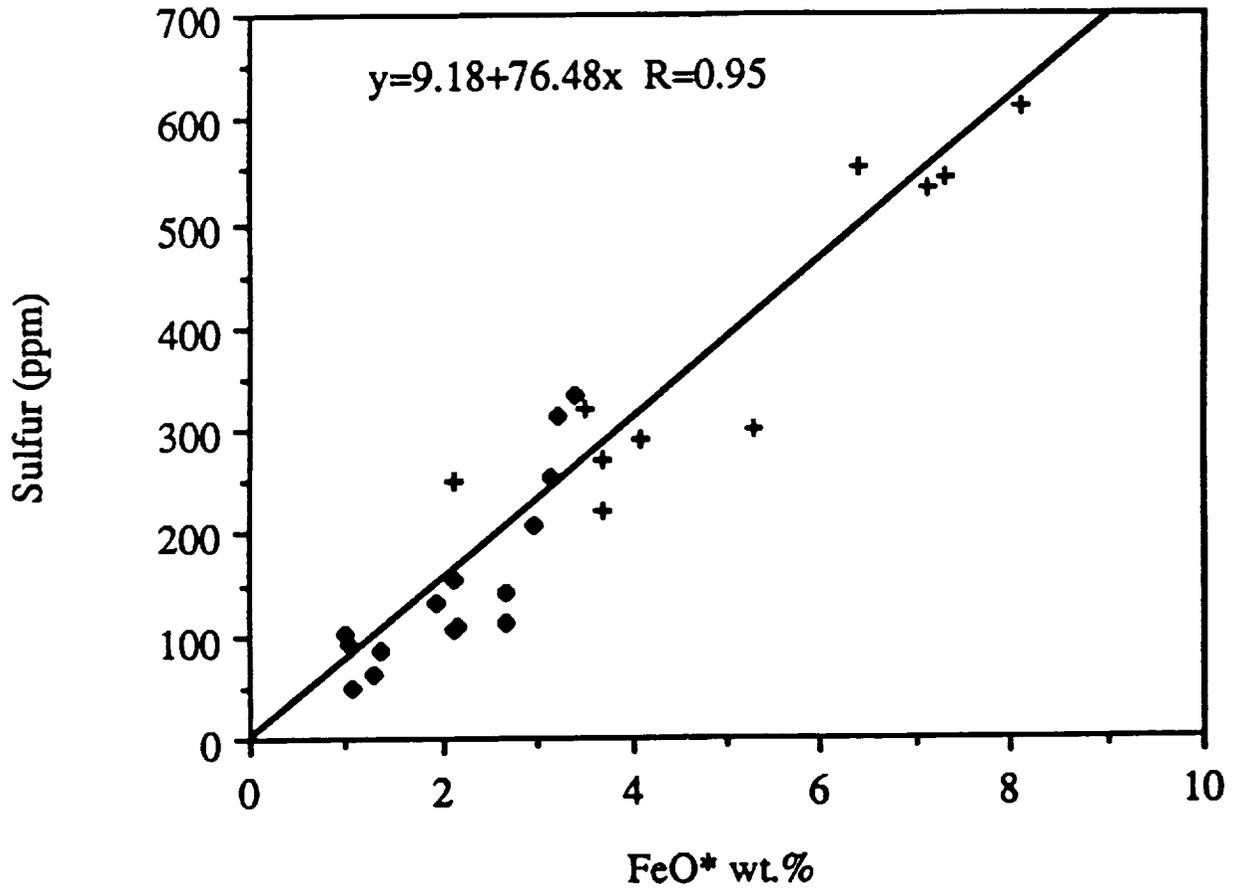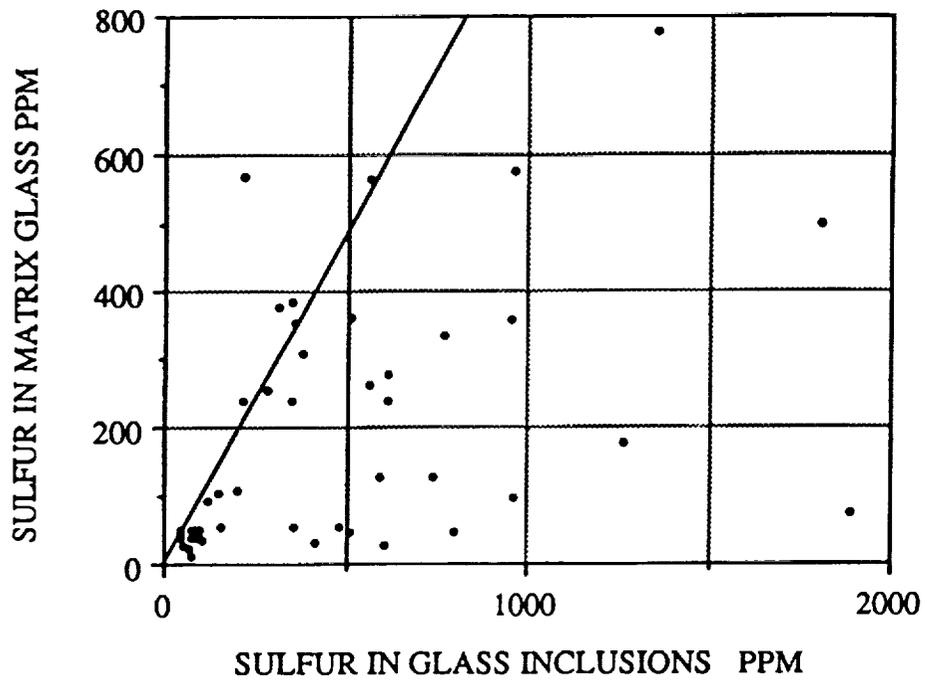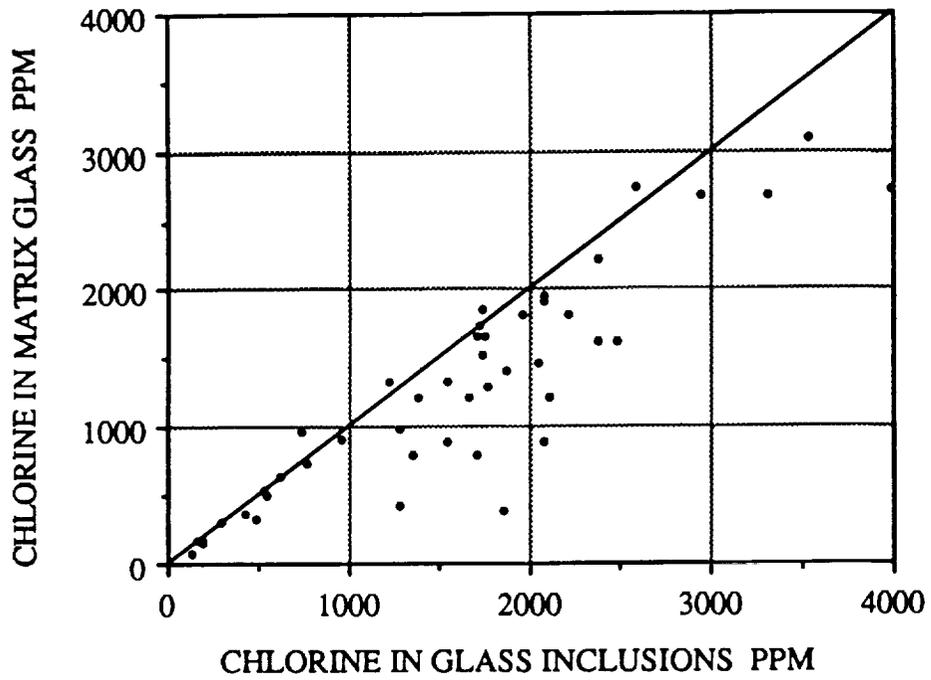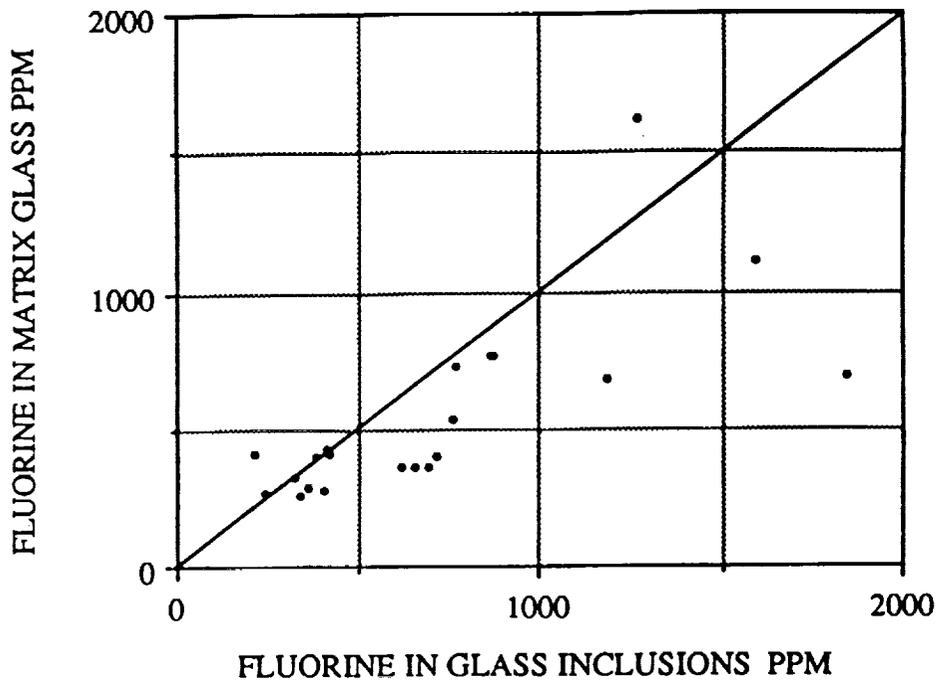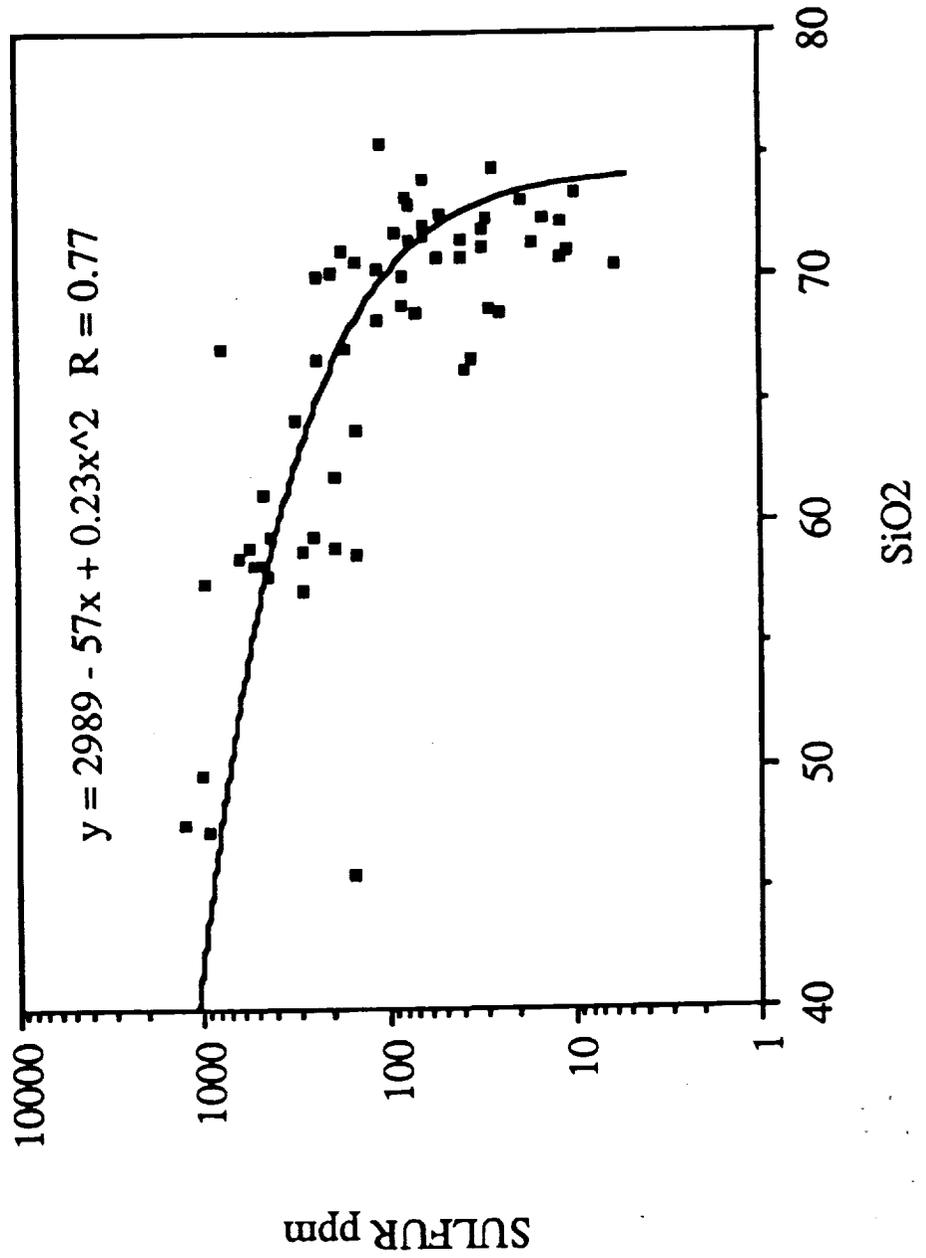
ACIDITY LAYER IN GREENLAND ICE CORE:
AEROSOL FROM THE 1783 ERUPTIONS OF LAKI
IN ICELAND AND ASAMA IN JAPAN
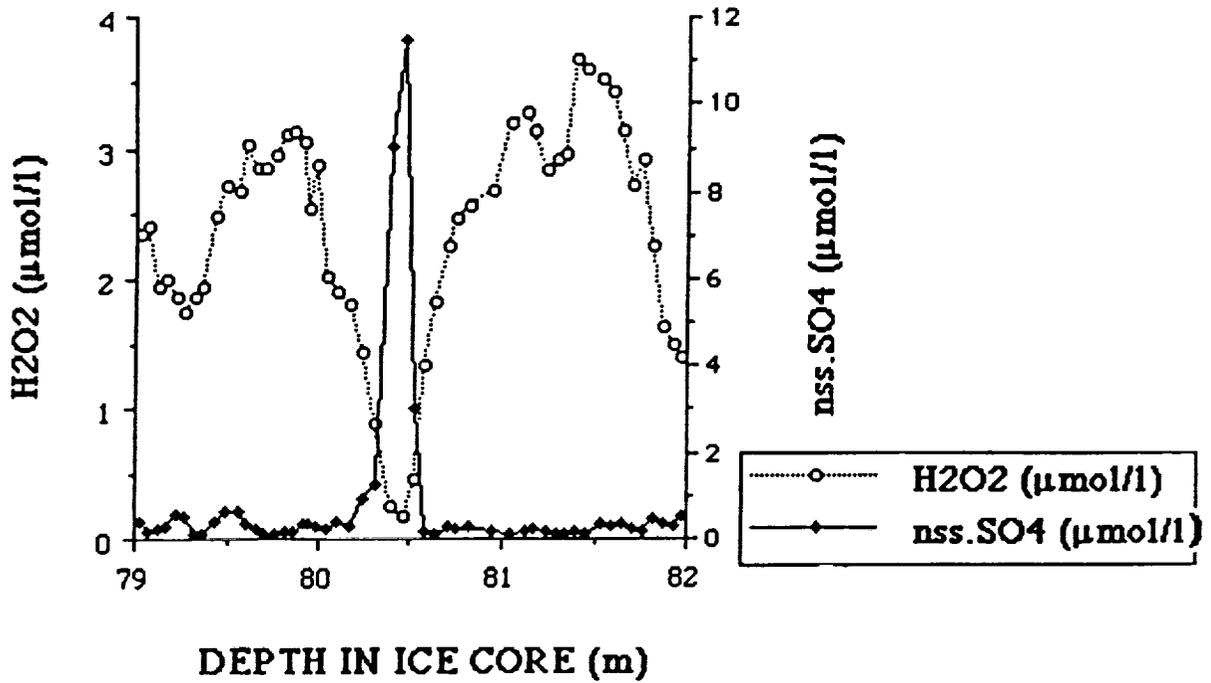
ACIDITY LAYER IN GREENLAND ICE CORE: AEROSOL DEPOSITION FROM THE 1815 ERUPTION OF TAMBORA IN INDONESIA

$y \; oC = 5.89e\text{-}5 \times SULFUR^{0.308} \quad R = 0.92$

LAKI
TAMBORA

KATMAI
KRAKATAU
ST MARIA
AGUNG
FUEGO

ST HELENS

TEMP DECREASE -oC

SULFUR YIELD (GRAMS)

C-32

FIGURE 2

FIGURE/3

SULFUR ppm

VESICLE vol %

Date AD (y-axis)
H2SO4 kg (x-axis)

Date	Volcano
1982	El Chichon
1963	Agung
1956	Bezymianni
1912	Katmai
1902	Santa Maria
1886	Tarawera
1883	Krakatau
1815	Tambora
1783	Laki
1259	?
934	Eldgja
550	Rabaul
131	Taupo
-850	Hekla-3
-1615	Thera - Minoan
-1645	Ice Core

