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7

Emissions from volcanoes

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1. GLOBAL VOLCANISM

Around 380 volcanoes were active during the last century, with around 50 volcanoes active per year (Andres and Kasgnoc 1998). Volcanic activity is not randomly distributed over the Earth, but is linked to the active zones of plate tectonics, as shown in figure 1. More than 2/3 of the world's volcanoes are located in the northern hemisphere, and in tropical regions. The emission of volcanic gases depends on the thermodynamic conditions (pressure, temperature) and on the magma type (i.e., its chemical composition, which in turn depends on the tectonic environment).

Globally, most of the magma mass erupted is of basaltic composition. Basaltic volcanoes erupt most frequently along mid-oceanic ridges in deep ocean water, where magma rises along the oceanic plate boundaries. On very rare locations, these volcanoes erupt into the atmosphere (called subaerial eruptions), such as in Iceland and the Azores, where the eruption rate is so large that volcanic islands have been formed along the mid-Atlantic ridge. Their basaltic magmas are primitive melts from the Earth's mantle. Basaltic magma is rich in magnesium and iron, and poor in silicate. In general, this magma type is characterized by low viscosity and low gas content, and eruptions are mostly effusive. They consist of a high portion of CO_2 and sulfur in the gas fraction. Long lasting basaltic lava streams can cover large areas (e.g., the Deccan traps in India, Yellowstone in North America, and the Laki fissure in Iceland). Basaltic magmas are also typical for intra-plate volcanoes that evolve over very hot regions in the Earth's mantle at around 100 km depth. The island chain of Hawaii is an example of this volcano type.

Basaltic magma contributes only a minor fraction to the volcanic sulfur emissions into the atmosphere and only in rare cases reaches the stratosphere.

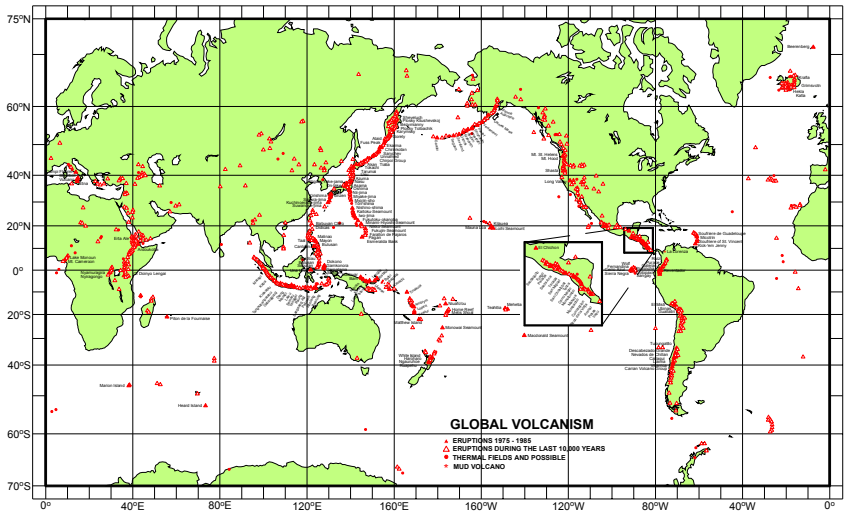

Figure 1: Active volcanoes from 1975-1985 (solid triangles) and sites with volcanic activity during the last 10,000 years (open triangles). Taken from Graf et al. (1997), based on McClelland et al. (1989), adapted by permission of Prentice Hall.

Felsic magma stems from differentiation processes (i.e., chemical alteration) in the magma chamber, or from melting of earth crust material. This highly differentiated magma is rich in silicate and alkali. It contains a higher content of dissolved gases, especially water. It is highly viscous and eruptions are generally more explosive. In the petrological nomenclature, magma of this type is called rhyolite, dacite, or phonolite, depending on the specific chemical composition. Magmas of intermediate silicate content are andesitic magmas, and are typical for volcanoes at convergent plate boundaries. Subduction zone volcanoes develop in regions where the continental plate overrides the oceanic plate, such as in the Andes. When oceanic plates converge, island arc volcanoes evolve (e.g., Indonesia). Felsic and andesitic volcanoes erupt less frequently than basaltic volcanoes. They can release high amounts of magma and energy on short time scales, however, sometimes injecting ashes and gases directly into the stratosphere. In addition, many permanently emit gases during non-explosive phases. Subduction zone volcanoes contribute the largest part to the total global volcanic sulfur emission.

2. CHEMICAL SPECIES EMITTED BY VOLCANOES

The composition of volcanic gases at the volcanic vent is in general controlled by the equilibrium between a hydrous fluid (exsolved gas) at the top and the silicate melt in the magma chamber below (Symonds et al. 1994). It varies widely between volcanoes depending on the magma type, and is also dependent on the individual volcano's state of activity. An overview is given in table 1.

Table 1: Characteristic composition of volcanic gases at the vent (e.g., Symonds et al. 1988, Cadle 1980, Symonds et al. 1994, and Chin and Davis 1993, see also table 2).

Species	H_2O	CO_2	SO_2	H_2S	COS	CS_2	HCl	HBr	HF
%/vol	50-90	1-40	1-25	1-10	10^{-4}-10^{-2}	10^{-4}-10^{-2}	1-10	?	$< 10^{-3}$
Tg/year	?	75	1.5-50	1-2.8	0.006-0.1	0.007-0.096	0.4-11	0.0078-0.1	0.06-6

Water vapor (H_2O) is the most prevalent volcanic gas, contributing between 50 and 90% by volume, however the contribution to the global H_2O inventory is negligible in comparison to the atmospheric concentration. The second important volcanic gas is carbon dioxide (CO_2), which ranges from 1 to 40% by volume. Volcanic emissions contribute less than 1% to the total global CO_2 emission (Cadle 1980, Gerlach 1991). Anthropogenic annual CO_2 emissions are by a factor of 100 higher than total natural degassing of the Earth (Schmincke 1993).

Sulfur gases contribute typically 2 to 35%/vol of volcanic gas emissions. They are the most relevant species concerning the climatic impact of volcanic events. The dominant sulfur component is sulfur dioxide (SO_2), with yearly emissions ranging from 1.5 to 50 Tg SO_2. These data are discussed in more detail below in section 5. The second important S-species is hydrogen sulfide (H_2S), which is only observed by direct sampling. It is often co-emitted and subsequently converted to SO_2 in the atmosphere (e.g., Bluth et al. 1995). The H_2S fraction increases with increasing pressure (i.e., depth of the magma chamber) and with decreasing temperature and oxygen concentration of the magma (Gerlach et al. 1986). Stoiber et al. (1987) have estimated that the amount of H_2S and SO_4^{2-} is commonly less than 1%/vol, although it may reach 10% in some cases. Berresheim and Jaeschke (1983) estimated total sulfur emissions (H_2S plus SO_4^{2-}) to be about 4.2 Tg S per year, but these figures are highly variable and uncertain. In the case of the El Chichón eruption, possibly most of the ~3.5 Tg S was emitted as H_2S. Considering all emitted sulfur as SO_2 will not lead to a big error, however, since H_2S oxidizes to SO_2 within a few days. Other sulfur species add minor portions. Carbonyl sulfide (COS) and its precursor carbon disulfide (CS_2)

contribute a small fraction of 10^{-4} to 10^{-2}%/vol. However, COS has a residence time of several years in the atmosphere (e.g., Kjellström et al. 1998). Because of its high stability it is an important source for the sulfate aerosol layer in the stratosphere. Volcanoes contribute less than 1% to the total global atmospheric COS emission (Cadle 1980, Andres and Kasgnoc 1998). The data suggest that volcanoes contribute insignificant amounts of COS and CS_2 to the atmosphere, but the exact quantity has not yet been determined.

The main halogen component of volcanic emissions is hydrogen chloride (HCl), with 1-10 %/vol (Symonds et al. 1988). The volcanic contribution of 0.4-11 Tg HCl per year (Symonds et al. 1988, Cadle 1980) to the total chlorine budget is approximately equal to the anthropogenic emissions, but the emissions from oceans are orders of magnitudes higher. HCl is highly soluble and is therefore rapidly washed out of the atmosphere. Hence, small eruptions and silent degassing will not be of importance for atmospheric composition. Hydrogen bromide (HBr) contributes a small proportion in the range of 10^{-6} parts per volume. Quantification of global annual emissions needs to be improved, in particular because of its enormous ozone depletion potential, which is higher than that of chlorine (e.g., McElroy et al. 1992). Estimates for HBr range from 0.0078 Tg to 0.1 Tg per year (Cadle 1980, Bureau et al. 2000). Its fate in the atmosphere is similar to that of HCl. Only under very specific conditions can its lifetime be significant, as discussed in section 4.

Very little information is available on hydrogen fluoride (HF). Its fraction in volcanic gas emissions is less than 1 ppm; the annual global emission is 0.06-6 Tg (Symonds et al. 1988). HF is not of importance in general, but during specific events (e.g., Laki 1783, Mt. Hudson 1990) HF emissions may be extreme and lead to severe environmental contamination with hazards to plants and livestock.

A variety of other gaseous components is erupted by volcanoes. Volcanic emissions can be detrimental for the local environment. However, this paper focuses on impacts on the atmosphere on larger scales in time and space.

3. OBSERVATIONAL TECHNIQUES

The determination of gas and particle concentrations in a plume is extremely difficult because of the cloud's opaqueness and the inherent risks of directly observing and sampling the volcanic cloud. Volcanic emissions can be studied remotely by airborne and ground-based instruments and by satellite observations.

Among the most abundant volcanic gases, SO_2 is the species that has been mainly measured remotely, because its concentration in a plume exceeds atmospheric background concentration. Ultraviolet (Correlation Spectrometer, COSPEC) and infrared (Fourier Transform Infrared Spectrometer, FTIR) spectroscopy have been employed (e.g., Symonds et al. 1988, Andres and Rose 1995). These techniques are most important for the detection of injections into the troposphere.

In recent years, the launches of new satellites and new developments in remote-sensing techniques have expanded the capability to monitor volcanoes from space (Rose et al. 2000). Satellite observations (e.g., by TOMS (Total Ozone Mapping Spectrometer), AVHRR (Advanced Very High Resolution Radiometer), GOME (Global Ozone Monitoring Experiment), SCIAMACHY (SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY)) for SO_2 and ash particles are only useful for strong sources (Bluth et al. 1993). SO_2 is the only volcanic gas so far monitored operationally by satellite. Since the first TOMS data in 1979, which could only measure the presence of SO_2 from larger eruptions, improved instruments and retrieval algorithms today can detect SO_2 gas from smaller eruptions and from the passive degassing of some volcanoes if the detection limit of 5-20 kt SO_2 is exceeded. Satellites have even detected SO_2 from several eruptions not known from ground observations.

SO_2 emission rates cannot be measured directly, but have to be inferred from a multitude of other data. The errors vary with the volcanic plume mass and area, and they depend on the quality of the estimation of the plume's trajectory and dilution. Since these instruments only measure the SO_2 abundance in a vertical (satellites) and/or horizontal/slant (COSPEC and FTIR) path, the flux must be estimated using information on the wind field. This is not always available with the necessary accuracy. In addition, the results are disturbed by meteorological clouds and affected by the high optical depth of the plume and the reflectivity of the underlying surface. For example, the error of the total SO_2 mass retrieved from TOMS data ranges from 15 to 30% (Krueger et al. 1995). In addition, most of the techniques mentioned above work only during the day.

The petrologic method is employed to estimate magmatic gas content by comparing pre- and post-eruptive volatile concentrations trapped in ash deposits. One compares the gas content of matrix glass inclusions (representative of the primitive magma) to that in the degassed magma. This technique works better for sulfur for volcanoes erupting along spreading ridges, such as the Icelandic volcanoes, where the erupted material comes

directly from primitive magma, as compared to volcanoes along subduction zones, such as El Chichón or Pinatubo, where there is a relatively high concentration in the atmospheric plumes but little sulfur in the inclusions, which come from reprocessed material. Palais and Sigurdsson (1989) compared petrological data to ice core estimates of volatile emissions for several major eruptions. They found that the atmospheric yield of sulfur, chlorine and fluorine is not only dependent on total erupted mass, but largely determined by the composition of the erupting magmas. The "excess sulfur" found in satellite data after Pinatubo was explained by a separate SO_2-rich fluid phase which coexists with the magma (Gerlach et al. 1996). This means that gases can dissolve from un-erupted magma, and form a separate fluid in the magma chamber which is emitted co-eruptive, or by fuming (non-steady degassing during phases of higher activity, but not highly explosive) and fumarole (steady degassing) activity.