x-axis: 10^8 10^9 10^{10} 10^{11} 10^{12}

C-37
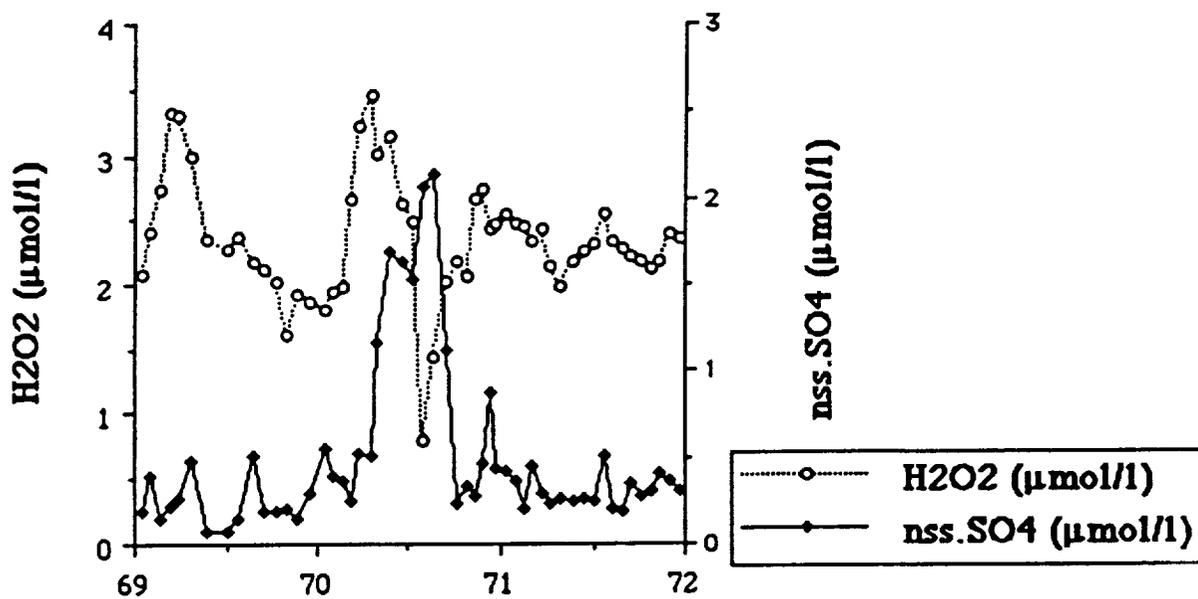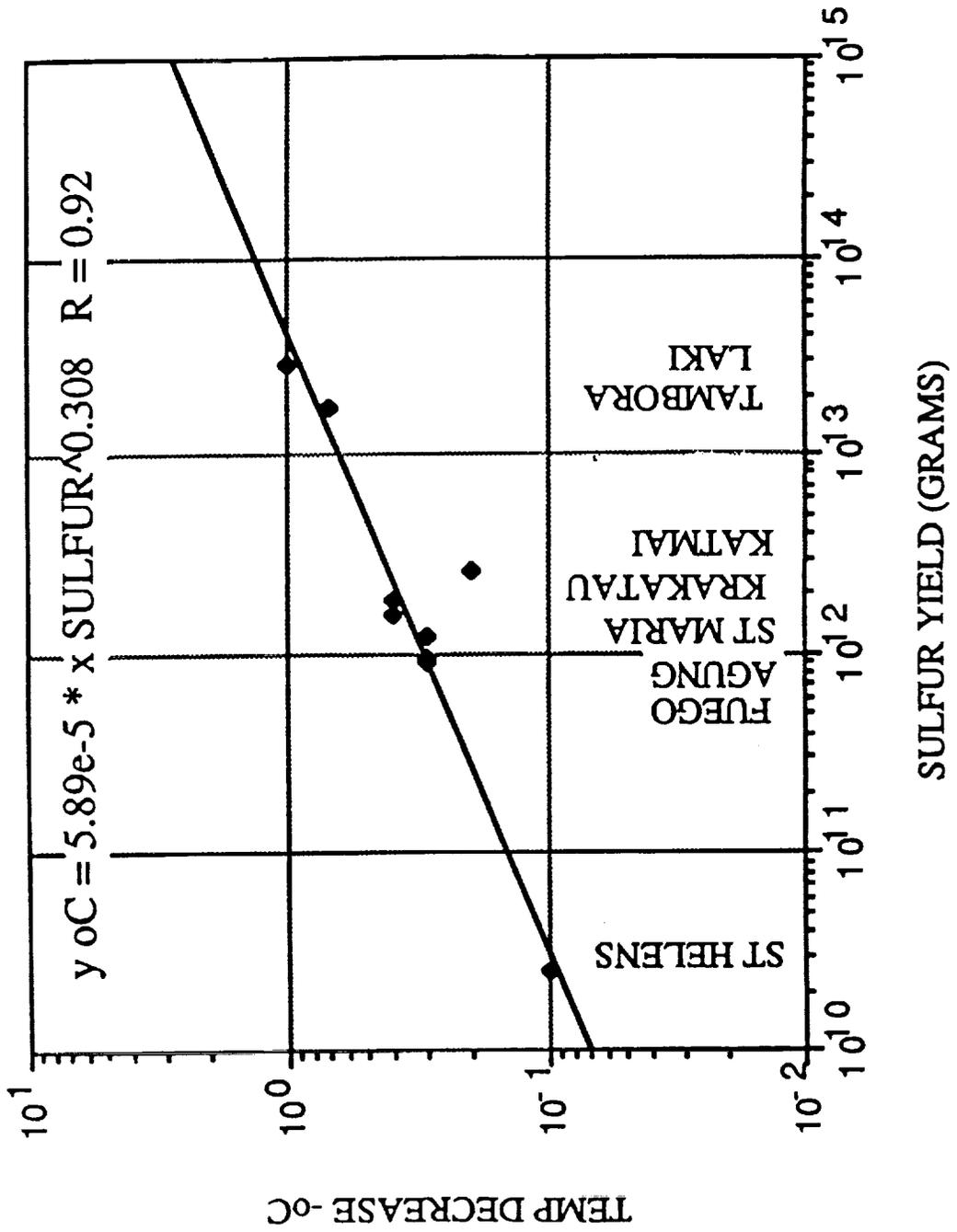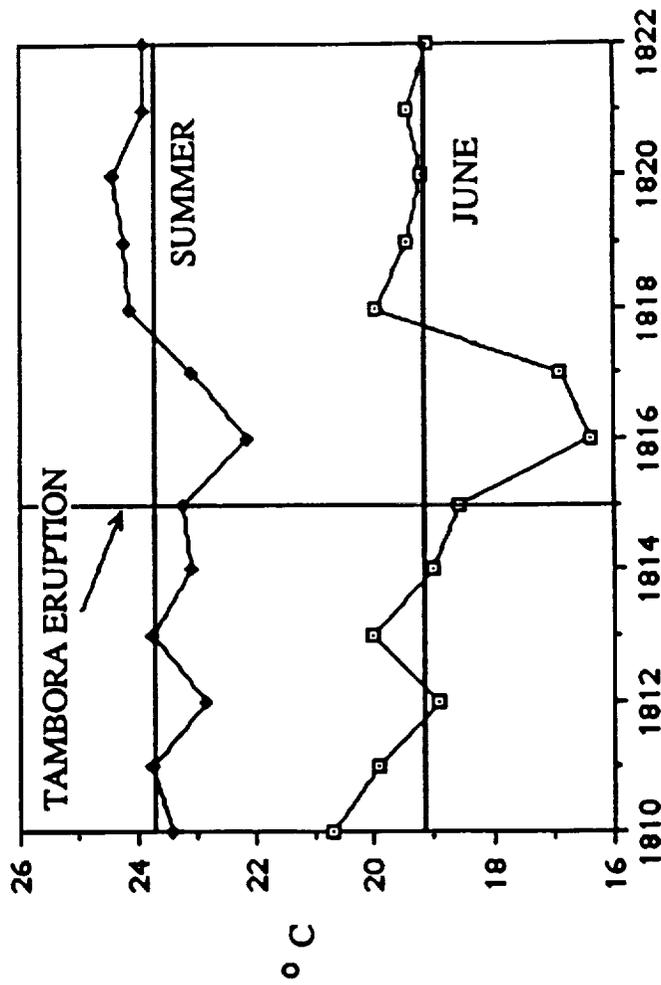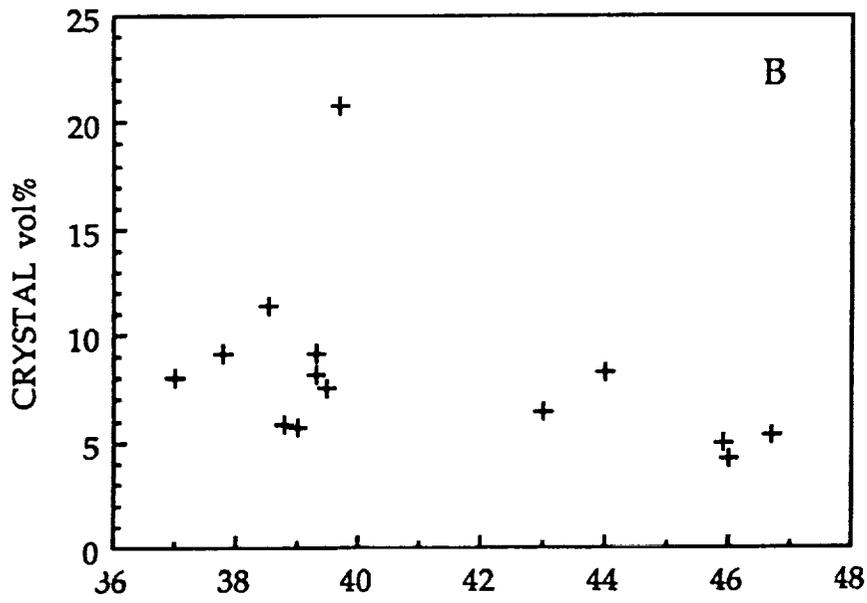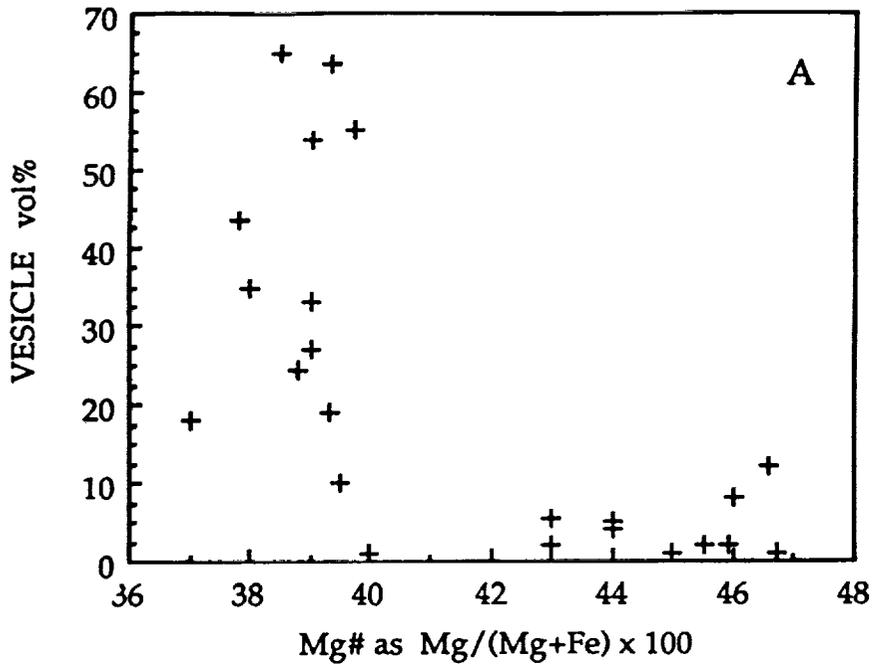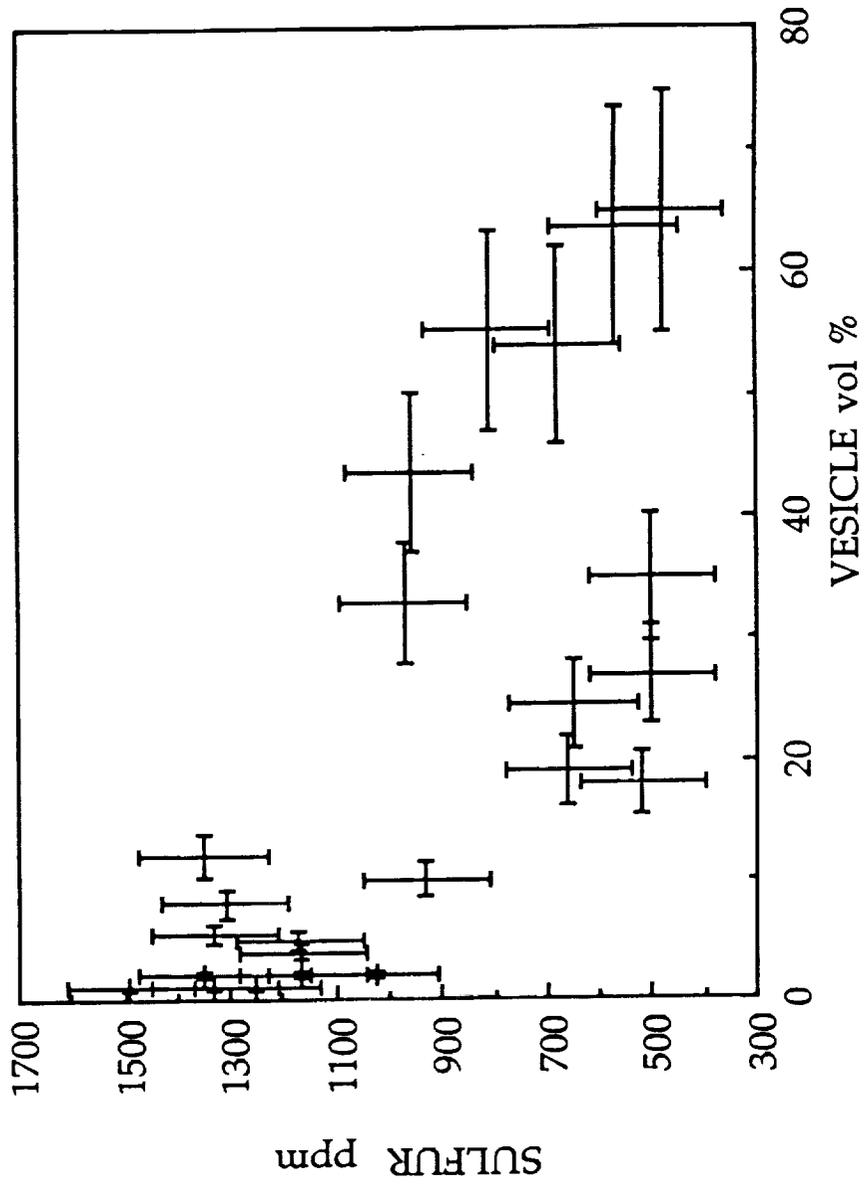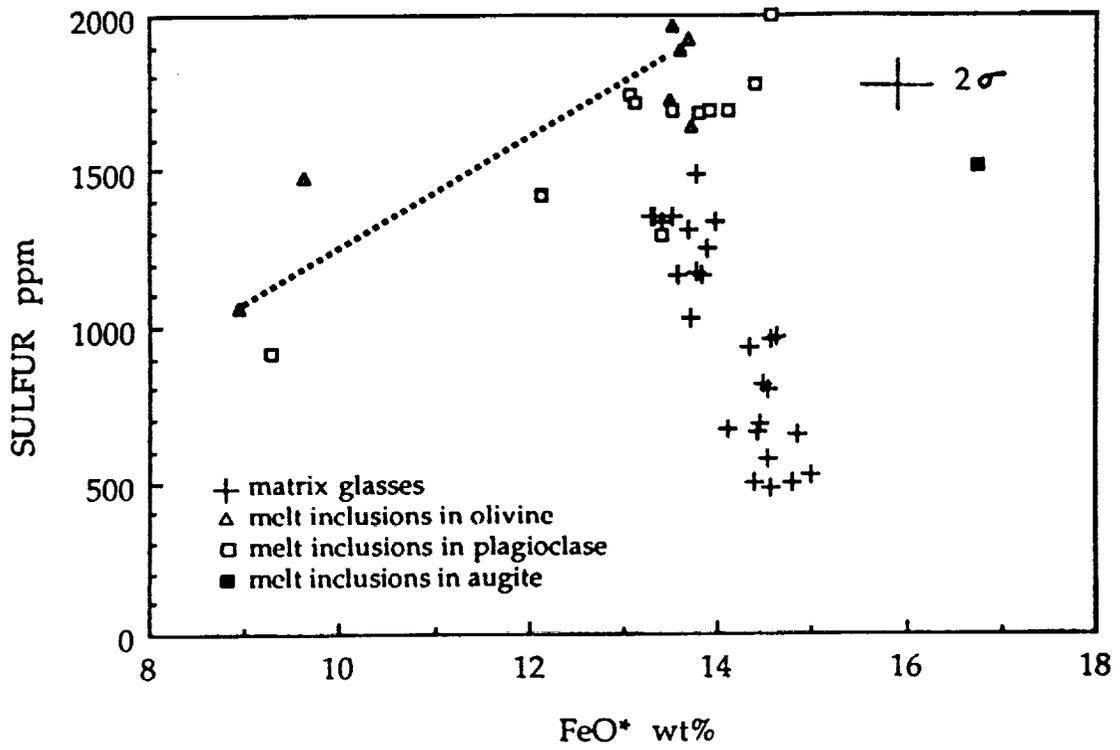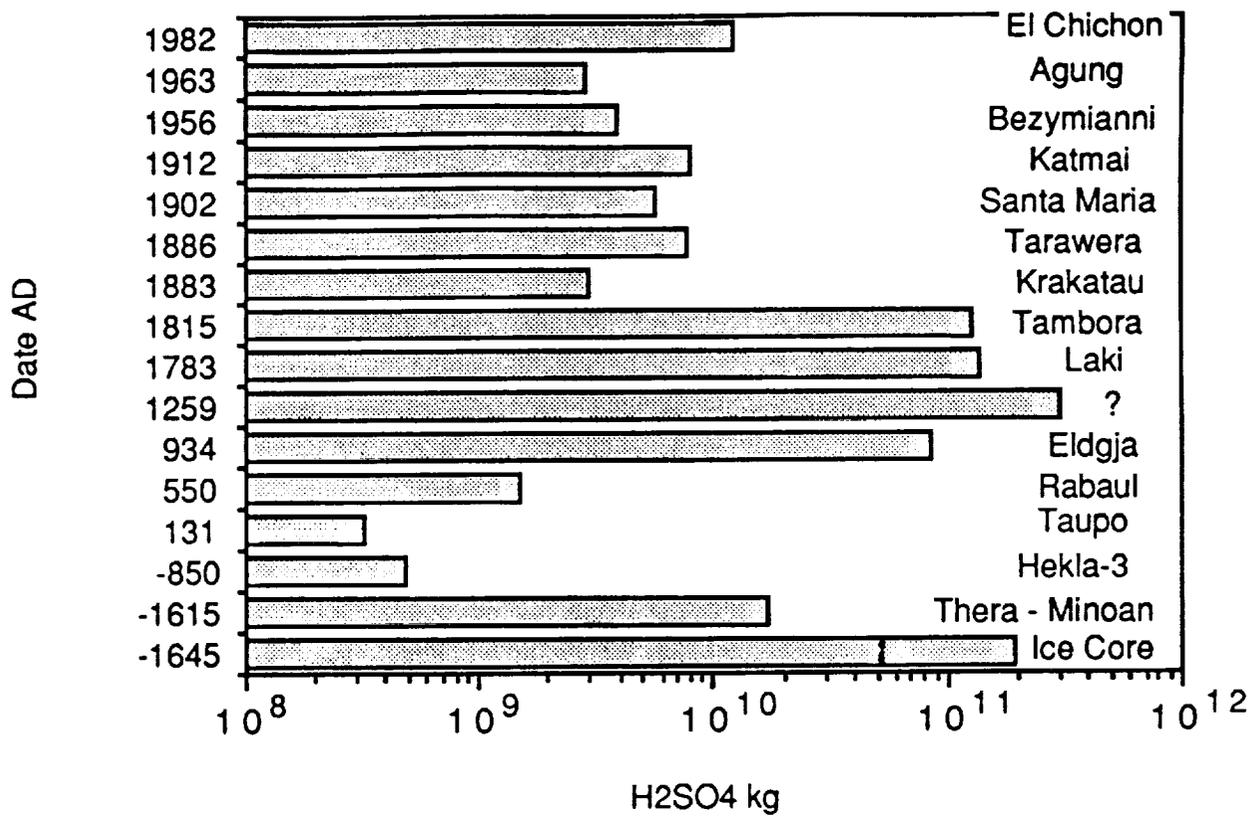
VOLCANIC AEROSOLS:
CHEMISTRY, MICROPHYSICS, EVOLUTION AND EFFECTS

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CONTENTS

Introduction

Stratospheric aerosols have been the subject of scientific speculation since the 1880's, when the powerful eruption of Krakatoa attracted worldwide attention to the upper atmosphere through spectacular optical displays. The presence of a permanent tenuous dust layer in the lower stratosphere was postulated in the 1920's following studies of the twilight glow. Junge collected the first samples of these 'dust' particles and demonstrated that they were actually composed of sulfates, most likely concentrated sulfuric acid (Junge and Manson, 1961; Junge, 1963). Subsequent research has been spurred by the realization that stratospheric particles can influence the surface climate of earth through their effects on atmospheric radiation. Such aerosols can also influence, through chemical and physical effects, the trace composition of the atmosphere, ozone concentrations, and atmospheric electrical properties.

The properties of stratospheric aerosols -- both the background particles and those enhanced by volcanic eruptions -- have been measured in situ by balloon ascents and high-altitude aircraft sorties. The aerosols have also been observed remotely from the ground and from satellites using both active (lidar) and passive (solar occultation) techniques (remote sensing instruments have been carried on aircraft and balloon platforms as well). In connection with the experimental work, models have been developed to test theories of particle formation and evolution, to guide measurement strategies, to provide a means of connecting laboratory and field data, and to apply the knowledge gained to answer practical questions about global changes in climate, depletion of the ozone layer, and related environmental problems.