The enormous differences in the observational data are due to uncertainties in each individual measuring technique, but they also result from the fact that the plume is investigated at different distances from the crater and during different states of volcanic activity or of the eruption. Changes in sulfur emissions at (and between) single sources over orders of magnitude can take place depending on the state of activity (e.g., Augustine or Etna) and magma type. Only a few of the about 560 potential sources have ever been measured, and only a handful of these were observed more than episodically. For important regions (e.g., Kamchatka) there are no published data at all. In some cases (e.g., Etna), the emission at the single volcano can reach mean values of highly polluted industrial areas (Langmann and Graf 1997, Graf et al. 1998).

Seasonal and annual variations of stratospheric aerosols have been observed by satellite instruments (Hitchman et al. 1994), by balloon-borne particle counters (Hofmann et al. 1983), and by lidars (Menzies and Tratt 1995). Each of these techniques, however, has problems. The Stratospheric Aerosol and Gas Experiment (SAGE) and Stratospheric Aerosol Measurement (SAM) projects (McCormick et al. 1979, Mauldin et al. 1985, McCormick 1987, Thomason 1991, Veiga 1993) have provided more than 20 years of three-dimensional data of stratospheric aerosol spectral extinctions, the longest such record. Hitchman et al. (1994) and Stevermer et al. (2000) used these data to study the zonal mean aerosol climatology. However, a significant level of uncertainty exists in aerosol characterization during the period of SAGE observations from 1978 to present. For example, because of the "saturation" effect, SAGE II fails to measure aerosols in a large part of the most important equatorial region for almost a year after the Mt. Pinatubo eruption. The eruption of El Chichón in 1982 was not covered by SAGE ob-

servations because the SAGE I instrument failed in 1981, and SAGE II was only launched in 1984. Therefore, to produce a data set of stratospheric aerosols, SAGE observations would have to be enhanced and blended with other available satellite and ground-based measurements. For example, Antuña et al. (2002) used lidar data to fill gaps in SAGE II observations after the Mt. Pinatubo eruption. After Pinatubo, 29 balloon flights carrying aerosol counting instruments were made at Laramie, Wyoming (41.3ºN, 105.6ºW) by Deshler et al. (1992, 1993), but there were no observations from other latitudes. Lidars give excellent vertical distribution of aerosols, but until recently they could only operate under clear sky conditions and during the night. The distribution of lidar observatories is uneven, with none between 19°N and 23°S, with the exception of the one in Bandung, Indonesia (6.9°S, 107.6°E), which is plagued by bad weather. There are at least three lidar networks worldwide, one in Asia (Uchino et al. 1992) and two in Europe (Fiocco et al. 1996, Bösenberg et al. 1998), but none of these operates using standardized instruments and processing software. A data assimilation system, incorporating all these different observations in the context of a model of stratospheric dynamics is needed to produce a uniform aerosol data set for use in climate studies.

4. PROCESSES IN THE ERUPTION COLUMN

Little is known about the processes in volcanic plumes that determine the amount and specification of emissions into the atmosphere due to the difficulties with observations described in the previous section. Many eruptions are accompanied by a water cloud. Hydrometeors and ash particles are able to scavenge volcanic emissions (e.g, Turco et al. 1983). Emissions are altered by chemical reactions if the plume is not too opaque to prevent photochemical activity (Textor et al. 2002). For explosive eruptions, the time for the rise from the crater to the stratosphere is less than 10 min. Hence, the residence time in the plume is comparatively much too short for chemical transformations of the volcanic gases to take place before the reaching the stratosphere.

Sulfur species (SO_2 and H_2S) are only slightly soluble in liquid water; hence, they are only slightly removed by cloud and rain drops at lower heights. HCl, on the other hand, is highly soluble in liquid water. Volcanic gases can also be scavenged by frozen hydrometeors via direct gas incorporation during diffusional growth of ice as proposed by Textor et al. (2002, 2003). The mechanism of gas trapping in ice within a volcanic plume is supported by observational evidence of eruptions, where a lot of water was present in the plume due to interaction with sea water. For example, in one observation

extremely high amounts of ice were accompanied by unusually low SO_2 concentrations (Rose et al. 1995).

The scavenging of volcanic gases in a plume has been investigated through numerical simulations by Tabazadeh et al. (1993), who used a one-dimensional plume model (Wilson 1976; Woods 1988, 1993) without ice microphysics, and by Textor et al. (2002, 2003), who worked with a more complex model (Oberhuber et al. 1998, Herzog et al. 1998) including ice microphysics and ash aggregation. The results of the two studies are in general accordance, however, the latter authors showed that scavenging efficiency is determined by the amount of condensed water or ice, which in turn depends on the volcanic conditions (composition of the magma, strength of the eruption) and on the meteorological conditions (stability of the atmosphere, wind shear) in the ambient atmosphere (Graf et al. 1999). These simulations showed that a large portion of gaseous SO_2 and H_2S reaches the umbrella region of the plume. The scavenging efficiency for HCl is much higher, but it might not be completely removed from the gas phase under dry conditions (Textor et al. 2002, 2003). Furthermore, the simulations showed that hydrometeors containing dissolved volcanic gases can reach the stratosphere where sublimation is possible, releasing them into the stratosphere.

5. VOLCANIC SULFUR EMISSIONS INTO THE TROPOSPHERE

The total amount of volcanic tropospheric sulfur emissions per year was estimated by several authors (table 2). There is a large range in estimates, from 0.75 Tg (Kellogg et al. 1972) to 25 Tg (Lambert et al. 1988) sulfur per year. Lambert et al. (1988) have employed the atmospheric concentration of the polonium isotope ^{210}Po to estimate the volcanic output of SO_2. The other authors used emission data from monitored volcanoes to extrapolate to global volcanic emissions. The contribution of silent degassing to volcanic sulfur input into the troposphere is still under discussion, the numbers vary between 1% (Andres and Kasgnoc 1998) and 95% (Berresheim and Jaeschke 1983). This great uncertainty is due to the poor database, the suspicious consideration of silent degassing, and to different extrapolation techniques to include non-monitored volcanoes. Graf et al. (1997) estimated 14 ± 6 Tg S per year, which is in the upper level among other recent estimates. Halmer et al. (2002) have performed a comprehensive literature survey on volcanic activity of the last century. Based on volcanological parameters and on the volcanic SO_2 index (VSI) (see section 8), the information about 50 monitored

volcanoes was extrapolated to include 310 unmonitored active subaerial volcanoes.

Table 2: Annual global volcanic sulfur emission fluxes into the atmosphere by different authors.

Authors	S (in 10^{12} g/yr)
Kellogg et al. (1972) excluded silent degassing (0.5% gas content)	0.75
Bartels (1972)	17.0
Friend (1973)	2.0
Stoiber and Jepsen (1973) based on only 5 central and south American volcanoes	5.0
Cadle (1975) excluded silent degassing, but used higher magma gas content (2.5%)	3.8
Naughton et al. (1976)	23.5
Granat et al. (1976)	3.0
Le Guern (1982)	5.0
Berresheim and Jaeschke (1983) 95% from silent, 5% from explosive, incl. $H_2S + SO_4$	7.6
Stoiber et al. (1987) 35 volcanoes, short time (1 year) 35% silent, 65% explosive	9.3
Lambert et al. (1988) uses ^{210}Po (50% of all from volcanic sources), but only for very few volcanoes $^{210}Po/SO_2$ ratio	25.0
Andres and Krasgnoc (1998) mainly explosive, only 1% silent	10.4
Graf et al. (1997) includes silent (fumaroles (5±2) post and extra-eruptive) (fuming (5±2) pre- and intra-eruption) and explosive (4±2 Mt S), incl. $H_2S + SO_4$	14.0 ± 6
Halmer et al. (2002)	9.0 ± 1.5

Volcanoes contribute about 36% to the tropospheric sulfur burden (Graf et al. 1997). The proportion of H_2S in sulfur emissions is still under discussion. However, H_2S is quickly oxidized to SO_2 in about two days in the troposphere (e.g., Seinfeld and Pandis 1998). SO_2 in the atmosphere is transferred to sulfate by chemical reactions – promoted by the presence of water drops – within some days in the troposphere. Although a variety of SO_2 oxidation rates have been determined, an agreement on the correct rate has still not been found (Bluth et al. 1992, Facchini et al. 1992, Rani et al. 1992,

Fung et al. 1991, Grgi et al. 1991, Hansen et al. 1991, Joos and Baltensperger 1991, Gallagher et al. 1990, Oppenheimer et al. 1998). The difficulties are due to various conditions that volcanic SO_2 encounters in the atmosphere. Sulfate, which is highly water soluble, is removed from the troposphere within less than a week by wet and dry deposition, but in dry conditions can remain longer, especially if it reaches higher tropospheric levels.

Sulfate aerosols in the atmosphere have effects altering the Earth's radiation balance and cloud microphysics, discussed in section 7. In addition, tropospheric sulfate aerosols have an impact on atmospheric chemistry. Acid rain, caused by scavenging of sulfate aerosols and damaging the vegetation, has been a problem for many years.

6. VOLCANIC EMISSIONS INTO THE STRATOSPHERE

Volcanic injections of sulfur into the stratosphere are sporadic and unpredictable. Explosive volcanic eruptions reach the stratosphere in general at least once every two years (Simkin et al. 1993). Toba 71-73 kyr ago is the largest eruption believed to have occurred since the beginning of the Quaternary period, ca. 2 million years ago. Rose and Chesner (1990) analyzed the deposits and with simple assumptions estimated that Toba released about 6000 Tg of SO_2 into the atmosphere, but Zielinski et al. (1995, 1996) used ice core data to estimate a release of 1000-2000 Tg SO_2 and Scaillet et al. (1998) using experimental petrology methods estimated only about 70 Tg SO_2 (Oppenheimer 2002).

Volcanic sulfur is not only directly injected into the stratosphere from explosive eruptions, but also from emissions of continuously degassing non-eruptive volcanoes and from small eruptive events (Graf et al. 1998). Volcanoes substantially contribute to the stratospheric sulfur burden (e.g., Bluth et al. 1997). The average input of volcanic sulfur over the last 200 years has been estimated as 1 Tg per year ranging from 0.3-3 Tg per year (Pyle et al. 1996). A minimum flux of 0.5-1.0 Tg S for the past 9000 years has been derived from ice core sulfate data. This mean flux, however, is highly variable due to singular explosive events. During periods of little volcanic activity, the background stratospheric load of sulfate is in the order of 0.15 Tg S (Kent and McCormick 1984). In case of cataclysmic volcanic eruptions, this stratospheric sulfate mass can be increased for short periods of time by one to two orders of magnitude.

Table 3: The largest volcanic eruptions of the last 250 years. The Volcanic Explosivity Index (VEI) is a measure of explosivity (Newhall and Self 1982, see section 8). Opacity is normalized to the Krakatau eruption and derived from geological data. The SO_2 data marked with a * are derived from the petrologic method, which gives a lower limit of the possible emission (see section 8). Table modified from Robock (2000).

Volcano	Year	VEI	Opacity	SO_2 [Tg]
Laki fissure, Iceland	1783	4	2.3	100*
Tambora, Indonesia	1815	7	3.0	130*
Cosiguina, Nicaragua	1835	5	4.0	
Askja, Iceland	1875	5	1.0	
Krakatau, Indonesia	1883	6	1.0	32*
Tarawera, New Zealand	1886	5	0.8	
Santa Maria, Guatemala	1902	6	0.6	13*
Ksudach, Kamchatka	1907	5	0.5	
Katmai, Alaska, USA	1912	6	0.5	12*
Agung, Indonesia	1963	4	0.8	5-13*
St. Helens, USA	1980	5	0.5	1
El Chichón, Mexico	1982	5	0.8	7
Pinatubo, Philippines	1991	6	1.0	17

Volcanic sulfur is emitted in the form of SO_2 and H_2S. H_2S is oxidized to SO_2 in the stratosphere, and H_2S has a chemical lifetime of only about three days (McKeen et al. 1984). From stoichiometric considerations it can be assumed that during oxidation one molecule of H_2O per H_2S is produced. This could be an important source for water in the stratosphere after volcanic eruptions. The effect of H_2S oxidation on the HO_x balance has been discussed by McKeen et al. (1984) and is still not known. SO_2 is transferred

to sulfate by chemical reactions with an e-folding time of approximately 35 days in the dry stratosphere (Bluth et al. 1992). It is assumed that no net gain or loss of HO_x results from SO_2 oxidation (Read et al. 1993, McKeen et al. 1984). Two-dimensional model simulations of the Toba eruption (Bekki et al. 1996) showed that the injection of 200 Mt SO_2 could dehydrate the stratosphere, because approximately three molecules of H_2O are needed to convert SO_2 to sulfate, strongly reducing the HO_x-concentration.