Figure 1 illustrates the various kinds of aerosols that have been identified in the stratosphere. The figure provides information on the size dispersion and concentration of these diverse particulates.
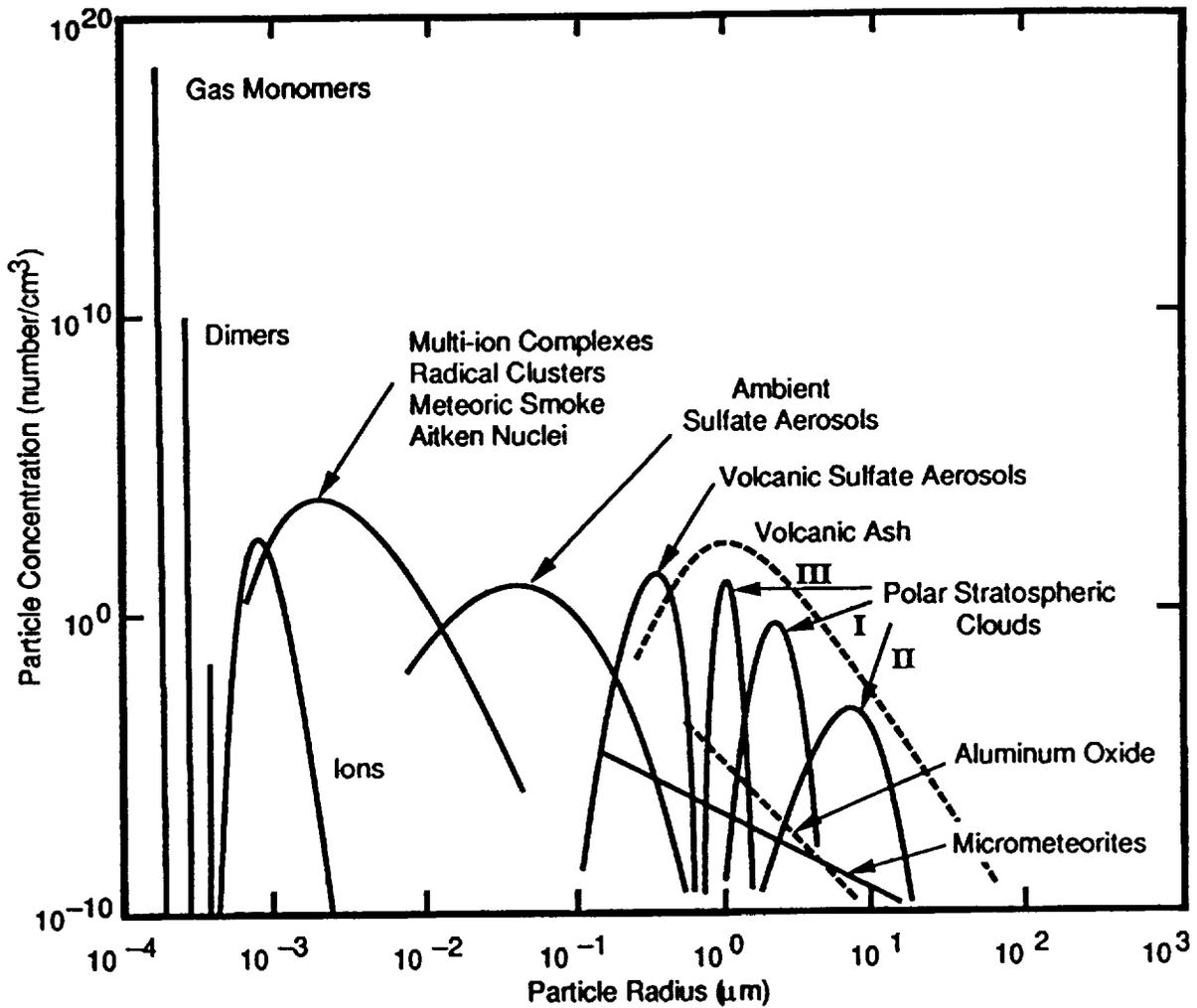
Figure 1. Spectrum of particles in the earth's stratosphere. Shown are the approximate size dispersions of particles of different origin. The total number concentration of each type of particle is roughly indicated by the peak value on the vertical scale. Some aerosols are highly variable in concentration and properties, particularly the 'volcanic ash' component of volcanic eruption clouds; typically, these particles will fall out of the stratosphere within a few months of an eruption.

C-2

Stratospheric Sulfate Aerosols and Precursor Gases

The environmental conditions that exist in the stratosphere favor the formation of sulfuric acid aerosols in the presence of sulfur-bearing precursor gases. Because such gases exist in the background atmosphere, the stratosphere always contains a tenuous, ubiquitous sulfate haze. The processes that control this haze also control the formation and evolution of volcanically-induced aerosols. Accordingly, investigations of the ambient stratospheric aerosol layer provide insights into the behavior of volcanic eruption clouds above the tropopause.

Background Aerosols

It is now fairly well established that the ambient stratospheric sulfate layer is formed as a result of the chemical transformation and condensation of sulfur-bearing gases (OCS, SO_2) transported into the stratosphere from the troposphere or injected there by large volcanic eruptions. The general properties of these aerosols are summarized in Table 1.

Table 1. Properties of Stratospheric Sulfate Aerosols

Composition -- H_2SO_4/H_2O (~70%/30%); Traces of sulfates, nitrates, nitryls, chlorides

Origin -- Tropospheric sulfides; OCS, CS_2; Volcanic sulfur emissions; SO_2 photochemical oxidation to H_2SO_4 via OH

Properties -- Liquid spheres, perhaps slurries and some solids; < 1 ppbm; ~$1/cm^3$, ~ 0.1 μm radius

Distribution -- Global; 12 to 30 km altitude; Latitudinal and seasonal variations

Effects -- Shortwave radiation scattering ($\tau < 0.01$); Longwave absorption/emission ($\tau \ll 0.01$); Heterogeneous chemical activation of chlorine (?)

Influences -- Natural and anthropogenic OCS sources; Volcanic activity; Stratospheric dynamics

Trends -- Global increase of ~ 6%/yr in mass

In addition to the chemical reactions of precursor sulfur gases -- which are illustrated in Figure 2 -- that lead to the production of condensible sulfur compounds -- primarily H_2SO_4 -- evolution of the sulfate particle layer involves a number of microphysical processes; these include aerosol nucleation, growth by condensation, evaporation of volatiles, coagulation, and gravitational sedimentation.
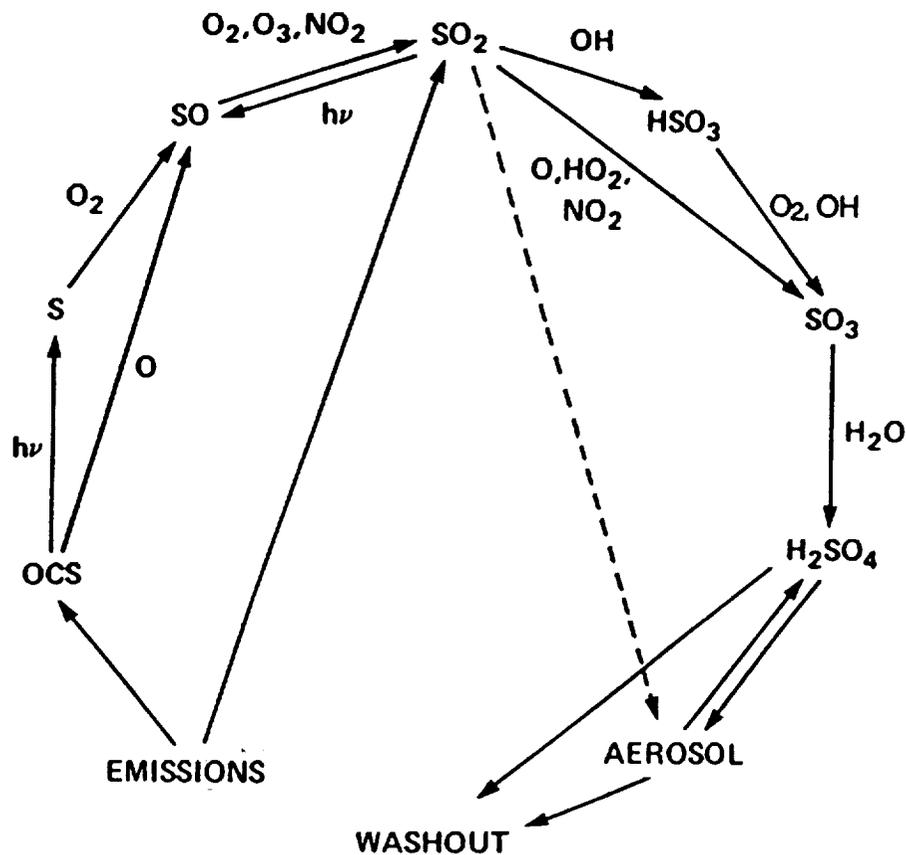
Figure 2. Gas phase sulfur reaction pathways that lead to the production of sulfuric acid vapor, which condense to form sulfate aerosols. The reactions are essentially irreversible in the lower stratosphere, and sulfur is ultimately lost through transport of stratospheric aerosols into the upper troposphere. In the upper stratosphere, enough ultraviolet radiation is present that H_2SO_4 may be photodissociated, thus recyling sulfur into simpler forms (however, the H_2SO_4 photodissociation rate has never been measured).

Some of the complex physicochemical processes responsible for the formation of the stratospheric aerosol layer are illustrated schematically in Figures 3 and 4. The potential contribution of meteoritic debris and ion clusters in sulfate particle formation are also indicated in Figure 4.
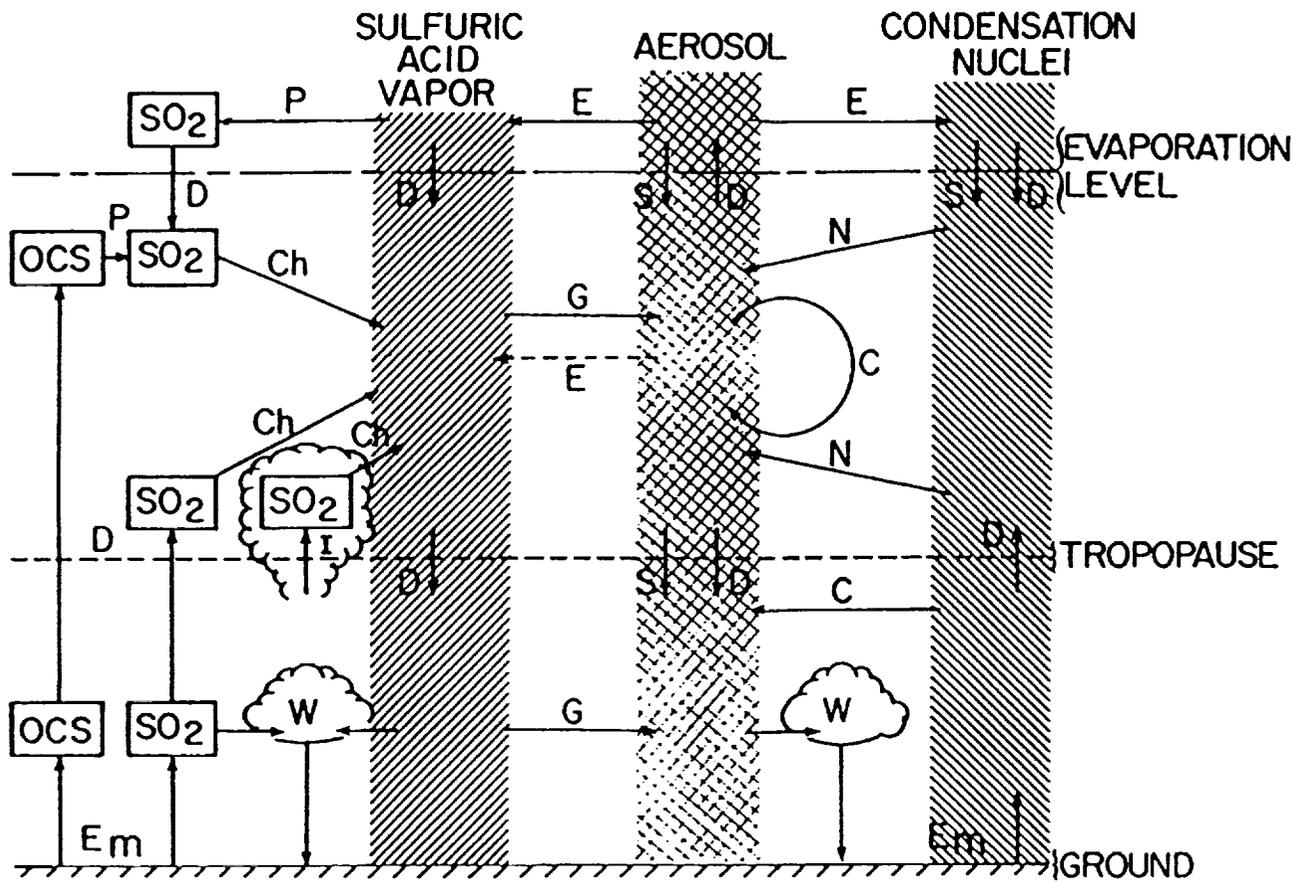
Figure 3. Schematic diagram of the microphysical and chemical processes that control the formation and evolution of stratospheric sulfate aerosols. The principal compositional elements are: precursor sulfur-bearing gases; sulfuric acid vapor; condensation (Aitken) nuclei; and sulfate aerosols. The letters represent specific processes and interactions: C = coagulation, Ch = chemical reaction, D = diffusion (turbulent), E = evaporation, Em = emission, G = growth by condensation, I = injection (e.g., by volcanic eruptions), N = nucleation, P = photolysis, S = sedimentation, and W = washout.

Figure 4. Diagram depicting the physical and chemical processes which affect stratospheric aerosols. Meteoritic debris, positive and negative ions, and tropospheric Aitken nuclei can serve as nucleation sources for sulfate aerosols and ice clouds. These particles can grow, evaporate, coagulate and fall vertically. The particles are also advected in stratospheric winds and diffuse under the influence of small scale turbulent eddies.

Volcanic Aerosols

Table 2 summarizes the general properties of the sulfate aerosols generated by a large volcanic eruption (that is, one for which the eruption column penetrates the tropopause and deposits substantial quantities of gaseous and particulate emisssions in the stratosphere).

Table 2. Properties of Volcanic Aerosols

Composition -- Silicates; H_2SO_4/H_2O (~70%/30%); Traces of sulfates, nitrates, chlorides, etc.

Origin -- Volcanic SO_2 fumes; H_2S ?, OCS, CS_2; Photochemical oxidation to H_2SO_4 via OH

Properties -- Liquid spheres; Solid mineral particles dominant the first month;
~ 100-1000 ppbm; (highly variable); ~1-10/cm^3; ~ 0.3 μm radius

Distribution -- Regional (days); Zonal (weeks); Hemispheric (months); Global (year)

Effects -- Shortwave radiation scattering ($\tau > 0.1$) leads to surface cooling;
Longwave absorption warms the stratosphere;
Injection of HCl, H_2O, etc., alter air composition;
Heterogeneous reactions on sulfate aerosol surfaces;
Ozone decrease from ΔT, ΔHCl, $\Delta Aerosol$, etc.;
Stratospheric stability / tropospheric dynamics affected;
Nuclei for upper tropospheric cirrus (?)

Influences -- Geophysical; No anthropogenic influences; Geological setting determines effects;
Impact on ozone affected by future CFC buildup

Trends -- Significant eruption every ~20 years on average; Major eruptions are ~100 years apart

---

The microphysical development and properties of volcanic eruption clouds can be illustrated through model simulations that include all of the relevant physical and chemical processes (Turco et al., 1982, 1983; also see Figures 2-4). In the following series of figures, simulations of the evolution of the El Chichon eruption cloud are used to indicate the behavior of volcanic aerosols. The primary eruption of El Chichon occurred on April 4, 1982; debris and gases were injected to altitudes of about 30 km over the Yucatan penninsula.
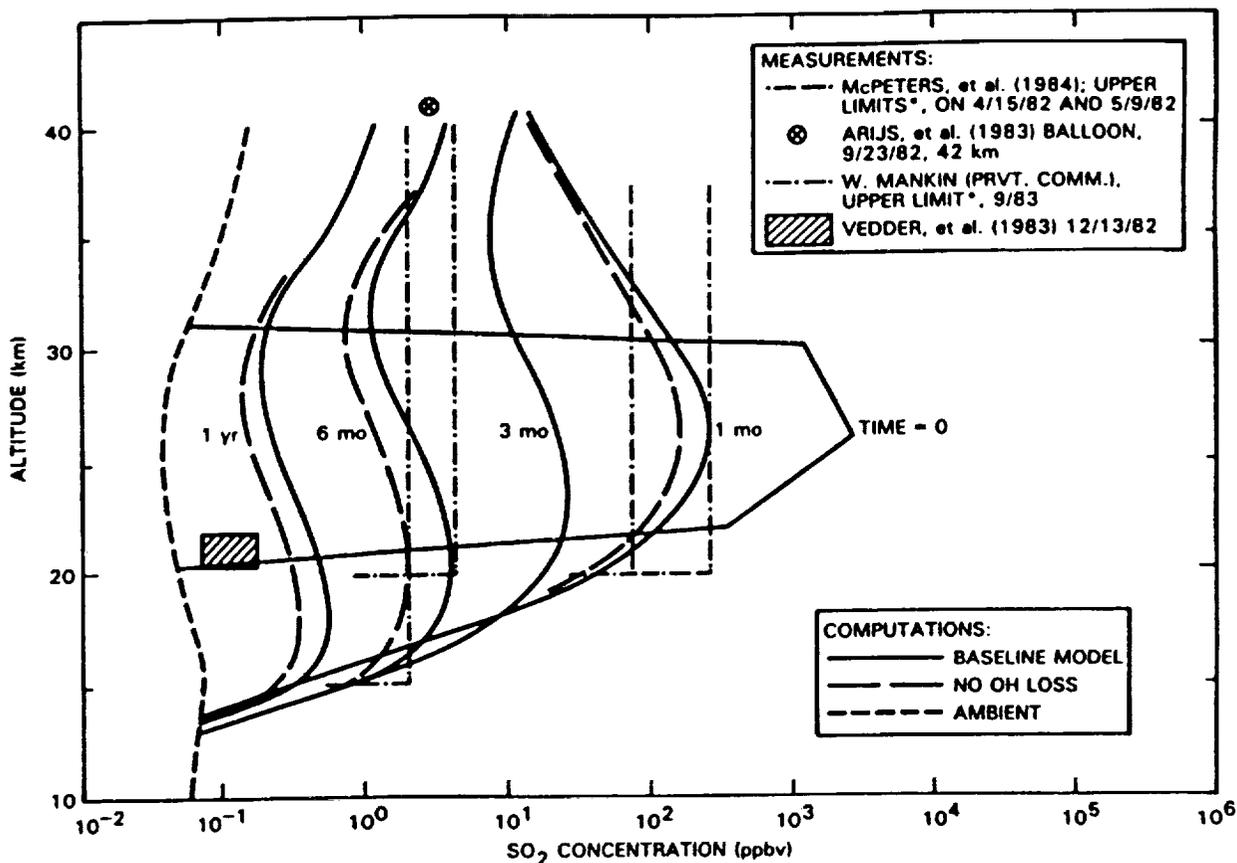
Figure 5. Simulation of the El Chichon volcanic eruption cloud SO_2 concentration using a one-dimensional aerosol/photochemistry model. The initial SO_2 injection profile (reconstructed from sparse data defining the injection heights) is shown, along with vertical profiles during the first year following the eruption. Some data on observed SO_2 concentrations are plotted.

Figure 5 illustrates the vertical diffusion of the initially-injected SO_2 over time, and the SO_2 depletion by chemical transformation into H_2SO_4. Two depletion scenarios are shown. The results corresponding to "NO OH LOSS" are probably the more accurate, although the differences between the simulations shown are relatively small. These results seem to fit the limited constraints imposed by the few direct SO_2 concentration measurements available. The "NO OH LOSS" computation includes the key sulfur reaction,

$$HSO_3 + O_2 \longrightarrow SO_3 + HO_2.$$
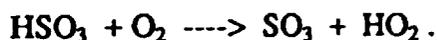
The reaction is so rapid that sulfur radicals (i.e., HSO_3) never achieve a significant concentration, and thus do not play a role in the chemical evolution of the aerosol cloud (except as an intermediary sulfur species). More importantly, this reaction does not consume odd-hydrogen, HO_x, as would the competing process,

$$HSO_3 + OH \longrightarrow SO_3 + H_2O.$$
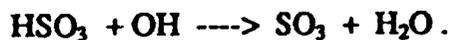
Hence, HO_x is not consumed as it proceeds to oxidize SO_2 catalytically.
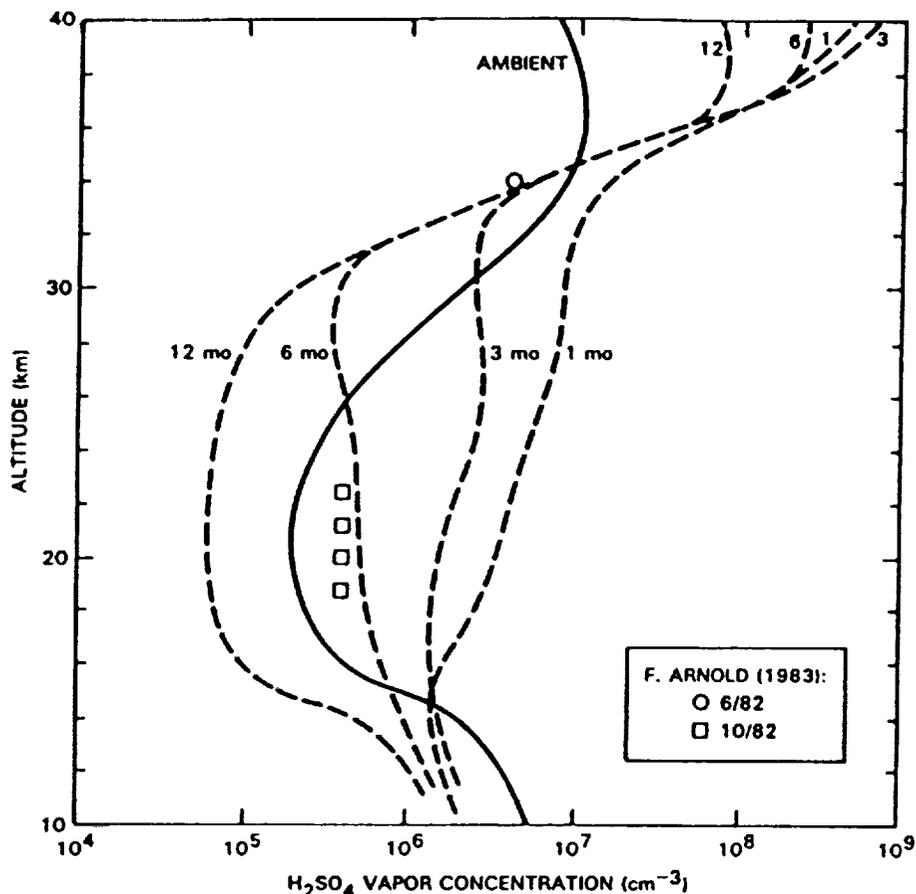
Figure 6. Sulfuric acid vapor concentrations resulting from the oxidation of SO_2 injected by El Chichon (as described in Figure 5). Some observational data are compared. The ambient H_2SO_4 profile is shown for comparison (the ambient profile comes from a model that has been validated against measurements; Turco et al., 1982).