Sulfate has residence times of a few years in the stratosphere. New particles are formed due to binary homogenous nucleation of sulfuric acid and water vapor. After the eruption of Mt. Pinatubo, 98% of the observed stratospheric aerosol was volatile (Deshler et al. 1992, Sheridan et al. 1992), which indicates that homogeneous nucleation is the most important process for stratospheric aerosol formation in the disturbed atmosphere. Deshler et al. (1993) found an increase of 1-2 orders of magnitude in the concentration of condensation nuclei in the volcanic layers. The aerosol size distribution is shifted to greater radii due to condensation of the vapor on existing aerosol particles. Model calculations that successfully reproduced Pinatubo sulfate aerosol (Weisenstein et al. 1997, Timmreck and Graf 2000) were based on homogeneous nucleation.

Observations (Russell et al. 1996) and model studies (Zhao et al. 1995, Stenchikov et al. 1998, Timmreck and Graf 2000) show that it takes about three months to build up the sulfate peak after big volcanic injections of SO_2 in the stratosphere. A volcanically enhanced stratospheric aerosol layer can be observed for about four years. Measurements of peak backscatter and mass, and columnar quantities (mass, backscatter and optical depth) indicate that the volcanic aerosol is removed with an e-folding time of approximately 1±0.2 years (Jäger et al.1995, Ansmann et al. 1997, Deshler et al. 1997). The decay rates of vertical integrated quantities are strongly influenced by variations in the tropopause height and stratosphere-troposphere exchange processes, while the decay of peak parameters is smoother and primarily reflects gravitational removal by sedimentation. The relatively faster sedimentation of larger particles from the stratosphere leads to a rapid decrease in mass, but not in surface area (Ansmann et al. 1998). The e-folding time of surface area concentration is about 20 to 30% larger (16-17 months) than the decay of the aerosol backscatter and mass, since the ratio of surface area to mass increases with decreasing particle size. Similar decay rates have been observed after the eruption of Mt. Pinatubo and El Chichón (Rosen et al. 1994, Barnes and Hoffmann 1997). Chazette et al. (1995), however, found for the eruption of El Chichón a 25% slower decay of integrated backscatter in the lower stratosphere than for Pinatubo. This was possibly caused by the lofting of the particles to higher altitudes.

7. ATMOSPHERIC EFFECTS OF VOLCANIC EMISSIONS

Sulfate aerosols in the atmosphere have radiative effects that alter the Earth's radiation balance (e.g., Franklin 1784; Charlson et al. 1991, 1992). While their optical depth is higher in visible wavelengths and the net effect in general is cooling, sulfate aerosols also absorb and emit in the longwave, heating the layer where they are and increasing the downward flux of radiation at the surface (for a review see Robock 2000). The scattering of incoming solar radiation (direct effect) leads to cooling at the surface.

Sulfate aerosols in the troposphere act as cloud condensation nuclei and modify the radiative properties and the lifetime of clouds (indirect, or Twomey effect) (Twomey 1974). The increase of the number of cloud droplets due to an increased number of condensation nuclei increases the albedo of clouds, enhancing surface cooling. It may also lead to changes in precipitation rate or rain suppression in deep convective clouds, thus changing the spatial and temporal distribution of latent heat release. This could have a significant effect on the global circulation, as was shown for aerosols in general by Nober et al. (2002).

Volcanic sulfur emissions in the troposphere have a disproportionate effect on the atmosphere, as shown by numerical experiments with an atmospheric general circulation model including a simplified sulfur cycle (Graf et al. 1997). Active volcanoes generally reach considerable elevations and most of their emissions, even during non-explosive events, are injected into the free troposphere, well above the planetary boundary layer. At this height, removal processes are slower and volcanic sulfur has longer atmospheric residence times than anthropogenic sulfur emitted from low elevations. In the mean, volcanic sulfur dominates the sulfate concentration in the middle and upper troposphere while anthropogenic emissions control the boundary layer (Graf et al. 1998). Table 4 shows the global sulfur budget and the radiative effect of different sulfur sources taken from Graf et al. (1997), who used a particular climate model for their calculations. Volcanoes contribute about 4.5 times less to the global sulfur emissions than anthropogenic emissions. But the atmospheric SO_2 burden is only 1.3 times less, and the the global tropospheric sulfate burden is of the similar size as the anthropogenic one. Hence, due to the longer life time of their sulfur emissions, volcanoes have a much higher relative radiative effect on global climate. Since the treatment of sulfur species, especially their vertical transport and cloud interactions, is not

very realistic in current climate models, these results may be highly model-dependent. This is especially true for the transfer of troposphere sulfur to the stratosphere, which is discussed next.

Table 4: Global annual mean sulfur budget in Tg sulfur per year (from Graf et al. 1997) and top-of-the-atmosphere (TOA) radiative forcing in percentage of the total (101.8 Tg S/yr emission, 0.52 Tg S SO_2 burden, 0.78 Tg S SO_4 burden, -0.65 W/m^2 total direct global forcing). The efficiency is the relative sulfate burden divided by the relative source strength (i.e., column 4/column 2).

Source	Sulfur emission [%]	SO_2 burden [%]	SO_4^{2-} burden [%]	Efficiency	Direct TOA forcing [%]
Anthropogenic	65.6	46.1	37.1	0.56	40
Biomass Burning	2.5	1.2	1.6	0.64	2
Dimethyl sulfide (DMS), mainly from oceans	18.2	17.8	25.3	1.39	26
Volcanoes	13.7	34.9	36.0	2.63	33

The injection of large quantities of gases (SO_2, H_2O, H_2S, CO_2, HCl) and volcanic ash (mainly silicate particles) into the stratosphere can have an important impact on the global climate. In recent years, the understanding of the global volcanism-climate system has improved significantly, as the very detailed summaries by Robock (2000) and Zielinski (2000) show. But there is still more to be learned about the factors controlling the chemical, physical and optical properties of the volcanic aerosol, its radiative forcing and its role in the global climate system. A schematic representation of the atmospheric effects of volcanic eruptions is shown in figure 2.

Fine volcanic ash injected into the stratosphere is characterized by grain sizes in the micrometer range. They are efficiently removed by sedimentation from the stratosphere within about a month after the eruption (e.g., Pinto et al. 1989) and therefore have only a limited local impact on the climate system.

Sulfur-containing volcanic gases, rather than ash, are responsible for the climatic effects of explosive volcanic eruptions. Sulfate aerosols in the stratosphere have radiative effects altering the Earth's radiation balance (Stenchikov et al. 1998). The scattering of incoming solar radiation (direct effect) leads to cooling at the surface and counteracts the greenhouse effect. The additional aerosol loading increases the temperature in the lower stratosphere through the absorption of near infrared and long wave radiation. According to the 1995 IPCC report (Schimel et al. 1996), the decadal mean

radiative forcing due to volcanic aerosols has varied by as much as 1.5 W/m^2 since 1850, which can be large compared to the decadal-scale variation in any other known forcing. Volcanoes are therefore an important part of the global climate system that substantially contribute to the observed natural variability.

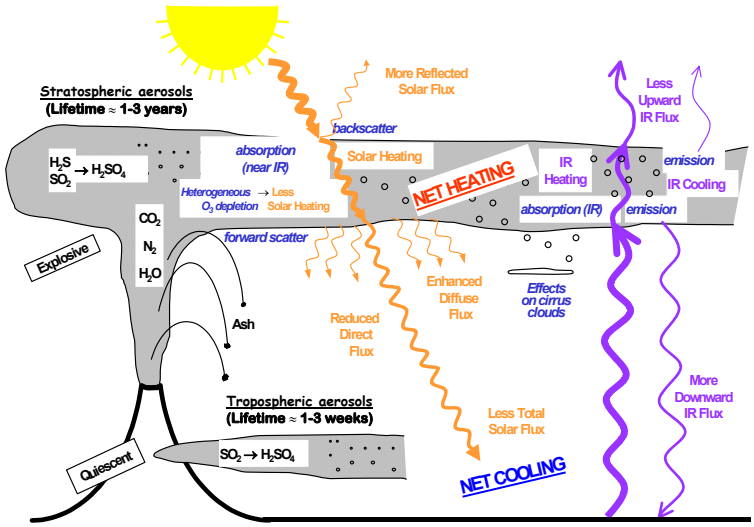

Figure 2: Schematic diagram of volcanic inputs to the atmosphere and their effects. Reproduced with permission from Alan Robock, Volcanic eruptions and climate, Reviews of Geophysics, 38, 2, 191-219, 2000. Copyright 2000 American Geophysical Union.

Stratospheric warming due to the presence of the volcanic aerosol in the stratosphere increases the pole-to-equator temperature gradient (e.g., Quiroz 1984, Parker and Brownscombe 1983, Angell 1997a). This has a dynamical feedback on the tropospheric circulation, leading to abnormally warm winters over the northern hemisphere continents in years following the eruption (Groisman 1985, Robock and Mao 1992). The mechanism, including the production of stronger westerly winds in the lower stratosphere and their effect on tropospheric planetary waves was explained first by Graf et al. (1993, 1994) and Kodera (1996) to work via exaggeration of the North Atlantic Oscillation.

The effect of the geographical location of an explosive volcanic eruption on its climatic impact has not yet been quantified. Tropical volcanoes have the capability to affect the global climate system, because the cloud can be

spread into both hemispheres. Mid to high latitude eruptions will primarily affect their own hemisphere (Graf and Timmreck 2001). The dispersal of the volcanic cloud is also strongly dependent on the time of the year, and the eruption height. The location of the intertropical convergence zone and the phase of the Quasi Biennial Oscillation (QBO) play an important role for the transport of volcanic clouds.

The eruption of halogen species at the crater is rather significant (e.g., Varekamp et al. 1984, Westrich et al. 1992, Bureau et al. 2000). Their direct injection by explosive volcanic eruptions into the stratosphere could lead to catastrophic ozone loss (Prather 1992). No severe increase of halogens was observed in the stratosphere after the eruption of Mt. Pinatubo (e.g., Mankin et al. 1992, Wallace and Livingston 1992), however, after the eruption of El Chichón in 1982 a clear increased chlorine concentration was detected (Mankin and Coffey 1984, Woods et al. 1985). The fraction of direct injection of halogen compounds depends on the magma composition and on the processes in the eruption column, as discussed in section 4.

Volcanic hydrated sulfate aerosols in the stratosphere can serve as sites for heterogeneous reactions which convert passive chlorine compounds (HCl, HOCl, $ClNO_3$) into active ones (ClO, Cl) (chlorine activation). After the Pinatubo eruption, the surface layer of the stratospheric sulfate aerosol was about 300 times higher than the usual value with peak concentrations of $>3x10^{-7}$ cm^2/cm^3 (Jäger et al. 1995, Thomason et al. 1997). The critical value for ozone destruction of 10^{-7} cm^2/cm^3 was reached (Jäger et al. 1995, Ansmann et al. 1993) for more than one year in northern hemisphere midlatitudes. Heterogeneous reactions deplete ozone in the presence of halogens like chlorine or bromine (e.g., Michelangeli et al. 1989, Hofmann and Solomon 1989, Granier and Brasseur 1992, Solomon et al. 1996). Since the human-induced increase of chlorine concentration in the stratosphere has peaked, the effect of ozone destruction at volcanic aerosol will probably decrease in the next few decades.