The H_2SO_4 concentrations exhibit an interesting behavior. At early times, the concentrations increase, which would be expected as excess SO_2 injected by the volcano is oxidized. However, at late times, the H_2SO_4 concentrations fall even below the ambient concentrations. The depletion occurs because the mass (and surface area) of the sulfate aerosols has increased substantiaally during this period (and will remain elevated for at least two years). The rate of H_2SO_4 vapor deposition (and loss) is roughly proportional to the aerosol surface area at any time. The maximum increase in H_2SO_4 vapor -- a factor of about 10 -- is limited by condensation. As can be seen in Figure 7, the maximum increase in sulfate mass is much greater for this case -- about a factor of 100.

ALTITUDE (km)

40

30

AMBIENT

1 yr   6 mo 1 wk   1 mo

3 mo

20

GANDRUD, et al. (1983) ~ 30°N
  □   SUMMER, 1982
  △   FALL, 1982
OBERBECK, et al. (1983) 26 - 42°N
  ⊢—⊣ 7/23/82
  ⟷   9/23/82
  ⟩—⟨ 11/4 - 5/82
MROZ, et al. (1983) ~ 30°N
  ○   8/5, 17/82

10

10⁻²   10⁻¹   10⁰   10¹   10²   10³   10⁴   10⁵

SO_4^{2-} CONCENTRATION (ppbm)
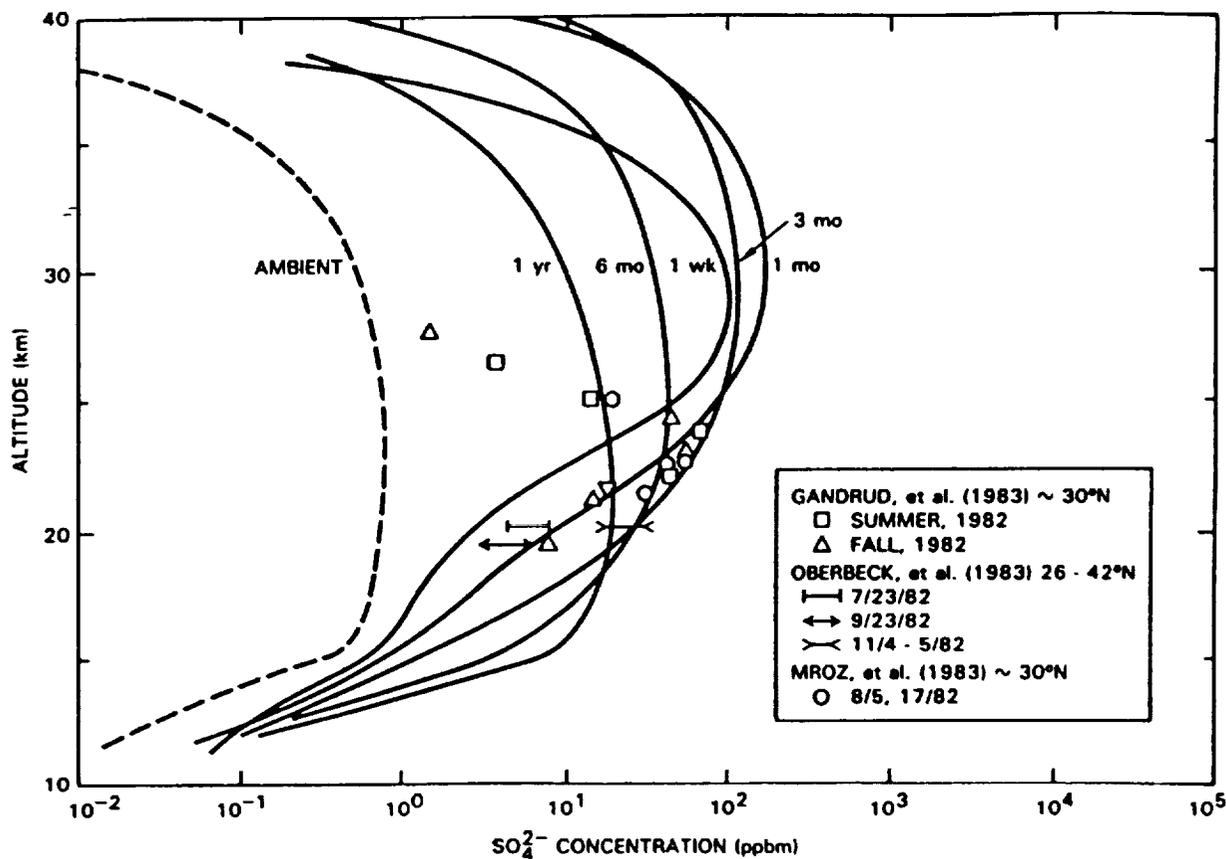
Figure 7. Sulfate mass concentrations (expressed in parts per billion by mass) are illustrated for the conditions corresponding to Figures 5 and 6. Data are compared. The ambient (computed) aerosol mass profile is also shown. The predicted mass profiles are shown at various times following the eruption.

The predicted aerosol sulfate mass profiles in Figure 7 compare favorably with observations below about 25 km, but overestimate the mass above this altitude, indicating a substantial aerosol burden up to about 40 km. It should be noted that high-altitude sampling of the volcanic clouds was very limited. Data collected by the Solar Mesosphere Explorer satellite suggest, indeed, that the aerosols of El Chichon reached altitudes of 40 km (Watson et al., 1990). However, the assumptions concerning vertical 'eddy' diffusion explicit in the one-dimensional model used in this study clearly causes the sulfur gases and aerosols to be transported too rapidly above 25 km. Lidar measurements indicated that the El Chichon particles remained below about 30 km for several months following the eruption. A point to note is that, although the aerosol mass mixing ratio is relatively large above 30 km, the absolute sulfate mass is relatively small.
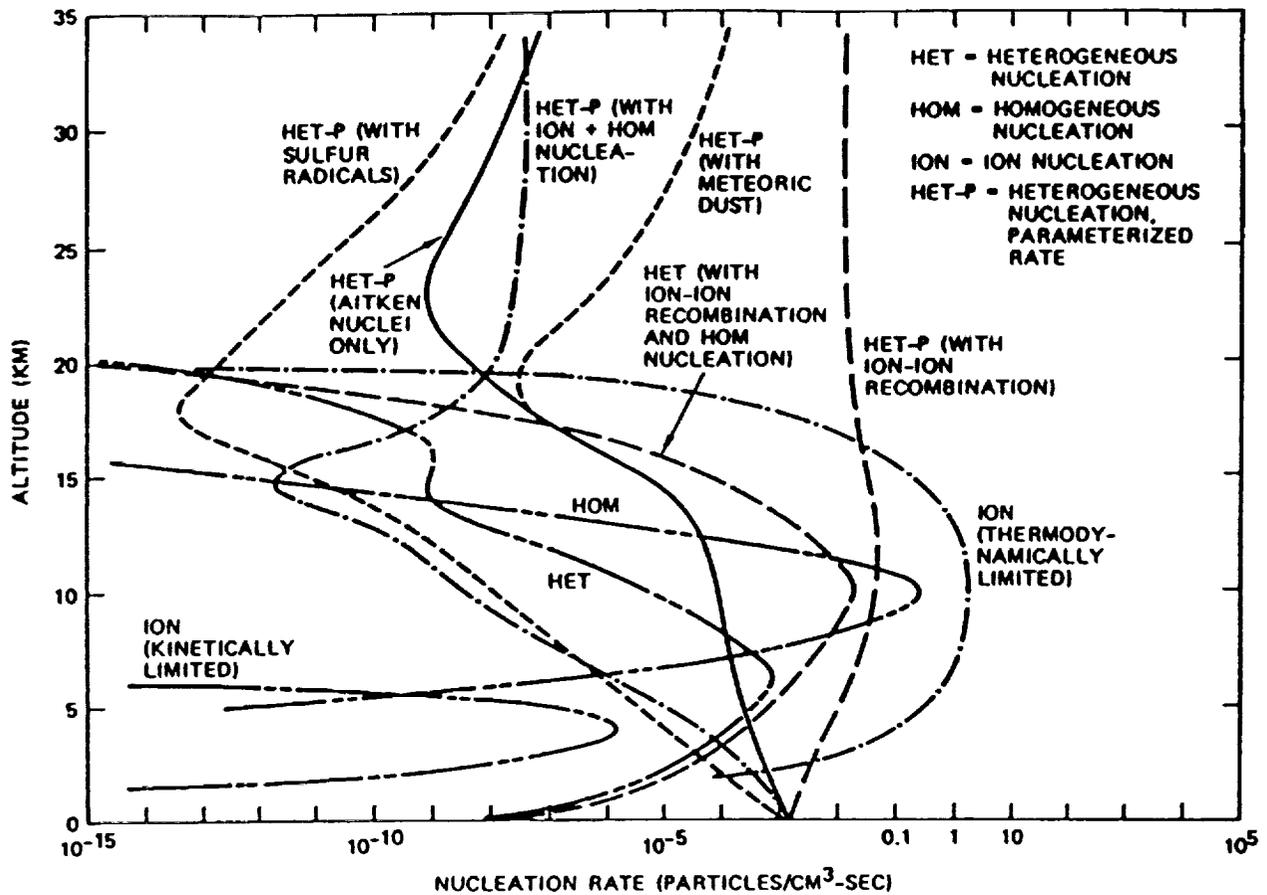
Figure 8. Sulfuric acid aerosol nucleation rates in the lower atmosphere for different nucleation mechanisms. A binary solution of H_2SO_4 and H_2O condenses; the solution composition is fixed thermodynamically by the local water vapor concentration and air temperature. The concentration of H_2SO_4 is similar to the ambient profile in Figure 6, and a standard H_2O profile is used. The most important processes are those labelled "HOM" (homogeneous nucleation of binary liquid embryos from the gas phase), "HET" (formation of embryos on pre-existing surfaces), and "ION (KINETICALLY LIMITED)" (nucleation of vapors around ion cores, stabilized by electrostatic energy).

It is imortant to note that, in the ambient stratosphere, nucleation rates that exceed about 10^{-6}/sec are significant because, over time, aerosol concentrations of $\sim 1/cm^3$ can accumulate. In the lower stratosphere, water vapor abundances are high enough and temperatures are cold enough that homogeneous nucleation can occur if the H_2SO_4 concentration is well above ambient. If the peak H_2SO_4 vapor concentrations in the core of a volcanic cloud exceed those in Figure 6, homogeneous nucleation can proceed rapidly and aerosol concentrations would be dominated by newly formed particles.

D-12
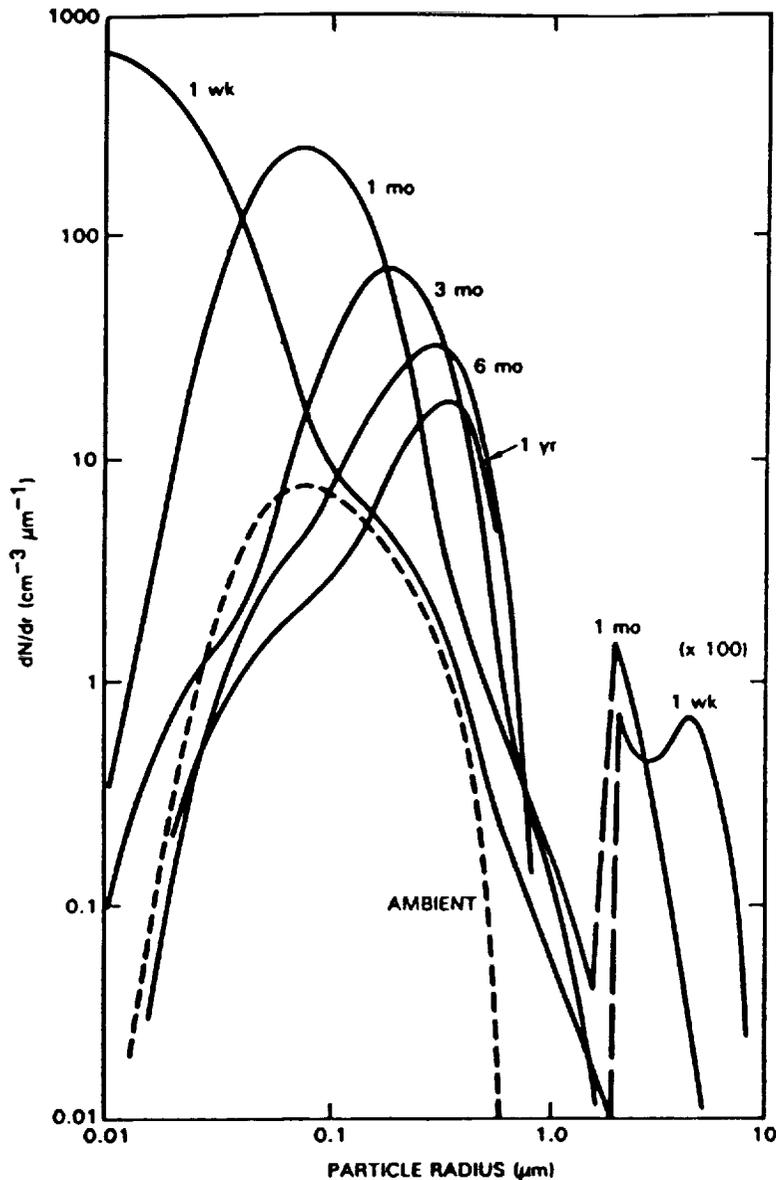
Figure 9. Evolution of the aerosol size distribution at 20 km in the simulated El Chichon eruption cloud. Size distributions are shown at early and late times, and are compared to the ambient size distribution.

The simulated size distributions of volcanic aerosols in Figure 9 exhibit a tri-modal structure that varies over time. The principal modes are: nucleation mode, which is most prominent at early times and at sizes near 0.01 μm; sulfate accumulation mode, which evolves initially from the nucleation mode (by coagulation and condensation) and increases in size to about 0.3 μm after one year; large-particle 'ash' mode (of solid mineral and salt particles) that settles out of the layer within the first month. A primary feature of the volcanic aerosol size distribution after several months is a greatly enhanced sulfate accumulation mode. The mass residing in this mode is indicated in Figure 7. The increased aerosol size is caused by an accelerated growth rate in the presence of enhanced sulfuric acid vapor concentrations maintained by SO_2 chemical conversion.

D-13
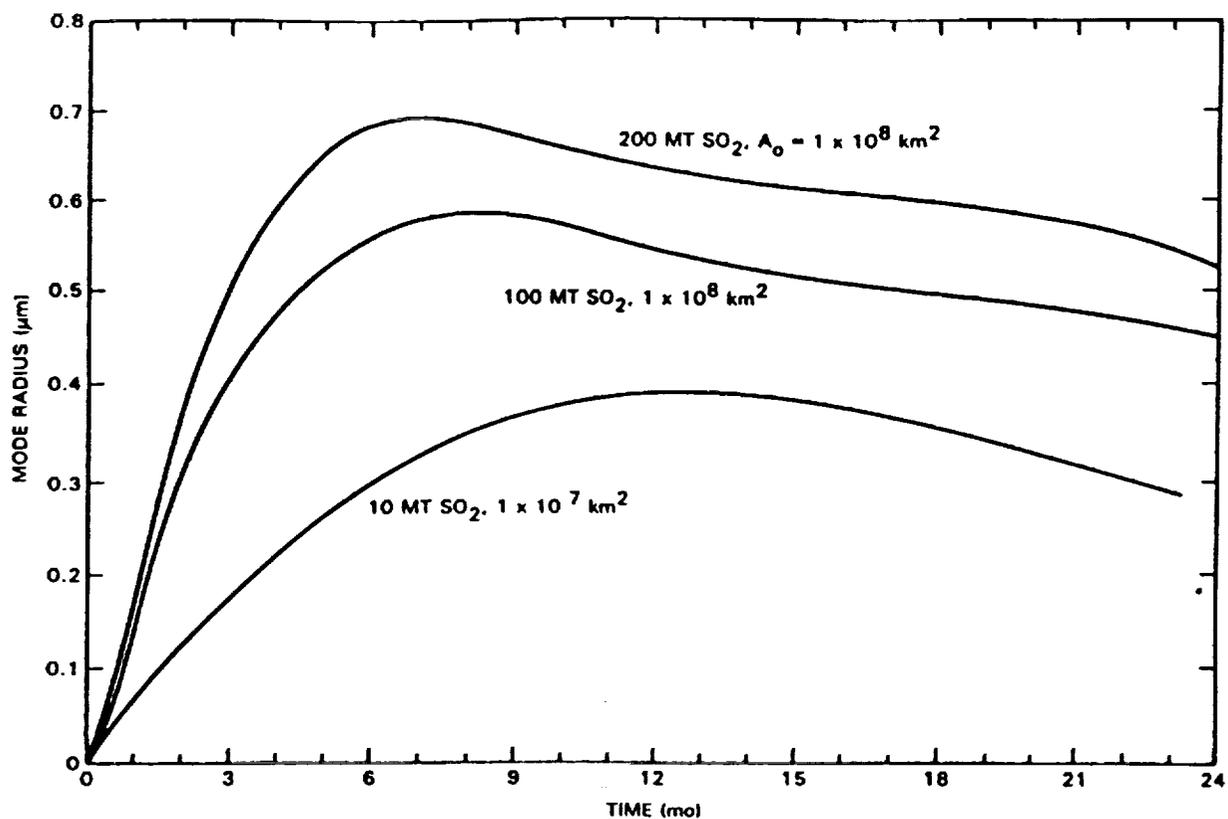
Figure 10. Mode radius of an equivalent log-normal size distribution that fits the computed sulfate accumulation mode versus time in a volcanic eruption cloud. The SO_2 injections that correspond to these simulations are indicated on the figure in megatons (MT) or 10^6 metric tons. The area, A_0, is the initial area over which the volcanic debris is assumed to be distributed; the cloud subsequently expands horizontally over the globe, with an equivalent horizontal diffusion coefficient of 1×10^{11} cm^2/sec. Note that the mode radius approaches zero at early times in these simulations because of the large number of extremely small freshly-nucleated particles. (Pinto et al., 1989)

The sulfate aerosol mode radii illustrated in Figure 10 correspond to SO_2 mass injections of 10 MT (1 MT = 10^6 metric tons) (i.e., like El Chichon), and 100 and 200 MT (more like Tambora). The mode radius peaks earlier with larger SO_2 injections and remains elevated throughout the history of the eruption cloud. In these simulations, the mode radius grew as large as 0.7 µm, which greatly exceeds the ambient mode radius of ~0.05 µm. In the case of large eruptions, the mode radius slowly returns to its ambient value, requiring a period of more than three years. The mode radius is important in determining the rate at which sulfate is removed from the stratosphere. The mass flux is proportional to the fallspeeds of the aerosols multipl;ied by their mass. Since $m \propto r^3$ and $v \propto r^{1 \to 2}$ in the regime of interest, the mass loss rate is, $\dot{m} \propto r^{4 \to 5}$

OPTICAL DEPTH AT 550 nm

3

$200 MT SO_2, A_o = 1 \times 10^8 km^2$

2

$100 MT SO_2, 5 \times 10^7 km^2$

$100 MT SO_2, 1 \times 10^8 km^2$

HEMISPHERIC
DISPERSION

1

$10 MT SO_2$
$1 \times 10^7 km^2$

GLOBAL DISPERSION

0

0   1   2   3   6   9   12   15   18   21   24

TIME (mo)
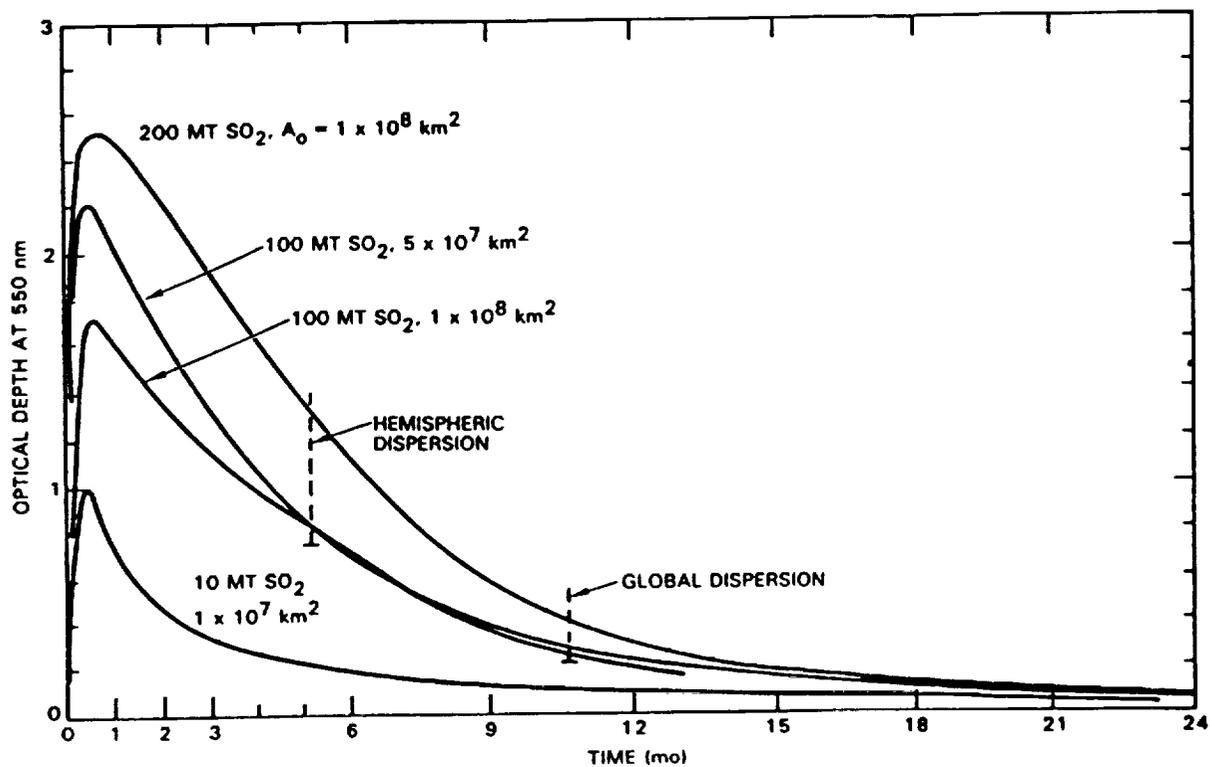
Figure 11. Volcanic aerosol optical depths (zenithal) versus time for the volcanic cloud simulations discussed in Figure 10. Indicated on the figure are the times required model times for the cloud to disperse over a hemisphere or over the globe. (Pinto et al., 1989)

Figure 11 illustrates the evolution of the volcanic aerosol optical depth at mid-visible wavelengths. The optical depth is associated with the scattering of light by the sulfuric acid droplets. At early times, and over limited geographical regions, the optical depths exceed a value of 2. However, after one year of evolution by growth, coagulation and fallout, the average optical depth for even the largest SO_2 injection has fallen to about 0.5. These results suggest that the nonlinear physical/chemical interactions occurring in volcanic eruption clouds will severely limit the aerosol optical depth that can be maintained over a period of several years -- i.e., the time span required to induce substantial climatic impacts. (Also see the following section)

Volcanic Aerosol Impact on Radiation and Climate

Many scientists, beginning with Benjamin Franklin, have studied the relation between volcanic explosions and climate/weather change (see Lamb, 1977; Robock, 1981). Existing research indicates that, in the years following a major volcanic eruption, anomalous weather patterns and regional- to global-scale cooling is likely (Robock and Mass, 1982; Rampino et al., 1988). The connection between volcanoes and climate is most directly made through the effect that volcanically-induced sulfate aerosols have on the global radiation budget (see Figure 12). The volcanic aerosols, when formed in the stratosphere where they can be rapidly dispersed around the earth, increase the reflectivity of the atmosphere and reduce incoming solar energy. The aerosols also produce a modest infrared trapping effect, which is not sufficient to reverse the cooling caused by the increased reflectivity (Pollack et al., 1976). The net effect of volcanic emissions of sulfur gases into the stratosphere is an average global cooling of ~ 0.5 $^{\circ}$C for 1-3 years. A number of statistical correlations have been uncovered between volcanic eruptions in one year and global cooling in the following years. For example, studies of tree ring records (LaMarche and Hirschboek, 1983) and similar climatic proxies (Lamb, 1977) support the volcanic cooling hypothesis (Hansen et al., 1978).

The violence of a volcanic eruption is not always directly related to its climatic impact. More important is the quantity of sulfur gases (particularly SO_2) injected into the stratosphere. However, the climate impact of an explosive eruption also appears to be limited to a temperature perturbations of ~ 1 $^{\circ}$C even for the largest eruptions. This fundamental limitation may be caused by highly nonlinear physical processes that restrict the buildup and persistence of the aerosol optical depth (Pinto et al., 1989; see Figure 11).
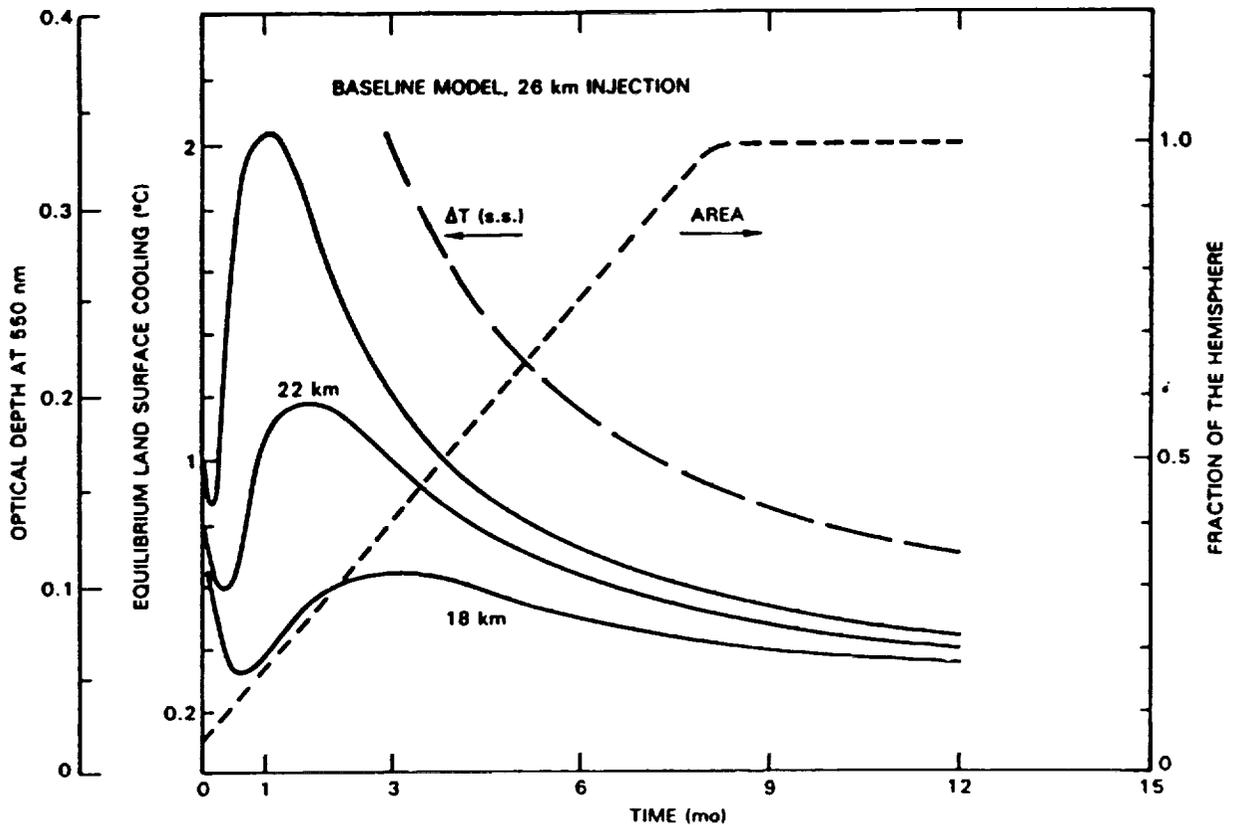
Figure 12. Time-dependent optical depth variations, and corresponding equilibrium surface temperature changes, for an El Chichon-like volcanic eruption of about 10 MT of SO_2 injected at stratospheric levels up to 26 km, 22 km, and 18 km, respectively, in three simulations. The steady-state (equilibrium) surface temperature variation is only shown for the 26 km injection. The horizontal area of the aerosol cloud -- as represented in the model -- is also given.

The temperature perturbations shown in Figure 12 correspond to the maximum attainable temperature decrease for an optical perturbation that is fixed long enough for the surface/atmosphere system to achieve its new equilibrium state. Thus, it is likely that an eruption such as El Chichon would produce global average coolings of perhaps 0.1-0.2 °C. The climatic impact decreases as the injection height of the precursor gases is lowered from 26 to 18 km (the latter value, for example, characterizes the height of sulfur injection in the Mt. St. Helens eruption).

D-17

Stratospheric Aerosols as Tropospheric Cloud Condensation Nuclei

After a major volcanic eruption, the stratospheric aerosols settle into the upper troposphere over a period of several years. These sulfate particles are highly soluble and thus enhance the cloud condensation nuclei (CCN) population of the upper troposphere (Pruppacher and Klett, 1978). It has also been suggested -- but remains speculative -- that such soluble aerosols may provide nuclei for cirrus cloud formation (through spontaneous freezing of supercooled droplets, which then act as ice nuclei). Meteoritic debris particles, which fall through the stratosphere, were once suggested as a source of cloud nuclei, but are unlikely to be important in this regard. Stratospheric particles, acting as nuclei, could modulate the microphysical and radiative properties of upper tropospheric clouds, and hence affect the global climate.

Polar Stratospheric Clouds

The properties of polar stratospheric clouds have been defined by a decade of satellite observations (McCormick et al., 1982; McCormick and Trepte, 1986, 1987; Hamill and Turco, 1988). On optical and physical evidence, PSC's fall into two broad categories, which we will refer to as Type-I and Type-II PSC's. Type-I PSC's consist of an aerosol haze of micron-sized nitric acid ice particles composed of HNO_3 and H_2O [in roughly a 50/50 mixture by weight, similar to the trihydrate $HNO_3 \cdot 3H_2O$] (Toon et al., 1986; Crutzen and Arnold, 1986). Type-II PSC's are apparently composed of water-ice crystals (Poole and McCormick, 1988; Hamill et al., 1986).

The Type-I PSC's are thought to be the most common form, perhaps accounting for 80-90% of all cloud sightings. These PSC's exhibit an onset at temperatures near 195 K, whereas the denser Type-II PSC's appear to condense at colder temperatures (<187 K) consistent with the measured frost point of water vapor in the polar stratosphere (Poole and McCormick, 1988; Hamill et al., 1988). It should be expected, therefore, that Type-I haze would predominate the totality of cloud observations in the earliest part of the Antarctic winter season, and that the frequency of Type-II clouds would increase with the progression of winter and cooling of upper air layers. On the other hand, the observed dehydration and denitrification of the polar winter stratosphere would, over the course of time, reduce the frequency of cloud formation at specific temperature thresholds. Hence, in the late winter and early spring, PSC's are predicted to dissipate abruptly when sunlight, absorbed by ozone, begins to warm the upper stratosphere (Toon et al., 1989).

The nuclei for nitric acid ice deposition are most likely to be the background sulfuric acid aerosols (Toon et al., 1986). Observational evidence on the extent to which sulfate particles are nucleated in PSCs is mixed. Aircraft measurements taken in the Antarctic ozone hole in September, 1987, suggest that many, if not most, of the sulfate particles may be activated into nitric acid haze particles (Fahey et al., 1989; Pueschel et al., 1989). On the other hand, balloonborne aerosol measurements conducted at McMurdo Station during the same period, and more recent data from the Arctic winter stratosphere, indicate that, although layers of ~1 micron-size haze particles are frequently present, on occasion the fraction of the sulfate particles activated into haze may be relatively small (e.g., Hofmann et al., 1989; Hofmann, 1990). If the sulfate particle nucleation is limited to a small fraction of the total number of particles, the resulting low concentration of haze particles will have

important implications for stratospheric denitrification (Salawitch et al., 1988; Toon et al., 1990).

In situ measurements of odd-nitrogen species provide direct evidence for the existence of nitric acid particles in the antarctic winter stratosphere (Fahey et al., 1989; Gandrud et al., 1989; and Pueschel et al., 1989). That evidence is supported by infrared spectroscopic measurements of condensed HNO_3 (Kinne et al., 1989) and column measurements of depletion of the HNO_3 vapor column amount (G. Toon et al., 1989; Coffey et al., 1989). In addition, Hanson and Mauersberger (1988a,b), in a series of detailed laboratory studies, have quantified the compositions and vapor pressures of the nitric acid ices under the conditions of interest, and concluded that the nitric acid trihydrate would be the stable form in the stratosphere.

Chemical transformations on PSC ice particles that lead to the formation of the ozone hole are discussed below. Extensive stratospheric ozone depletion may be influenced indirectly by volcanic sulfate aerosols through their role as PSC nuclei. However, sulfate aerosols may have a more direct role in ozone depletion chemistry by catalyzing special reactions (see below).

Aerosol Impact on Stratospheric Chemistry and Ozone

The recent discovery of an ozone "hole" over Antarctica in late austral winter and early spring (Farman et al., 1985) has led to considerable research on the causitive processes (e.g., Geophysical Research Letters, Volume 13, Number 12, 1986; Geophysical Research Letters, Volume 15, Number 8, 1988; Journal of Geophysical Research, Volume 94, Numbers D9 and D12, 1989; Geophysical Research Letters, Volume 17, Number 4, 1990). The leading chemical theories assume that "heterogeneous" reactions occur on polar stratospheric cloud (PSC) particles. Laboratory studies show that the ice-catalyzed reaction of HCl with chlorine nitrate ($ClONO_2$) produces 'active' chlorine. Through known homogeneous photochemical processes, this enhanced activated chlorine can explain most, if not all, of the measured ozone deficits (Solomon et al., 1986; Crutzen and Arnold, 1986; McElroy et al. 1986a,b).

The intense interest in polar stratospheric clouds has led to a new focus on the possible role of other stratospheric aerosols in heterogeneous chemical reactions that might affect stratospheric composition and ozone depletion. An early review of potential heterogeneous reactions on the background sulfate aerosols concluded that such reactions were too slow to be of any significance (Turco et al., 1982). However, that conclusion was based on early measurements of species reactivities on sulfuric acid surfaces that did not match precisely the properties of sulfuric acid particles under stratospheric conditions. New measurements of $ClONO_2$ reactivity on such surfaces (Tolbert et al., 1988) indicate a very strong dependence on solution composition, implying a potentially significant chemical impact. Accordingly, a reassessment of potential heterogeneous contributions to global stratospheric chemistry is timely.

Heterogeneous Chemical Processes on Stratospheric Aerosols

The key heterogeneous chemical processes that occur on polar stratospheric cloud particles are:

$$HCl(a) + ClONO_2(a) \longrightarrow Cl_2(g) + HNO_3(a) \tag{1}$$

$$HCl(a) + N_2O_5(a) \longrightarrow ClNO_2(g) + HNO_3(a) \tag{2}$$

$$ClONO_2(a) + H_2O(a) \longrightarrow HOCl(g) + HNO_3(a) \tag{3}$$

$$N_2O_5(a) + H_2O(a) \longrightarrow 2\,HNO_3(a)\,. \tag{4}$$
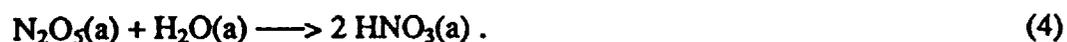
In these chemical equations, the abbreviation a indicates that the species is adsorbed on the particle surface or absorbed into the particle, and g indicates that the species rapidly desorbs from ice surfaces and is likely to be found in the gas phase. Chlorine activation via reactions 1-3 is very likely to proceed to completion on PSCs, with or without occasional solar illumination, in the early part of polar winter. This conclusion has recently been confirmed by laboratory studies (Moore et al., 1990). Chlorine activation -- in combination with denitrification, which is caused by PSC particle sedimentation -- results in a perturbed stratospheric composition that is extremely destructive toward ozone. A principal concern for the future is that increasing concentrations of background stratospheric chlorine from chlorofluorocarbons might be extensively activated over the entire globe on sulfate aerosols enhanced by anthropogenic activity (Hofmann, 1989) or generated suddenly by a major volcanic eruption.

Anthropogenic Influences on Stratospheric Aerosols

Human activities can affect the composition and structure of the stratosphere and stratospheric aerosols in a number of ways:

1) Carbon dioxide growth leads to a significant cooling of the middle and upper stratosphere thereby altering the dynamics, chemical kinetics and thermodynamics of sulfate particle formation;

2) Tropospheric emissions of sulfur-bearing gases such as OCS reach the stratosphere and upon decomposition modify the sulfate layer;

3) Direct emissions of sulfur vapors (SO_2) and soot from high-flying aircraft contaminate the natural sulfate layer;

4) Fluorocarbons released at the surface diffuse into the stratosphere and are decomposed into chlorine species that are subsequently activated on stratospheric aerosols and attack ozone;

5) Direct injection of rocket exhaust particles into the stratosphere, and the increasing rate of ablation of debris from orbiting platforms, enhances the concentration of stratospheric aerosols (Zolensky et al., 1989);

6) Other trace gases of tropospheric origin like methane and nitrous oxide are transported into the stratosphere and alter the ambient composition of the air in which stratospheric aerosols form and evolve (e.g., by increasing water vapor concentrations).

Figure 13 illustrates the potential change in the sulfate aerosol size distribution and optical depth resulting from an order-of-magnitude increase in either OCS or SO_2. The size distribution is noticeably skewed toward larger particle sizes, which have greater optical activity. The optical depth of these stratospheric aerosols increases substantially (by a factor of almost 5 for the OCS enhancement of a factor of 10), suggesting that their climatic impact could be significant (Turco et al., 1980).
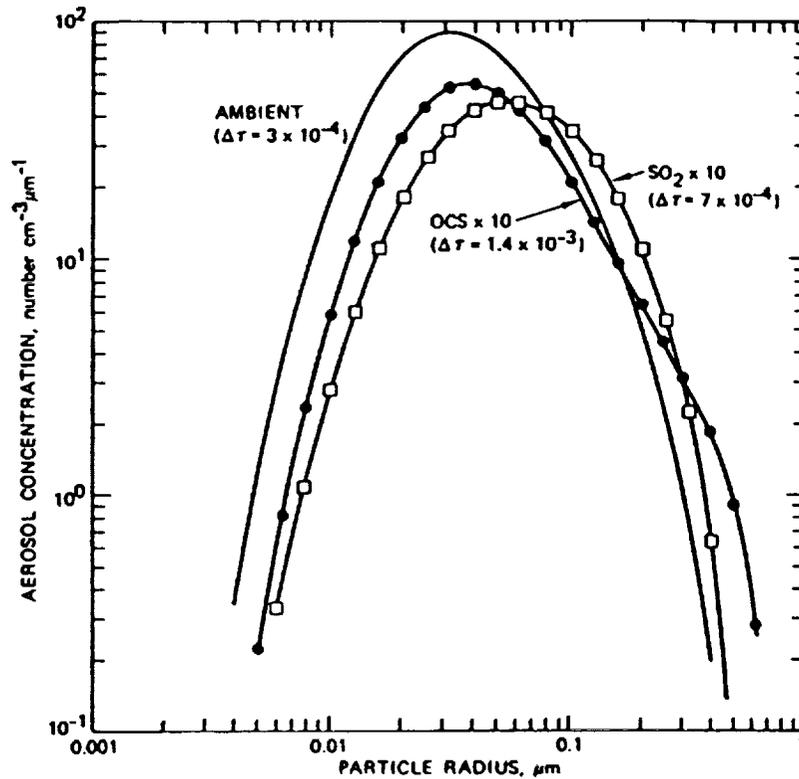
Figure 13. Calculated aerosol size distributions at 20 km for the background stratospheric sulfur gas abundances, compared to the size distributions corresponding to an increase by a factor of 10 in either the OCS or SO₂ ambient abundances. Also indicated in each case are the aerosol optical depths of a layer 1 km thick centered at a height of 20 km.

Stratospheric Aerosol Observations for Global Change

Stratospheric aerosols participate in global-scale processes that affect the troposphere and biosphere. The presence of aerosols directly influences solar radiation reaching the lower atmosphere and surface, and thus has a role in controlling the climate of earth. If stratospheric aerosols affect the properties of clouds in the upper troposphere, further climatic influences are implied. Sulfate aerosols may also modulate the chemistry of the ozone layer, leading to enhanced ozone depletion following major volcanic eruptions and an increase of harmful ultraviolet radiation at the ground. Stratospheric aerosols are subject to modification by a number of human activities, and these activities therefore may induce climate and other environmental changes. Table 3 lists a number of outstanding scientific questions regarding stratospheric sulfate aerosols -- and volcanic aerosols -- and their effects.