Column ozone reduction after the Mt. Pinatubo eruption ranged from about 2% in the tropics to about 7% in mid latitudes (Angell 1997b, Solomon et al. 1998). Very large ozone losses were observed in high northern latitudes in February and in March. Randel et al (1995) found losses of 10% in total column ozone in 1992 northward of 60°N and of 10-12% in 1993. Ozonesonde profiles show that the gas did not decrease uniformly at all altitudes. Hofmann et al. (1993) found at 20°N an ozone decrease below 20-24 km and an ozone increase about 26 km. Vertical profiles from the tropics (Grant et al. 1994) showed 10% losses of ozone below the peak and roughly 6% increases compared with SAGE data.

The observed ozone changes are a combined effect of heterogeneous chemistry and of perturbations in the heating rates and in the photolysis rates. The chemical composition of the atmosphere during the post-Pinatubo period has been analyzed in several publications (e.g., Solomon et al. 1998, Toohey et al. 1995). Model studies, in particular for the Pinatubo episode, were carried out to investigate the impact of the volcanic aerosol contribution to stratospheric trace gas concentrations (e.g., Pitari and Rizzi 1993, Bekki and Pyle 1994, Kinnison et al. 1994, Solomon et al. 1996, Rosenfeld et al. 1997). A systematic analysis of the contribution of several components to stratospheric ozone destruction after the Pinatubo eruption was carried out by Tie et al. (1994) with a 2-dimensional radiative chemical model, which suggests that in the first year after the eruption the radiative-dynamical effects and in the second year the chemical effects were dominant. Rosenfeld et al (1997) found with a 2-d interactive radiative-dynamical-chemical model, that 1-2% of the ozone depletion in the low latitudes after the Pinatubo eruption was due to the altered circulation (perturbation in the heating rate), and about 0.5% due to heterogeneous chemistry and perturbation in the photolysis rate each. However, global model studies considering the various interactions between aerosol microphysics, radiation, chemistry and dynamics, which are necessary to completely analyze the climate impact of a volcanic eruption, are still missing.

In the tropics and during tropopause folds in the midlatitudes, volcanic sulfate aerosol particles can be transported vertically across the tropopause. Important lidar data by Ansmann et al. (1993) showed that in more than 50% of the observations the stratospheric aerosol layer penetrated the tropopause and influenced the formation and maintenance of cirrus clouds in the upper troposphere. Graf et al. (1997) suggested the importance of volcanic sulfate aerosol in the upper troposphere for cirrus formation. Unusual high cloud particle number concentration (600 L^{-1}), and extremely supercooled drops at 223-223 K have been observed in the year after the Mt. Pinatubo eruption (Sassen 1992, Sassen et al. 1995). Song et al. (1996) suggested that the interannual variability of global high level clouds is related to explosive volcanism. The amount and persistence of such clouds increased by up to 10% after the eruptions of El Chichón and Pinatubo mainly in midlatitudes. These anomalies lasted for several years. Thus, violent volcanic eruptions lead to a change in radiative properties of cirrus clouds. Their impact on the climate is still not known, as it depends on changes in cloud microphysics. Scattering of solar radiation would lead to enhanced cooling, while absorption of terrestrial radiation would lead to warming.

8. VOLCANIC ACTIVITY INDICES

Multiple efforts have been made to compile a quantitative record of climatic impact of historical volcanic eruptions. When studying the effects of volcanic eruptions on climate, it is important to separate the volcanic forcing from the climatic response. Proxy records of past climate, such as tree rings and corals, measure climate change, not volcanism. Only if one already understands the climatic response, can these records be used to indicate past volcanism. Therefore, these records will be discussed in the next section as data to validate theories of the impact of eruptions on climate. Robock and Free (1995, 1996) described indices of past volcanism in detail and compare them, and Robock (2000) summarized their work. Here we even more briefly describe these indices. A perfect index would convey the radiative forcing associated with each explosive eruption. The radiative forcing is most directly related to the sulfur content of emissions that reach into the stratosphere, and not to the explosivity of the eruption. For all the indices, the problem of missing volcanoes and their associated dust veils becomes increasingly important the farther back in time they go. Even today, some indices may miss some southern hemisphere eruptions, as they may not be reported. Even in the 1980s, the December 1981 aerosols from the eruption of Nyamuragira were observed with lidar but were reported as the "mystery cloud" for several years until the source was identified by reexamining the TOMS satellite record (Krueger et al. 1996). The problem of missing an eruption does not exist for ice core records, except that associating acidity or sulfate peaks with particular eruptions may not be possible if the particular eruption is not known.

The first compilation of past volcanic eruptions, the dust veil index (DVI), was made by Lamb (1970, 1977, 1983). The methods used to create the DVI include historical reports of eruptions, optical phenomena, radiation measurements (for the period 1883 onward), temperature information, and estimates of the volume of ejecta. While the DVI has been criticized because of the circular reasoning used by including reports of temperature variations, for only a few eruptions between 1763 and 1882 was the northern hemisphere averaged DVI calculated based solely on temperature information. Robock (1981a) created a modified version of Lamb's DVI which excluded temperature information, and found that this had minor impacts on climate model simulations.

A second compilation is the volcanic explosivity index (VEI) (Newhall and Self 1982). As the VEI is based only on volcanological data (Simkin et al. 1981, Simkin and Siebert 1994), and included no atmospheric data, it cannot be used to estimate the height of the eruption columns. "Stratospheric injec-

tion" was the least reliable of 11 criteria for estimating VEI, and was never intended as a description of the eruption which had a VEI assigned from more reliable evidence. Therefore, "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." (Newhall and Self 1982). An example of a recent eruption for which VEI would be misleading, is the 1980 Mt. St. Helens eruption, with a large VEI of 5, but little stratosphere injection of sulfur and virtually no climatic impact (Robock 1981b).