Table 3. Outstanding Scientific Issues

Sulfate Aerosols

Why is the background sulfate mass increasing?

How do the sulfate particles act as PSC nuclei?

Is increased sulfate aerosol reactivity possible, for example, with increasing water vapor?

What is the chemical nature of the unusual aerosol layers observed near 30-35 km?

How do stratospheric aerosols affect remote sensing of the upper atmosphere?

Are stratospheric particles a significant component of the global radiative energy balance under normal conditions, and thus of global climate change?

Volcanic Particulates

What are the types and frequencies of volcanic eruptions that produce global effects?

What is the injection efficiency of SO_2, HCl, H_2O and other gases?

Are there strong non-linear limits to the global effects of volcanic injections?

What is the precise composition of volcanic aerosols for heterogeneous chemistry?

Do volcanic aerosols act as nuclei for the formation of cirrus in the upper troposphere?

Is there a measurable effect of volcanic eruptions on climate, or is the effect illusory?

Do volcanic eruptions affect tropospheric phenomena such as El Nino?

What has been the role of ancient volcanic eruptions on the course of geological/biological evolution?

Can volcanic eruptions affect stratospheric ionization, conductivity and electric fields?

To be more certain of the possible connections between the stratosphere and troposphere that involve stratospheric aerosols and that have global change implications, specific scientific programs could be developed:

1) Volcanic aerosol studies: A program should be established to delineate the connection between volcanic emissions, aerosol formation and dispersion, and radiative/climatic impacts of volcanic eruptions; these studies could in part be carried out using the existing data base for the El Chichon eruption of 1982, although the project would aim toward collecting new data -- particularly through long-term satellite observations -- on future eruptions;

2) Aerosol heterogeneous chemistry studies: Accurate laboratory studies on the reactivity -- particularly the chlorine reactivity -- of sulfate (sulfuric acid) aerosols should be carried out under realistic stratospheric conditions; as opportunities arose, in situ chemical perturbations associated with enhanceed sulfate aerosols could be studied directly (e.g., following a volcanic eruption, or in a planned sulfur release);

3) Predictive model development: Theoretical models capable of predicting the effects of volcanic eruptions and various human activities on stratospheric aerosols, upper tropospheric aerosols, the global radiation budget and climate should be developed; such models would be global in scale -- optimally three-dimensional -- and include accurate treatments of the appropriate dynamical, photochemical, microphysical and radiative processes;

4) Global aerosol monitoring program: A global monitoring program for stratospheric aerosol trends -- particularly trends that may be associated with anthropogenic emissions but also transients associated with volcanic eruptions -- could be built upon existing and planned satellite and lidar measurement projects; in situ measurements would be necessary for calibration and detailed microphysical/radiative/chemical defintion of the aerosols over time.

Finally, the recent conclusions of the Ozone Trends Panel (Watson et al., 1990) on the roles of stratospheric aerosols in the collection of data from space -- particularly data on ozone abundances -- are briefly stated in Table 4.

Table 4. Stratospheric Aerosol Impact on Remote Sensing of the Upper Atmosphere (Ozone Trends Panel Report) ·

Ozone satellite observations (SBUV) suffer minimal degradation, with care required during periods of stratospheric disturbance by volcanic eruptions.

Umkehr ozone measurements suffer major perturbations during periods when the stratospheric aerosols are enhanced by volcanic activity.

Stratospheric aerosol monitoring for radiative transfer compensation of remotely-sensed data and for long-term trends is recommended.

Recent References (Partial Listing)

Coffey, M. T., W. G. Mankin and A. Goldman, "Airborne measurements of stratospheric constituents over Antarctica in the austral spring of 1987, 2, Halogen and nitrogen trace gases", J. Geophys. Res., 94, 16,597-16,613 (1989).

Crutzen, P. J., and F. Arnold, "Nitric acid cloud formation in the cold Antarctic stratosphere: A major cause for the springtime 'ozone hole'", Nature, 324, 651-655 (1986).

Fahey, D. W., K. K. Kelly, G. V. Ferry, L. R. Poole, J. C. Wilson, D. M. Murphy, M. Loewenstein and R. K. Chan, "In-situ measurements of total reactive nitrogen, total water vapor, and aerosols in a polar stratospheric cloud in the Antarctic", J. Geophys. Res., 94, 11,299-11,315 (1989).

Farman, J. C., B. G. Gardiner and J. D. Shanklin, "Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction", Nature, 315, 207-210 (1985).

Gandrud, B. W., P. D. Sperry, K. K. Kelly, G. V. Ferry and K. R. Chan, "Filter measurement results from the Airborne Antarctic Ozone Experiment", J. Geophys. Res., 94, 11,285-11,297 (1989).

Hamill, P., and R. P. Turco, "The ozone hole: The role of polar stratospheric cloud particles", Paper AIAA-88-0211, American Institute of Aeronautics and Astronautics, 7 pp., Washington, D.C. (1988).

Hamill, P., O. B. Toon and R. P. Turco, "Characteristics of polar stratospheric clouds during the formation of the Antarctic ozone hole", Geophys. Res. Lett., 13, 1288-1291 (1986).

Hamill, P., R. P. Turco and O. B. Toon, "On the growth of nitric and sulfuric acid aerosol particles under stratospheric conditions", J. Atmos. Chem., 7, 287-315 (1988).

Hansen, J. E., W. Wang and A. A. Lacis, "Mt. Agung eruption provides test of a global climatic perturbation", Science, 199, 1065-1068 (1978).

Hanson, D., and K. Mauersberger, "Vapor pressures of HNO_3/H_2O solutions at low temperatures", J. Phys. Chem., 92, 6167-6170 (1988a).

Hanson, D., and K. Mauersberger, "Laboratory studies of the nitric acid trihydrate: Implications for the south polar stratosphere", Geophys. Res. Lett., 15, 855-858 (1988b).

Hofmann, D. J., "Increase in the stratospheric background sulfuric acid aerosol in the past 10 years", submitted to Science (1989).

Hofmann, D. J., "Stratospheric cloud micro-layers and small-scale temperature variations in the Arctic in 1989", Geophys. Res. Lett., 17, 369-372 (1990).

Hofmann, D. J., J. M. Rosen, J. W. Harder and J. V. Hereford, "Balloonborne measurements of aerosol, condensation nuclei, and cloud particles in the stratosphere at McMurdo Station Antarctica during the spring of 1987", J. Geophys. Res., 94, 11,253-11,269 (1989).

Jones, R. L., J. Austin, D. S. McKennaa, J. G.Anderson, D. W. Fahey, C. B. Farmer, L. E. Heidt, K. K. Kelly, D. M. Murphy, M. H. Proffitt, A. F. Tuck and J. F. Vedder, "Lagrangian

photochemical modeling studies of the 1987 Antarctic spring vortex. 1. Comparison with AAOE observations", J. Geophys. Res., 94, 11,529-11,558 (1989).

Junge, C. E., "Sulfur in the statosphere", J. Geophys. Res., 68, 3975 (1963).

Junge, C. E., and J. E. Manson, "Stratospheric aerosol studies", J. Geophys. Res., 66, 2163-2182 (1961)

Kinne, S., O. B. Toon, G. C. Toon, C. B. Farmer, E. V. Browell and M. P. McCormick, "Measurements of size and composition of particles in polar stratospheric clouds from infrared solar absorption spectra", J. Geophys. Res., 94, 16,481-16,491 (1989).

LaMarche, , and Hirshboek, " ", Science, (19).

Lamb, H. H., Climate: Present, Past and Future, Vol. 2, Climatic History and the Future, Methuen, London (1977).

McCormick, M. P., and C. R. Trepte, "SAM II measurements of Antarctic PSC's and aerosols", Geophys. Res. Lett., 13, 1276-1279 (1986).

McCormick, M. P., and C. R. Trepte, "Polar stratospheric optical depth observed between 1978 and 1985", J. Geophys. Res., 92, 4297-4306 (1987).

McCormick, M. P., H. M. Steele, P. Hamill, W. P. Chu and T. J. Swissler, "Polar stratospheric cloud sightings by SAM II", J. Atmos. Sci., 39, 1387-1397 (1982).

McElroy, M. B., R. J. Salawitch, S. C. Wofsy and J. A. Logan, "Reductions of Antarctic ozone due to synergistic interactions of chlorine and bromine", Nature, 321, 759-762 (1986a).

McElroy, M. B., R. J. Salawitch and S. C. Wofsy, "Antarctic O_3: Chemical mechanisms for the spring decrease", Geophys. Res. Lett., 13, 1296-1299 (1986b).

Moore, S. B., L. F. Keyser, M.-T. Leu and R. P. Turco, "Heterogeneous reactions on nitric acid trihydrate particles: Implications for the ozone hole", submitted to Nature (1990).

Pinto, J. P., R. P. Turco and O. B. Toon, "Self-limiting physical and chemical effects in volcanic eruption clouds", J. Geophys. Res., 94, 11,165-11,174 (1989).

Pollack, J. B., O. B. Toon, C. Sagan, A. Summers, B. Baldwin and W. Van Camp, "Volcanic explosions and climatic change: A theoretical assessment", J. Geophys. Res., 81, 1071-1083 (1976).

Poole, L. R., and M. P. McCormick, "Airborne lidar observations of arctic polar stratospheric clouds: Indications of two distinct growth stages", Geophys. Res. Lett., 15, 21-23 (1988).

Pruppacher, H. R., and J. D. Klett, Microphysics of Clouds and Precipitation, D. Reidel, Dordrecht, Holland (1980).

Pueschel, R. F., K. G. Snetsinger, J. K. Goodman, O. B. Toon, G. V. Ferry, V. R. Oberbeck, J. M. Livingston, S. Verma, W. Fong, W. L. Starr and R. K. Chan, "Condensed nitrate, sulfate and chloride in the Antarctic stratospheric aerosols", J. Geophys. Res., 94, 11,271-11,284 (1989).

Rampino, M. R., S. Self and R. B. Stothers, "Volcanic winters", Ann. Rev. Earth Plaet. Sci., 16, 73-99 (1988).

Robock, A., "A latitudinally dependent volcanic dust veil index, and its effect on climate simulations", J. Volcan. Geotherm. Res., 11, 67-80 (1981).

Robock, A., and C. Mass, "The Mount St. Helens volcanic eruption of 18 May 1980: Large short-term surface temperature effects", Science, 216, 628-630 (1982).

Salawitch, R. J., S. C. Wofsy and M. B. McElroy, "Influence of polar stratospheric clouds on the depletion of ozone over Antarctica", Geophys. Res. Lett., 15, 871-874 (1988).
Solomon, S., R. R. Garcia, F. S. Rowland and D. J. Wuebbles, "On the depletion of Antarctic ozone", Nature, 321, 755-758 (1986).

Tolbert, M. A., M. J. Rossi and D. M. Golden, "Heterogeneous interactions of chlorine nitrate, hydrogen chloride, and nitric acid with sulfuric acid surfaces at stratospheric temperatures", Geophys. Res. Lett., 15, 847-850 (1988).

Toon, G. C., C. B. Farmer, L. L. Lowes, P. W. Schaper, J.-F. Blavier and R. H. Norton, "Infrared aircraft measurements of stratospheric composition over Antarctica during September 1987", J. Geophys. Res., 94, 16,571-16,596 (1989).

Toon, O. B., P. Hamill, R. P. Turco and J. Pinto, "Condensation of HNO_3 and HCl in the winter polar stratospheres", Geophys. Res. Lett., 13, 1284-1287 (1986).

Toon, O. B., R. P. Turco, J. Jordan, J. Goodman and G. Ferry, "Physical processes in polar stratospheric ice clouds", J. Geophys. Res., 94, 11,359-11,380 (1989).

Toon, O. B., R. P. Turco and P. Hamill, "Denitrification mechanisms in the polar stratospheres", Geophys. Res. Lett., 17, 445-448 (1990).

Turco, R. P., R. C. Whitten, O. B. Toon, J. B. Pollack and P. Hamill ,"OCS, stratospheric aerosols and climate", Nature, 283, 283-286 (1980).

Turco, R. P., R. C. Whitten and O. B. Toon, "Stratospheric aerosols: Observation and theory", Rev. Geophys., 20, 233-279 (1982).

Turco, R. P., O. B. Toon, R. C. Whitten, P. Hamill and R. G. Keesee, "The 1980 eruptions of Mount St. Helens: Physical and chemical processes in the stratospheric clouds", J. Geophys. Res., 88, 5299-5319 (1983).

Watson, R. T., et al., Report of the Ozone Trends Panel, Chapters 10 and 11, Washington, D. C., in press (1990).

Zolensky, M. E., D. S. MacKay and L. A. Kaczor, "A tenfold increase in the abundance of large particles in the stratosphere, as measured over the period 1976-1984", J. Geophys. Res., 94, 1047-1056 (1989).

Volcanic Effects on Climate

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ABSTRACT

Volcanic eruptions which inject large amounts of sulfur-rich gas into the stratosphere produce dust veils which last several years and cool the earth's surface. At the same time, these dust veils absorb enough solar radiation to warm the stratosphere. Since these temperature changes at the earth's surface and in the stratosphere are both in the opposite direction of hypothesized effects from greenhouse gases, they act to delay and mask the detection of greenhouse effects on the climate system.

Tantalizing recent research results have suggested regional effects of volcanic eruptions, including effects on ENSO. In addition, a large portion of the global climate change of the past 100 years may be due to the effects of volcanoes, but a definitive answer is not yet clear. While effects of several years have been demonstrated with both data studies and numerical models, long-term effects, while found in climate model calculations, await confirmation with more realistic models. Extremely large explosive prehistoric eruptions may have produced severe weather and climate effects, sometimes called a "volcanic winter."

Complete understanding of the above effects of volcanoes is hampered by inadequacies of data sets on volcanic dust veils and on climate change. Space observations can play an increasingly important role in an observing program in the future. The effects of volcanoes have not adequately been separated from ENSO events, and climate modeling of the effects of volcanoes is in its infancy. Specific suggestions are made for future work to improve our knowledge of this important component of our climate system.

1. Introduction

Since 1784, when Benjamin Franklin suggested that the Hecla eruption in Iceland in 1783 might have been responsible for the abnormally cold winter of 1783-4 (Franklin, 1784), emissions from volcanoes have been implicated as a possible cause of weather and climate variations. Although conventional wisdom (Ramanathan, 1988; Self and Rampino, 1988) holds that the effects of volcanoes on climate have not yet been demonstrated, much work has already shown that volcanoes can be important causes of hemispheric temperature changes for several years following large eruptions and even on a 100-year time scale, when their cumulative effects are taken into account. It has also been suggested that they can influence atmospheric circulation, and interact with the El Niño/Southern Oscillation (ENSO) in complex ways, perhaps even causing ENSOs. Because the climatic signal of volcanic eruptions is of approximately the same amplitude as

that of ENSO, and because there have been so few large eruptions in the past century, it has been difficult to separate the volcanic signal from that of other simultaneous climatic variations. Large prehistoric eruptions, such as those of Toba, must have had severe climatic consequences, and such eruptions will occur in the future. In order to improve our understand of the effects of volcanoes on climate, more research is necessary.

2. Data on Volcanic Eruptions

It has become clear in the last decade (*e.g.*, Rampino and Self, 1984) that the effect of a volcano on climate is most directly related to the sulfur content of emissions that reach into the stratosphere, and not to the explosivity of the eruption. These sulfur gases convert to small sulfate particles, which persist for several years in the stratosphere and efficiently scatter the incoming sunlight, reducing the direct and total solar radiation reaching the ground.

In order to investigate the effects of volcanic eruptions on climate, it would be desirable to have a volcanic index that is proportional to the physical effect of the volcanic dust veil on climate, namely the net radiation deficit. If the index is incomplete in its geographical or temporal coverage, if it assumes that surface air temperature drops after an eruption and uses this information to create the index, or if it is a measure of some property of volcanic eruptions other than its long-term stratospheric dust loading, it will be unsuitable for this type of study. All volcanic indices produced so far suffer from one or more of these problems. Yet if the various deficiencies of each index are kept in mind, they can be used cautiously, which has not been the case in many instances, as discussed in the next section.

The first extensive modern compilation of past volcanic eruptions is the classic study of Lamb (1970), updated by Lamb (1977, 1983). Lamb created a volcanic Dust Veil Index (DVI), specifically designed for analyzing the effects of volcanoes on "surface weather, on lower and upper atmospheric temperatures, and on the large-scale wind circulation." (Lamb, 1970, p. 470) The methods used to create the DVI are described by Lamb (1970), and in more detail by Kelly and Sear (1982), and include historical reports of eruptions, optical phenomena, radiation measurements (for the period 1883 onward), temperature information, and estimates of the volume of ejecta.

The formula for the DVI includes a term Emax, which gives an estimate of the fraction of the globe covered by the dust veil. In order to compare the amount of material emitted from volcanoes, it is convenient to present DVI = d.v.i./Emax, as was done in Table 1. Although the DVI for the Mt. St. Helens eruption of 1980 was 500, and DVI for El Chichón of 1982 was 800, Emax for Mt. St. Helens was 0.3, while Emax for El Chichón was 1. Therefore, Lamb's (1983) estimate of the relative climatic effect of the two volcanoes was different by a factor of more than 5.

Lamb's DVI has been often criticized (*e.g.*, Bradley, 1988) as having used climatic information in its derivation, thereby resulting in circular reasoning if the DVI is used as an index to compare to temperature changes. In fact, for only a few eruptions between 1763 and 1882 was the Northern Hemisphere (NH)

averaged DVI calculated based solely on temperature information. Robock (1981a) created a modified version of Lamb's DVI which excluded temperature information. When used to force a climate model, the results did not differ significantly from those using Lamb's original DVI, demonstrating that this is not a serious problem.

Mitchell (1970) also produced a time series of volcanic eruptions for the period 1850-1968 using data from Lamb. As discussed by Robock (1978, 1981a), the Mitchell volcanic compilation for the NH is more detailed than Lamb's, because Lamb excluded all volcanoes with DVI < 100 in producing his NH annual average DVI (Table 7(a), p. 526). Thus Mitchell's volcanic series has proven to be very useful as a climatic volcanic index.

More recently, a comprehensive survey of past volcanic eruptions (Simkin *et al.*, 1981) produced the Volcanic Explosivity Index (VEI) (Newhall and Self, 1982) which gives a geologically-based measure of the power of the volcanic explosion. Unfortunately, this index has been used in many studies (see next section) as an index of the climatological impact of volcanoes without any modification. A careful reading of Newhall and Self (1982), however, will find the following quotes: "We have restricted ourselves to consideration of volcanological data (no atmospheric data). . ." and "Since the abundance of sulfate aerosol is important in climate problems, VEI's must be combined with a compositional factor before use in such studies." In their table 1 they list criteria for estimating the VEI in "decreasing order of reliability," and the very last criterion out of 11 is "stratospheric injection." For VEI of 3, this is listed as "possible," for 4 "definite," and for 5 and larger "significant." If one attempts to work backwards, and use a geologically determined VEI to give a measure of stratospheric injection, serious errors can result. Not only is this the least reliable criterion for assigning a VEI, but it was never intended as a description of the eruption which had a VEI assigned from more reliable evidence.

Eruptions with a high VEI may also have a large stratospheric impact, such as Tambora (1815, VEI = 7) or Krakatau (1883, VEI = 6), but 3 recent examples demonstrate the danger in using the VEI for climate studies. Mt. St. Helens in 1980 had a high VEI of 5, and while it had a large local temperature impact (Robock and Mass, 1982; Mass and Robock, 1982), it had a negligible stratospheric impact (Robock, 1981b)., Agung in 1963 and El Chichón in 1982, on the other hand, had a very large stratospheric impact (Robock, 1983a) but a smaller VEI of 4. Several studies (discussed below) have been done using the VEI as an index for the climatic effect of volcanoes, and then excluded Mt. St. Helens as a special case. This example raises the question of the possibility of other special cases in the past for which we do not have the additional information as in this case.

Schönwiese (1988) has even created a Smithsonian Volcanic Index (SVI) which takes 10 to the VEI power, and includes volcanoes with VEI of 3 and greater. This is clearly not justified.

As mentioned by Newhall and Self (1982), by combining information about the typical type of eruption that each volcano produces with the VEI, it may be possible to produce a "climatic VEI," but it will probably be necessary to include additional information to produce a good index of the climatic impact of past

eruptions.

Ice core analysis (*e.g.*, Lyons *et al.*, 1988) can give the chemistry and particle content of well-dated layers, which can give a measure of the important volcanic parameters. Xu (1988) has actually used the Acidity Index of Hammer *et al.* (1980) as a volcanic index for comparison to climatic data. Unfortunately, small local eruptions can give as large a signal as distant large eruptions. By comparing the acidity and particle records from Greenland, Antarctic, and tropical ice cores, it may be possible to produce a global or hemispheric record from signals that appear simultaneously in all 3 regions or in the tropics and one of the high latitude cores.

Radiation measurements of the transmission of the direct solar beam give indications of the atmospheric turbidity. By combining measurements from many locations to eliminate local influences, Pollack *et al.* (1976), Pivovarova (1977), and Bryson and Goodman (1980) presented time series of radiation, interpreted as the volcanic loading of the atmosphere. Xu (1985) created a volcanic index based on radiation data. and Mass and Portman (1988) also give other sources of actinometric data. Because each of these data sets is incomplete spatially and temporally, and because local influences may not have been completely eliminated, by themselves they are not sufficient as a measure of volcanic influence on the atmosphere. By combining all the available radiation information, however, they would be a valuable input to a volcanic index.

Another source of information comes from lunar brightness during eclipses (Keen, 1983). In addition, lidar measurements (*e.g.*, McCormick and Osborn, 1986), balloon sampling (*e.g.*, Hoffmann and Rosen, 1984a,b), and aircraft sampling (*e.g.*, Sedlacek *et al.*, 1983) can all now give detailed measurements of the stratospheric aerosol concentration. Satellite data from the SAM II (*e.g.*, McCormick and Brandl, 1986) and SAGE measurements (*e.g.*, McCormick, 1987) also give measures of stratospheric aerosols. An instrument designed for measuring ozone, TOMS on Nimbus 7, can also pick up the signal of sulfur dioxide from volcanic eruptions, and in fact has been used by Krueger (personal communication) to identify the source of the "mystery cloud" of early 1982 as the Nyamuragira eruption of 26 December 1981 in eastern Zaire.

Until a good climatic-volcanic index is developed, all previous studies using inadequate indices must be evaluated cautiously. Since DVI, VEI and acidity index are correlated (Schönwiese, 1988), the results presented below are in partial agreement even if based on different indices. An objective, quantitative measure of the effects of volcanoes on climate, however, will require a better volcanic index.

3. Volcanic Effects on Temperature

There have been many studies in the past attempting to link climatic changes with large volcanic eruptions. (Lamb (1970) even took temperature drops as indications of the size of volcanic eruptions, in a few cases without any other evidence, when creating his volcanic index.) These studies range from case studies of a single large eruption or a few eruptions, to comparisons of time series of temperature to the timing of eruptions, to superposed epoch analyses combining the signals of many eruptions. The different studies distinguish themselves from each

other by their choices of volcanoes (or volcanic indices), temperature data sets (usually air at the surface, but also upper air and sea surface), time resolution, analysis technique (especially whether climatic data are normalized by their standard deviation for monthly data), and treatment of ENSO signal. In this section the effects of the 1815 eruption of Tambora are discussed, and then analyses of more recent eruptions are compared.

A. Tambora – Cause of the "Year Without a Summer?"

The book by Stommel and Stommel (1983), which is subtitled, "The Story of 1816, The Year Without a Summer," presents the fascinating story of the severe weather disruptions in New England and Western Europe, which also resulted in 1816 being called, "Eighteen Hundred and Froze-to-Death" and "Poverty Year" (Humphreys, 1940). Stommel and Stommel's book includes the stories of the record price of grain in London, Mary Shelley writing Frankenstein influenced by the terrible summer weather on the shores of Lake Geneva, and the killing summer frosts in the United States and Canada (Robock, 1984c). They conclude that the case cannot be proven that the great eruption of Tambora in 1815 was responsible for the extreme weather of the next year because evidence was only available from a small region of the globe (eastern North America and Western Europe).

Recent studies, however, present new evidence of climate effects both in China and India in 1816, although Kondo (1988) found no evidence of effects in Japan. Hameed et al. (1989) found evidence in Chinese documents of abnormally cold weather from the winter of 1815-16 through the summer of 1817, manifesting itself in crop failures and snow in June, 1816. Sigurdsson and Carey (1988) point out the bad harvests in India in 1816 that led to a famine which was followed by a serious cholera outbreak. In the next 2 decades the cholera spread as the greatest pandemic of the century to Europe and Asia.

One aspect of the 1816 events that is not widely recognized is that their were significant volcanic eruptions in each of the 4 years preceding Tambora (Sabrina in 1811, Soufriere and Awu in 1812, Vesuvius in 1813, and Mayon in 1814), so that any effects felt in 1816 were the cumulative effect of 5 years of enhanced stratospheric aerosol loading. Stothers (1984) and Rampino and Self (1982), who presented a detailed geological description of the Tambora eruption, found a NH temperature depression of approximately 0.7°C in 1816 from a limited network of stations. Humphreys (1940) similarly found a depression of about 1.0°C in 1816. Both studies and Groveman and Landsberg (1979) all found that the NH temperature was cool for several years before 1816, and then rose rapidly, by more than 1°C, during the next 10 years. The antecedent cooling can be easily explained by the effects of the preceding volcanoes, but the subsequent strong warming is more difficult to understand. How good were the temperature records? Was this a response to a dust-free atmosphere? Were internal oscillations becoming dominant? Was there strong tropical ocean forcing of the climate system during this period? If so, was it made stronger by the volcanoes? Quinn et al. (1987) report no ENSO events from 1814 to 1828, but their record may be incomplete.

Thus, the case of the volcanic eruptions of 1811-1815 and the severe weather of 1816 is strongly suggestive of the large potential short-term effect of volcanoes on climate. One individual case cannot prove the relationship, however, since

other causes of interannual variability were undoubtedly playing a part simultaneously, but with unknown amplitude. Next, studies which combine the effects of several eruptions (although none with as large a stratospheric impact as Tambora) and study the effects on temperature with the improved data network of the past 100 years are presented.

B. Comparative Studies

Humphreys (1940), Yamamoto *et al.* (1975), Angell and Korshover (1985), Kondo (1988), Angell (1988), and Xu (1988) all present time series of volcanic eruptions and climate change and comment on the correspondence. Robock (1990) contains a table comparing all these studies and the volcanoes used in each one. In each case the evidence is suggestive of a cause and effect, with varying degrees of agreement. All use surface air temperatures, except Kondo also used reports of crop failures and famines in Japan, Xu used reports of cold summers in China, and Angell also used sea surface and upper air temperatures.

Although visually comparing time series can suggest agreements, the superposed epoch technique, discussed next, can objectively filter out other effects and give a quantitative measure of the volcanic effect. Of course, if some other cause of climate variation is correlated with volcanic eruptions, the superposed epoch technique will not remove it. This seems to be the case with ENSOs, and Angell (1988) showed that, by removing a signal correlated with SSTs in the tropical Pacific with a 6-month lag, the volcanic signal is made clearer. This argument still may suffer from circular reasoning, since the SST is also part of the signal being measured.

C. Superposed Epoch Analyses

In superposed epoch analysis, a key date is identified for each volcanic eruption, the resulting temperature data are superposed on each other, and then the average is used to measure the signal of volcanic eruptions. This has been done for data-averaging periods on time scales ranging from 5 years (Mitchell, 1961) to 1 year (Mass and Schneider, 1977; Schönwiese, 1988) to 1 season (Taylor *et al.*, 1980; Angell and Korshover, 1985; Lough and Fritts, 1987) to 1 month (Self *et al.*, 1981; Kelly and Sear, 1984; Sear *et al.*, 1987; Bradley, 1988; Mass and Portman, 1989).

When a 1 month averaging period is used, the months are counted starting with the month of the eruption, so that if two volcanoes occurred in different years, say one in April and one in August, then the effects after three months would be an average of July and November data. Thus, if there is a seasonal component to the response of the climate system, it cannot be identified with this technique. Recognizing that climate variability is larger in the winter, in order to avoid averaging large and small variations together, Kelly and Sear (1984), Sear *et al.* (1987), and Bradley (1988) have all looked at normalized monthly surface air temperatures, with the temperature anomalies divided by the standard deviation of temperature for that month. However, this analysis technique gives less weight to winter temperature fluctuations and also works to obscure the seasonal cycle of temperature response.

Mass and Portmann (1989) have removed an ENSO signal, in a manner similar to Angell (1988), and find a definite volcanic signal.

D. Seasonal Cycle Analysis

It has been shown by Robock (1983b) that the sea ice/thermal inertia feedback is responsible for the amplification of climate response in high latitudes and in winter for equilibrium climate simulations with an energy-balance climate model. This also explains the results Manabe and Stouffer (1980) obtained with a GCM. For transient experiments with volcanic eruptions (Robock, 1981b, 1984a; discussed below) and for nuclear winter forcing (Robock, 1984b; Vogelmann et al., 1988) it has also been shown with an energy-balance model that the sea ice/thermal inertia feedback causes an amplification of the seasonal cycle when the climate system cools, resulting in more cooling in the polar regions in the winter. Yamamoto et al. (1975) also found winter polar enhancement of the volcanic cooling for several eruptions.

Since volcanic eruptions are thought to result in cooling of the climate system for a few years, Robock (1985) has presented a preliminary analysis in which the amplitude of the seasonal cycle in high latitudes is examined by doing an analysis of surface temperature that compares all months from different years. This analysis also solves another problem of previous studies, namely that large volcanic eruptions can sometimes occur close to each other in time, and a superposed epoch analysis must make the assumption that the year or years before the key date (date of the eruption) represent a normal climate. Since the climate system is constantly fluctuating, this analysis examines the overall level of volcanic forcing and compares it to the corresponding response of the climate system.

Robock (1985) presented a volcanically-weighted temperature variation, but it seems more straight-forward to simply present the correlation coefficient between temperature variations and a volcanic index. It is expected that if such an analysis were done with the ENSO signal removed from an improved temperature data set, such as the global set from the University of East Anglia, (Jones et al., 1988), with an improved volcanic index, that it will be possible to establish a typical volcanic signal in a more definitive manner than before.

E. Stratospheric Effects

Even though the data record is shorter, large warming beyond the level of the quasi-biennial oscillation has been measured in the stratosphere following the Agung and El Chichón eruptions (e.g., Labitzke et al., 1983, Parker and Brownscombe, 1983; Angell and Korshover, 1983; Quiroz, 1983, 1984; Fujita, 1985; Wendler and Kodama, 1986). This is in the opposite direction of anticipated effects from greenhouse cases which will be "virtually certain" to cause large stratospheric cooling (National Research Council, 1987). Thus these large stratospheric effects of volcanoes, while only lasting a few years, will have to be dealt with when modeling or interpreting climatic data in attempts to identify a greenhouse signal.

5. Climate Model Calculations

A. Energy-Balance Models

Energy-balance climate model calculations by Schneider and Mass (1975); Oliver (1976); Robock (1978, 1979, 1981b, 1984a); Gilliland (1982); and Gilliland and Schneider (1984) have all shown cooling effects due to volcanic eruptions for several years. As mentioned above, Robock (1984a) demonstrated the winter polar enhancement of cooling due to the sea ice/thermal inertia feedback, the same pattern seen in the observations (Fig. 1).

Robock (1978, 1979, 1981b) demonstrated a long-term effect of volcanic eruptions with the warming of the 1920s and 1930s resulting from the lack of significant eruptions. Oerlemans (1988) coupled an energy-balance climate model with a glacier model and found that about half of the observed long-term behavior of glaciers for the past 100 years can be explained by volcanoes and half by greenhouse gases. (Porter (1981) had previously found a relationship between glacier advances and volcanoes for the past 100 years.) The models used for these calculations had only a 75-m mixed layer ocean, and these longer time scales of the climate system may depend on deep ocean circulation (Hansen et al., 1985). Experiments with coupled atmospheric and oceanic GCMs will be necessary to confirm this result.

B. Radiative-Convective Models

Hansen et al. (1978) and Vupputuri and Blanchet (1984) have used radiative-convective models to calculate the vertical distribution of the climatic effect of the Agung and El Chichón eruptions, respectively. They both found cooling at the surface and warming in the stratosphere, which corresponds to the observations mentioned above.

C. Zonally-Averaged Model

MacCracken and Luther (1984) used a zonally-averaged dynamic climate model to calculate the vertical and latitudinal response to the El Chichón eruption. They found cooling at the surface in agreement with the energy-balance calculations of Robock (1984a), but in addition found intriguing precipitation and circulation anomalies caused by a shift in the ITCZ, which they suggested may be related to El Nino generation.

D. General Circulation Model (GCM) Simulations

Hunt (1977) presented the first GCM calculation of the effects of a volcano on climate, but it was done with a crude model and did not examine seasonal effects. Hansen et al. (1988) recently performed time-dependent simulations of climate from 1960 through 2050 with a GCM at the NASA Goddard Institute for Space Sciences (GISS). In three different simulations with different amounts of greenhouse gases, the effects of a total of 12 large and 9 small volcanoes were shown to cause cooling for several years. Although Hansen et al. present hemispheric, annual average results of these simulations, showing cooling from the volcanoes, the seasonal and latitudinal signal of the volcanoes has not been analyzed in their output. In addition, the simple ocean model used precluded the precise determination of long-time scale effects.

It is obvious that only with GCM studies of the effects of volcanic eruptions will the subtle interactions of the climate system, including possible ENSO or monsoon relationships, and the geographical distribution of effects be determined. Currently ongoing experiments include those of Graf (1989) at the Max Planck Institute in Hamburg and of Hansen, McCormick and Pollack (Hansen *et al.*, 1988) at GISS in New York.

5. Relationship to ENSO

On 4 April 1982, the El Chichón volcano in Mexico erupted, putting more aerosols into the stratosphere than any other volcano in this century, with the possible exception of Agung in 1963 (Robock, 1983). Although climate model calculations (Robock, 1984a; Hansen *et al.*, 1988) suggested a large drop in hemispheric average surface air temperatures for several years following the eruption, 1983 was in fact one of the warmest years on record (Hansen and Lebedeff, 1988). It is believed that this was due to the unprecedented ENSO of 1982.

This is only the most dramatic example of the combination of ENSO and volcanic signals in the climate system. Even if the effects are completely independent physically, they complicate the interpretation of either. Therefore, both modeling and data analysis studies (such as Angell, 1988) are necessary to separate and understand the climate responses to this combination of externally and internally-caused variations.

It has even been suggested that the El Chichón eruption produced the 1982-83 ENSO (*e.g.*, Hirono, 1988). This idea merits further investigation.

6. Recommendations: Volcanoes and Climate

The following projects should receive high priority in future research:

1. **An index of volcanic aerosols should be produced that is related to the climatic effect of volcanoes, not their explosivity.** It would start with the Dust Veil Index and Volcanic Explosivity Index, and then modify them with information obtained from ice cores, radiation measurements, surveys of the sulfur content of volcanic emissions, descriptive reports of eruptions and of optical effects, and modern information from lidars and satellite measurements.

2. **Comprehensive analysis of past temperature, pressure and precipitation data should be performed, searching for the volcanic signal while accounting for the internal climate variations related to ENSO.**

3. **General circulation model simulations of volcanic eruptions should be carried out.** These would produce the signal of a volcanic eruption that

would be compared to that found in the analysis above. Simultaneous forcings by tropical sea surface temperature anomalies and volcanoes should also be used to attempt to understand whether the climatic response is related or independent. The 1982-83 El Chichón-ENSO would be a good case to study. The case of an extremely large eruption should also be studied.

4. **The global aerosol monitoring effort should be continued, with special consideration to allow comprehensive observations of future volcanic eruptions.** A combination of spacecraft, airplanes, balloons and ground-based observations will allow complete coverage of the background aerosol distribution and perturbations due to volcanoes and other causes, such as dust storms and smoke.

REFERENCES

Angell, J. K., 1988: Impact of El Niño on the delineation of tropospheric cooling due to volcanic eruptions. *J. Geophys. Res.*, 93, 3697-3704.

Angell, J. K., and J. Korshover, 1983: Comparison of stratospheric warmings following Agung and El Chichón, *Mon. Wea. Rev.*, 111, 2129-2135.

Angell, J. K., and J. Korshover, 1985: Surface temperature changes following the six major volcanic episodes between 1780 and 1980. *J. Clim. Appl. Meteor.*, 24, 937-951.

Angione, Ronald J. and G. de Vaucouleurs, 1986: Twenty years of atmospheric extinction at McDonald Observatory. *Pub. Astron. Soc. Pac.*, 98, 1201-1207.

Asaturov, M. L., M. I. Budyko, K. Ya. Vinnikov, P. Ya. Groisman, A. S. Kabanov, I. L. Karol, M. P. Kolomeev, Z. I. Pivovarova, E. V. Rozanov, and S. S. Khmelevtsov, 1986: *Volcanics, Stratospheric Aerosol and Earth's Climate.* (Gidrometeoizdat, Leningrad), 256 pp. (in Russian)

Bradley, Raymond S., 1988: The explosive volcanic eruption signal in Northern Hemisphere continental temperature records. *Climatic Change*, 12, 221-243.

Bryson, R. A. and B. M. Goodman, 1980: Volcanic activity and climatic changes. *Science*, 207, 1041-1044.

Ellsaesser, Hugh W., 1983: Isolating the climatogenic effects of volcanoes. UCRL-89161, (Lawrence Livermore National Laboratory, Livermore, CA), 29 pp.

Franklin, B., 1784: Meteorological imaginations and conjectures. *Manchester Literary and Philosophical Society Memoirs and Proceedings*, 2, 122 or 375.

Fujita, Toshio, 1985: The abnormal temperature rises in the lower stratosphere after the 1982 eruptions of the volcano El Chichón, Mexico. *Papers in Meteor. and Geophys.*, 36, No. 2, 47-60.

Gilliland, R. L., 1982: Solar, volcanic and CO_2 forcing of recent climatic changes. *Clim. Change*, 4, 111-131.

Gilliland, R. L. and S. H. Schneider, 1984: Volcanic, CO_2 and solar forcing of Northern and Southern Hemisphere surface temperatures. *Nature*, 310, 38-41.

Graf, Hans-F., 1989: Forced cooling of the polar T21 atmosphere and tropical climate variability. Report No. 45, Max-Planck-Institut für Meteorologie, 25 pp.

Groveman, B. S., and H. E. Landsberg, 1979: Simulated Northern Hemisphere temperature departure 1579-1880. *Geophys. Res. Lett.*, 6, 767-769.

Hameed, Sultan, P.-Y. Zhang, and W.-C. Wang, 1989: Climate in China after the Tambora Eruption of 1815. *CDIAC Communications*, Winter 1989, 6-8.

Hammer, C. U., H. B. Clausen, and W. Dansgaard, 1980: Greenland Ice Sheet

evidence of post-glacial volcanism and its climatic impact. *Nature*, 288, 230-235.

Hansen, J., I. Fung, A. Lacis, S. Lebedeff, D. Rind, R. Ruedy, and G. Russell. 1988: Global climate changes as forecast by the GISS 3-D model. *J. Geophys. Res.*, 93, 9341-9364.

Hansen, James and Sergej Lebedeff, 1988: Global surface air temperatures: update through 1987. *Geophys. Res. Let.*, 15, 323-326.

Hansen, J., G. Russell, A. Lacis, I. Fung and D. Rind, 1985: Climate response times: dependence on climate sensitivity and ocean mixing. *Science*, 229, 857-859.

Hansen, J. E., Wei-Chung Wang and A. A. Lacis, 1978: Mount Agung provides a test of a global climatic perturbation. *Science*, 199, 1065-1068.

Hirono, M., 1988: On the trigger of El Niño Southern Oscillation by the forcing of early El Chichón volcanic aerosols. *J. Geophys. Res.*, 93, in press.

Hofmann, D. J., and J. M. Rosen, 1984a: Balloonborne particle counter observations of the El Chichón aerosol layers in the 0.01 - 1.8 um radius range. *Geofísica Internacional*, 23, 143-153.

Hofmann, D. J., and J. M. Rosen, 1984b: Measurement of the sulfuric acid weight percent in the stratospheric aerosol from the El Chichón eruption. *Geofísica Internacional*, 23, 309-310.

Humphreys, W. J., 1940: *Physics of the Air*. (Dover, New York), 676 pp.

Hunt, B. G., 1977: A simulation of the possible consequences of a volcanic eruption on the general circulation of the atmosphere. *Monthly Weather Review*, 105, 247-260.

Jones, P. D., 1988: Hemispheric surface air temperature variations: recent trends and an update to 1987. *J. Climate*, 1, 654-660.

Keen, R. A., 1983: Volcanic aerosols and lunar eclipses. *Science*, 222, 1011-1013.

Kelly, P. M. and C. B. Sear, 1982: The formulation of Lamb's dust veil index. In *Atmospheric Effects and Potential Climatic Impact of the 1980 Eruptions of Mount St. Helens*, Adarsh Deepak, Ed., NASA Conference Publication 2240, (NASA, Washington, DC), pp. 293-298.

Kelly, P. M. and C. B. Sear, 1984: Climatic impact of explosive volcanic eruptions. *Nature*, 311, 740-743.

Kondo, Junsei, 1988: Volcanic eruptions, cool summers, and famines in the Northeastern part of Japan. *J. Climate*, 1, 775-788.

Kontratyev, K. Ya., 1988: Volcanoes and Climate, WCP-54, WMO/TD - No. 166, (World Meteorological Organization, Geneva), 103 pp.

Labitzke, K., B. Naujokat, and M. P. McCormick, 1983: Temperature effects on the stratosphere of the April 4, 1982 eruption of El Chichón, Mexico.

Geophys. Res. Lett., **10**, 24-26.

Lamb, H. H., 1970: Volcanic dust in the atmosphere; with a chronology and assessment of its meteorological significance. *Phil. Trans. Roy. Soc. London*, **A266**, 424-533.

Lamb, H. H., 1977: Supplementary volcanic dust veil index assessments. *Climate Monitor*, **6**, 57-67.

Lamb, H. H., 1983: Update of the chronology of assessments of the volcanic dust veil index. *Climate Monitor*, **12**, 79-90.

Lough, J. M. and H. C. Fritts, 1987: An assessment of the possible effects of volcanic eruptions on North American climate using tree-ring data, 1602 to 1900 A.D. *Climatic Change*, **10**, 219-239.

Lyons, W. B., P. A. Mayewski, M. J. Spencer and M. S. Twickler, 1988: A Northern Hemisphere volcanic chemistry (1869-1984) and climatic implications using a South Greenland ice core. Submitted to *J. Geophys. Res.*

MacCracken, M. C. and F. M. Luther, 1984: Preliminary estimate of the radiative and climatic effects of the El Chichón eruption. *Geofísica Internacional*, **23**, 385-401.

Manabe, S., and R. J. Stouffer, 1980: Sensitivity of a global climate model to an increase of CO_2 concentration in the atmosphere. *J. Geophys. Res.*, **85**, 5529-5554.

Mass, Clifford F., and David A. Portman, 1989: Major volcanic eruptions and climate: a critical evaluation. *J. Climate*, **2**, 566-593.

Mass, Clifford and Alan Robock, 1982: The short-term influence of the Mount St. Helens volcanic eruption on surface temperature in the Northwest United States. *Mon. Wea. Rev.*, **110**, 614-622.

Mass, C. and S. H. Schneider, 1977: Statistical evidence on the influence of sunspots and volcanic dust on long term temperature trends. *J. Atmos. Sci.*, **34**, 1995-2004.

McCormick, M. Patrick, 1987: SAGE Aerosol Measurements, Volume III-January 1, 1981, to November 18, 1981. NASA Reference Publication 1173. 269 pp.

McCormick, M. Patrick, and M. T. Osborn, 1986: Airborne Lidar Measurements of the El Chichón Stratospheric Aerosols, May 1983. NASA Reference Publication 1172. 87 pp.

McCormick, M. Patrick, and David Brandl, 1986: SAM II Measurements of the Polar Stratospheric Aerosol, Volume VIII-April 1982 to October 1982. NASA Reference Publication 1165. 74 pp.

Mitchell, J. M., Jr., 1961: Recent secular changes of the global temperature. *Ann. N. Y. Acad. Sci.*, **95**, 235-250.

Mitchell, J. M., Jr., 1970: A preliminary evaluation of atmospheric pollution as a cause of the global temperature fluctuation of the past century. In *Global*

Effects of Environmental Pollution, S. F. Singer (ed.), (Reidel, Dordrecht), 139-155.

National Research Council. 1987: *Current Issues in Atmospheric Change.* National Academy Press, Washington, D.C. 39 pp.

Newhall, C. G. and S. Self, 1982: The volcanic explosivity index (VEI) – an estimate of explosive magnitude for historical volcanism. *J. Geophys. Res.*, **87**, 1231-1238.

Oerlemans, Johannes, 1988: Simulation of historic glacier variations with a simple climate-glacier model. *J. Glaciology*, **34**, 333-341.

Oliver, R. C., 1976: On the response of hemispheric mean temperature to stratospheric dust: an empirical approach. *J. Appl. Meteor.*, **15**, 933-950.

Parker, D. E., and J. K. L. Brownscombe, 1983: Stratospheric warming following the El Chichón volcanic eruption, *Nature*, **301**, 406-408.

Pivovarova, Z. I., 1977: Utilization of surface radiation data for studying atmospheric transparency. *Meteorology and Hydrology*, No. 9, 24-31. [in Russian]

Pollack, J. B., O. B. Toon, C. Sagan, A. Summers, B. Baldwin and W. Van Camp, 1976: Volcanic explosions and climatic change: a theoretical assessment. *J. Geophys. Res.*, **81**, 1071-1083.

Porter, Stephen C., 1981: Recent glacier variations and volcanic eruptions. *Nature*, **291**, 139-142.

Quinn, William H., Victor T. Neal, and Santiago E. Antunez de Mayolo, 1987: El Niño occurrences over the past four and a half centuries. *J. Geophys. Res.*, **92**, 14449-14461.

Quiroz, R. S., 1983: The isolation of stratospheric temperature change due to the El Chichón volcanic eruption from nonvolcanic signals. *J. Geophys. Res.*, **88**, 6773-6780.

Quiroz, R. S., 1984: Influence of the El Chichón eruption on the stratosphere after summer 1982. *Geofísica Internacional*, **23**, 363-372.

Ramanathan, V., 1988: The greenhouse theory of climate change: a test by an inadvertant global experiment. *Science*, **240**, 293-299.

Rampino, M. R. and S. Self, 1982: Historic eruptions of Tambora (1815), Krakatoa (1883) and Agung (1963), their stratospheric aerosols and climatic impact. *Quat. Res.*, **18**, 127-163.

Rampino, M. R. and S. Self, 1984: Sulphur-rich volcanic eruptions and stratospheric aerosols. *Nature*, **310**, 677-679.

Robock, Alan, 1978: Internally and externally caused climate change. *J. Atmos. Sci.*, **35**, 1111-1122.

Robock, Alan, 1979: The "Little Ice Age": Northern Hemisphere average observations and model calculations. *Science*, **206**, 1402-1404.

Robock, Alan, 1981a: A latitudinally dependent volcanic dust veil index, and its effect on climate simulations. *J. Volcanol. Geotherm. Res.*, 11, 67-80.

Robock, Alan, 1981b: The Mount St. Helens volcanic eruption of 18 May 1980: minimal climatic effect. *Science*, 212, 1383-1384.

Robock, Alan, 1983a: The dust cloud of the century. *Nature*, 301, 373-374.

Robock, Alan, 1983b: Ice and snow feedbacks and the latitudinal and seasonal distribution of climate sensitivity. *J. Atmos. Sci.*, 40, 986-997.

Robock, Alan, 1984a: Climate model simulations of the effects of the El Chichón eruption. *Geofísica Internacional*, 23, 403-414.

Robock, Alan, 1984b: Snow and ice feedbacks prolong effects of nuclear winter. *Nature*, 310, 667-670.

Robock, Alan, 1984c: Review of *Volcano Weather*. *Nat. Wea. Dig.*, 9, 5-6.

Robock, Alan, 1985: Detection of volcanic, CO_2 and ENSO signals in surface air temperature. *Adv. Space Res.*, 5, No. 6, 53-56.

Robock, Alan, 1990: The volcanic contribution to climate change of the past 100 years. in *Greenhouse-Gas-Induced Climatic Change: A Critical Appraisal of Simulations and Observations*, Michael E. Schlesinger, Ed., (Elsevier), in press.

Robock, Alan and Clifford Mass, 1982: The Mount St. Helens volcanic eruption of 18 May 1980: large short-term surface temperature effects. *Science*, 216, 628-630.

Schneider, S. H. and C. Mass, 1975: Volcanic dust, sunspots and temperature trends. *Science*, 190, 741-746.

Schönwiese, Christian-D., 1988: Volcanic activity parameters and volcanism-climate relationships within recent centuries. *Atmósfera*, 1, 141-156.

Sear, C. B., P. M. Kelly, P. D. Jones and C. M. Goodess, 1987: Global surface-temperature responses to major volcanic eruptions. *Nature*, 330, 365-367.

Sedlacek, W. A., E. J. Mroz, A.,L. Lazrus, and B. W. Gandrud, 1983: A decade of stratospheric sulfate measurements compared with observations of volcanic eruptions. *J. Geophys. Res.*, 88, 3741-3776.

Self, Stephen and Michael R. Rampino, 1988: The relationship between volcanic eruptions and climate change: still a conundrum? *Eos*, 69, 74-75, 85-86.

Self, S., M. R. Rampino and J. J. Barbera, 1981: The possible effects of large 19th and 20th century volcanic eruptions on zonal and hemispheric surface temperatures. *J. Volcanol. Geotherm. Res.*, 11, 41-60.

Sigurdsson, Haraldur and Steven Carey, ·1988: The far reach of Tambora. *Natural History*, No. 6, 66-73.

Simkin, T., L. Siebert, L. McClelland, D. Bridge, C. Newhall and J. H. Latter, 1981: *Volcanoes of the World*. (Hutchinson Ross, Stroudsburg, PA), 240 pp.

Stommel, Henry, and Elizabeth Stommel, 1983: *Volcano Weather*. (Seven Seas Press, Newport, RI), 177 pp.

Stothers, Richard B., 1984: The great Tambora eruption in 1815 and its aftermath. *Science*, **224**, 1191-1198.

Taylor, B. L., Tsvi Gal-Chen and S. H. Schneider, 1980: Volcanic eruptions and long-term temperature records: an empirical search for cause and effect. *Q. J. Roy. Meteor. Soc.*, **106**, 175-199.

Toon, Owen B., 1982: Volcanoes and climate. In *Atmospheric Effects and Potential Climatic Impact of the 1980 Eruptions of Mount St. Helens*, Adarsh Deepak, Ed., NASA Conference Publication 2240, (NASA, Washington, DC), pp. 15-36.

Toon, Owen B. and James B. Pollack, 1980: Atmospheric aerosols and climate. *American Scientist*, **68**, 268-278.

Vogelmann, A. M., A. Robock and R. G. Ellingson, 1988: Effects of dirty snow in nuclear winter simulations. *J. Geophys. Res.*, **93**, 5319-5332.

Vupputuri, R. K. R. and J. P. Blanchet, 1984: The possible effects of El Chichón eruption on atmospheric thermal and chemical structure and surface climate. *Geofísica Internacional*, **23**, 433-447.

Wendler, G. and Y. Kodama, 1986: Some remarks on the stratospheric temperature for Fairbanks, Alaska, after the El Chichón eruption. *Atmosphere-Ocean*, **24**, 346-352.

Xu, Qun, 1985: The assessment of volcanic dust veil index of Northern temperate zone in the recent 100 years. *Acta Geophysica Sinica*, **28**, 558-568.

Xu, Qun, 1988: The abnormally cold summers of central China and their relation to volcanic eruptions. in *Aerosols and Climate*, Peter Hobbs and M. Patrick McCormick, Eds., (Deepak, Hampton, VA), 223-232.

Yamamoto, Ryozaburo, Tatsuya Iwashima, and Makoto Hoshiai, 1975: Change of the surface air temperature averaged over the Northern Hemisphere and large volcanic eruptions during the year 1951-1972. *J. Meteor. Soc. Japan*, **53**, 482-486.

THE STUDY OF ATMOSPHERIC VOLCANIC EMISSIONS
by
J.P. Friend, Drexel University

It is an oddity that in examination of climate records the two most readily identifiable causes of 1 - 2 year fluctuations in global surface temperatures are volcanic eruptions and El Nino Southern Oscillations. Yet there has been little regard paid to volcanoes in the face of impending climate change due to increases in the atmospheric abundances of greenhouse gases. Another oddity connected with volcanoes is that society's interest in them waxes with the occurrence of a large explosive eruption in which hundreds or thousands of lives are lost and rapidly wanes afterwards. This behavior, perhaps all too human, leaves peoples and their governments in a state of perpetual unpreparedness to deal with the cataclysms which sooner or later occur. It is understandable that society will not support costs to remain prepared for years at a time while other problems and programs clamor for financial resources. Understanding of volcanic phenomena and the associated emissions of chemical substances that effect the environment and climate will permit rational planning with regard to societal concerns and will likely provide scientists with important clues about the nature of climate changes.

Science has long recognized that volcanic emissions of volatile substances have played a vital role in determining the composition of the earth's crust, the oceans and the atmosphere. Furthermore, their role continues to have such influence. Much of the influence is being exerted in extensive submarine releases of magma which occur out of sight and in regions not readily accessible for scientific study. Consequently, atmospheric emissions represent the best subjects for scientific scrutiny of volatile volcanic materials, the processes that affect them and their effect on the surface environment.

The key quantities of interest in the studies of volcanic emissions are the fluxes of individual components. Sulfur compounds are most important because they are the most abundant of the reactive substances in volcanic plumes and it is through their oxidation to sulfate particulate material that it may influence climate by changing the global albedo directly or through the formation of clouds. Berresheim and Jaeschke,1983 estimated the volcanic flux of sulfur compounds to have a lower limit of 12 Tg/yr. The NATO Workshop (Galloway et al, 1985) assessment estimated a factor of 2 uncertainty in the total global natural S-emissions of 80 Tg/yr. Thus it can be appreciated that in years with large explosive eruptions, the volcanic contribution to the total can be a substantial fraction of the total.

Friend, 1989 reviewed the global fluxes of chlorine and fluorine and suggests that emission from volcanoes could comprise the major part of atmospheric F flux. Symonds et al, 1985 estimated 0.06 - 6.0 Tg/yr. If the true flux is greater than about 1 Tg/yr, volcanic sources would dominate over sea salt aerosols, blowing soil dust and anthropogenic sources. By contrast, volcanoes contribute relatively little to the global fluxes of chlorine; sea salt aerosols being the dominant source. Nonetheless, Cl emissions from large explosive volcanoes may be of significance in perturbing stratospheric ozone chemistry.

The studies performed in the early 1980's by the group of scientists assembled in Project RAVE (Research on Atmospheric Volcanic Emissions) in combining airborne sampling of volcanic plumes with ground-based studies of the same materials provided insights concerning the gaseous and trace element compositions of volcanic plumes and their emission rates. Since only a few plumes (5) could be studied, the combined results of RAVE and all other volcanic flux studies do not provide the basis for making accurate estimates of the global fluxes of volcanic materials through the atmosphere. (See Galloway et al, 1985.) The experience gained by the RAVE investigators makes it possible to consider

undertaking a new study of volcanic emissions aimed toward the ends outlined above. It is suggested to assemble a group of potential principle investigators to consider writing a proposal for a such a study involving aircraft sampling of plumes of active volcanoes and sub-proposals for individual participation in the program. The investigators would be atmospheric chemists and physicists who have established airborne measurement capabilities for the important volcanic effluents and volcanologists who are experts in the phenomenology and chemistry of active volcanoes.

List of substances which could be studied.

Sulfur compounds: SO_2, H_2S, CS_2, OCS, particulate SO_4^{2-}.
Halogen compounds: HCl, HF, particulate Cl^- and F^-.
Nitrogen compounds: NO, NH_3, particulate NO_3^- and NH_4^+.
Trace elements: Particulate As, Sb, Se, Al, Fe, Ca, and others.
Radioactive elements: Rn and its progeny.

Measurements to permit flux estimates and spatial distribution of material in plumes: SO_2 column density (COSPEC), particulate vertical profile (LIDAR), inertial navigational derived parameters of position and wind speed.

REFERENCES

Friend, J.P., Natural chlorine and fluorine in the atmosphere, water, and precipitation, WMO, Global Ozone Research and Monitoring Project - Report No. 20: "Scientific Assessment of Stratospheric Ozone: 1989", Vol. II, Appendix: AFEAS Report, 1989.

Galloway, J.N., R.J. Charlson, M.O. Andreae and H. Rodhe, eds., "The Biogeochemical Cycling of Sulfur and Nitrogen in the Remote Atmosphere", NATO ASI Series, Series C: Mathematical and Physical Sciences Vol. 159, D. Reidel Publishing Co., Boston, 1985.

Symonds, R.B., W.I. Rose and M.H. Reed, Contribution of Cl- and F-bearing gases to the atmosphere by volcanoes, *Nature, 344*, 415-418, 1988.

EOS AND AEROSOLS

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PARAMETERS

1.) τ (optical depth)

   o spatial variability
   o vertical structure
   o temporal variability

2.) δ (asymmetry factor)

3.) ΔA_p (impact on planetary albedo)

$$A_p = \frac{1}{S_o} \int_{\cdot}^{\cdot} I_R(\mu)\, d\mu$$

APPROACHES

Type	EOS Instrument	Predecessor
1. Limb scanning	SAGE II	SAM, SAGE
2. Nadir imaging	MODIS	AVHRR
3. Polarization	EOSP	Venus polar.
4. Multiangle imaging	MISR	(none)
5. Laser sounding	LAWS	(none)

COMBINATION APPROACH

    o vertical structure: SAGE, LAWS

    o horizontal structure: MISR, LAWS, MODIS

    o total optical depth: MISR, MODIS, LAWS, EOSP

    o size, δ: MISR, SAGE, EOSP

    o albedo impact: MISR, MODIS

                other related measurements:

                      o cloud discrimination (HIRIS)
                      o albedo, fluxes (ERB)

O Polarization

    + very sensitive to aerosol properties

    -- very sensitive to everything (land surface)
    -- rather poor spatial resolution

O Laser sounding

    + direct measurement of backscatter vs. height

    -- uncertain conversion of backscatter to τ
    -- untested

O Multiangle imaging

    + global coverage of τ at high resolution
    + separates aerosol and surface reflectivity
    + direct measurement of $I_R(\mu)$

    -- very limited vertical resolution
    -- untested

O Limb scanning

    + direct measure of τ
    + indirect measure of size ($\tau(\lambda)$)
    + direct measure of $\Delta\tau(z)$

    -- long path-integrated measurement (~200 km)
    -- limited tropospheric information
    -- possible confusion with cirrus

O Nadir imaging

+ global coverage of τ at high resolution

-- inferred τ subject to error due to
  o ground reflectivity
  o cloud contamination

Appendix H

PARTICIPANTS

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Appendix I

WORKSHOP ON
VOLCANO CLIMATE INTERACTIONS
CENTER OF ADULT EDUCATION, UNIVERSITY OF MARYLAND
COLLEGE PARK, MARYLAND

18 - 19 NOVEMBER 1990

AGENDA

DAY 1 - Room No. 0105

08:00	Registration	
09:00	Welcoming Remarks	M. Baltuck
09:10	Agenda and Objectives	L. Walter
09:30	*Coffee Break*	
09:45	**VOLCANO PALEOCLIMATE**	M. Rampino
10:15	Informal Presentations/Discussion	
11:45	*Lunch*	
12:30	**PETROLOGY/TECTONICS**	H. Sigurdsson
13:00	Informal Presentations/Discussion	
14:30	*Break*	
14:45	**CLOUD DISSIPATION**	S. Thompson
15:15	Informal Presentations/Discussion	
18:30	*Reception (Chesapeake Room)*	
19:15	*Dinner*	
20:00	General Discussion	

DAY 2 - Room No. 0105

08:30	**VOLCANO MICROPHYSICS/ ATMOSPHERIC CHEMISTRY**	R. Turco
09:00	Informal Presentations Discussion	
10:30	*Coffee Break*	
10:45	**RADIATION/CLIMATE MODELLING**	A. Robock
11:15	Informal Presentations/Discussion	
12:45	*Lunch*	
13:30	**REVIEW, SUMMARY, AND PLANS**	All
15:30	Adjourn	

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<tr><th colspan="3">NNSN
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15. Supplementary Notes

Louis S. Walter--NASA/Goddard Space Flight Center, Greenbelt, Maryland;
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16. Abstract
An interdisciplinary workshop was held in June 1990 to assess the status of knowledge in, and to develop plans for research on the effects of volcanic eruptions on climate. Historically, very large eruptions have had tangible climatic effects. The effects of lesser eruptions in recent decades and in the future must be understood in order to be able to spot small, early effects of anthropomorphically induced climate change and to verify or adjust climate models.

The range of disciplines in the study of volcanism-climate interactions includes paleoclimate, volcanology, petrology, tectonics, cloud physics and chemistry, and climate and radiation modeling. Questions encountered in understanding the interactions include: 1) the source and evolution of sulfur and sulfur-gaseous species in magmas; 2) their entrainment in volcanic plumes and injection into the stratosphere; 3) their dissipation rates; and 4) their radiative effects. Other issues include modeling and measuring regional and global effects of such large, dense clouds.

The Workshop defined a broad-range plan of research designed to answer these questions. The plan includes observations of volcanoes, rocks, trees and ice cores, as well as satellite and aircraft observations of erupting volcanoes and resulting plumes and clouds.

17. Key Words (Suggested by Author(s)) Volcanism, Climate, Volcanoes, Geology, Climate Change, Paleoclimate Aerosols, Radiation, Stratosphere, Models, Research Plan, Workshop	18. Distribution Statement Unclassified - Unlimited Subject Category 46

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