Despite its weakness, the VEI provides the most comprehensive geological record of past volcanism. Schnetzler et al. (1997) developed a volcanic SO_2 index (VSI) based on the VEI and 15-year satellite samplings of volcanic eruptions. These authors distinguished between subduction zone volcanoes and others, but they excluded silent degassing like fuming, fumarolic activity and dissipative emissions at the volcano flanks. Halmer et al. (2002) extended Schnetzler et al.'s work by including more published observations of sulfur emissions and reports of volcanic activity, also including silent degassing. Observed activity reports were compared with available sulfur measurements. This information was projected to include unmonitored active sub-aerial volcanoes. Halmer et al.'s modified VSI shows the contribution of different volcano types to the injection of sulfur into the atmosphere at different altitudes. Basaltic magmas can be important local sources, but only subduction zone volcanoes have the potential to significantly directly contribute to the stratospheric sulfur budget, because of their higher explosivities and source altitudes. In spite of the careful data review, the range of order of magnitudes of SO_2 emission "typical" for a specific geological and ambient setting shows big uncertainties. These do not only result from too sporadic and inaccurate measurements, but also to a large degree from the large variability of the processes involved.

A zonally averaged time series of the optical depth due to volcanic eruptions from 1850 on was compiled by Sato et al. (1993), and updated by Stothers et al. (1996). This record is based on optical data, since 1979 satellite measurements have been included. Based on the Sato et al. data and on radiative forcing calculations for the Pinatubo aerosol, Stenchikov et al. (1998) and Andronova et al. (1999) provided a data set of the instantaneous and adjusted radiative forcing by volcanic aerosol from 1850-1994.

Acidity in ice cores from the deposition of volcanic sulfate on glacial ice in the years following an eruption has been used to construct a record of past volcanism. However, each ice core record is extremely noisy and may have other problems (Robock and Free 1995). For example, smaller local eruptions may have the same signal as distant larger eruptions, so estimating the

global stratospheric aerosol loading from high latitude ice cores is problematic. Hammer et al. (1980) produced a 10,000 year data set using the Crete and Camp Century ice cores from Greenland. Crowley et al. (1993) reanalyzed the Crete ice core data and constructed a 1420 year record of volcanic activity. The GISP2 ice core has been used to create a 2100 year estimate of stratospheric loading and optical depth, and a 110,000 record of explosive volcanism (Zielinski et al. 1995, 1996). Using multiple ices from both northern hemisphere ice sheets in Greenland and Ellesmere Island, and from Antarctica in the southern hemisphere, Robock and Free (1995) constructed a new Ice core Volcanic Index (IVI). Robock and Free (1996) extended this analysis back to 435 A.D., but the farther back one goes, the fewer ice core records are available. More cores are being drilled and analyzed as time goes on, and they will provide valuable new information. The IVI, which correlates well with the VEI but to a lesser extent with the DVI, was used in energy balance model simulations of the past six centuries (Crowley and Kim 1999, Free and Robock 1999).

Recently a new ice core volcanic index, the Volcanic Aerosol Index VAI, was introduced by Robertson et al. (2001). It combines historical observations, ice core data from both hemispheres and satellite data to obtain an estimation of the stratospheric optical depth for the past 500 years. The VAI is latitude dependent, with a variable zonal band width of the stratospheric volcanic aerosol.

Proxy data suggest an increase in the frequency of volcanic eruptions on secular time scales. However, the number of highly explosive eruptions shows no such trend and the increase seems to be related to historical events and the number of observers.

9. CONCLUDING REMARKS

Volcanic emissions are highly variable in space and time. To better quantify the impacts of volcanic eruptions on climate, there is a need to better constrain their sulfur emissions. Important future investigations that are needed include exemplary long-term measurements at the sources, better satellite observations, and improved volcanic plume models that simulate the transformation rates of SO_2 into SO_4^{2-} in warm moist plumes and the contribution of H_2S, SO_4^{2-} and SO_2 associated with plume particles, since these species are not measured by the conventional methods. The fate of volcanic aerosols in the stratosphere and their influence on chemistry, microphysics and dynamics are still under discussion.

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11. REFERENCES

Andres, R. J., and W. I. Rose, Remote sensing spectroscopy of volcanic plumes and clouds, in Monitoring Active Volcanoes: Strategies, Procedures and Techniques, edited by B. McGuire, C. Kilburn, and J. Murray, pp. 301-314, UCL Press Limited, London, 1995.

Andres, R. J., and A. D. Kasgnoc, A time-averaged inventory of subaerial volcanic sulfur emissions, Journal of Geophysical Research, 103, 25.251-25.261, 1998.

Andronova, N. G., E. Rozanov, F. Yang, M. E. Schlesinger, and G. L. Stenchikov, Radiative forcing by volcanic aerosols from 1850 through 1994, Journal of Geophysical Research, 104, 16807-16826, 1999.

Angell, J. K., Stratospheric warming due to Agung, El Chichón and Pinatubo taking into account the quasi-biennial oscillation, Journal of Geophysical Research, 102, 9479948, 1997a.

Angell, J. K., Estimated impact of Agung, El Chichón and Pinatubo volcanic eruptions on global and regional total ozone after adjustment for the QBO, Geophysical Research Letters, 24, 647-650,1997b.

Ansmann, A., U. Wandinger, and C. Weitkamp, One-year observations of Mount Pinatubo aerosol with an advanced Raman lidar over Germany at 53.5°N, Geophysical Research Letters, 20(8), 711-714, April 23, 1993.

Ansmann, A., I. Mattis, U. Wandinger, F. Wagner, J. Reichardt, and T. Deshler, Evolution of the Pinatubo Aerosol: Raman lidar observations of particle optical depth, effective radius, mass, and surface area over central Europe at 53.4°N, Journal of Atmospheric Sciences, 54, 2630-2641, 1997.

Ansmann, A., I. Mattis , H. Jäger, and U. Wandinger, Stratospheric Aerosol Monitoring With Lidar: Conventional Backscatter versus Raman Lidar Observations of Mount- Pinatubo, Contributions to Atmospheric Physics. 213-222, 1998.

Antuña, J. C., A. Robock, G. L. Stenchikov, L. W. Thomason, and J. E. Barnes, Lidar validation of SAGE II aerosol measurements after the 1991 Mount Pinatubo eruption. Journal of Geophysical Research, 107 (D14), 2002.

Barnes, J.E., and D. J.Hofmann, Lidar measurements of stratospheric aerosol over Mauna Loa Observatory, Geophysical Research Letters, 24, 1923-1926, 1997.

Bartels, O. G., An estimate of volcanic contributions to the atmosphere and volcanic gases and sublimates as the source of the radio isotopes ^{10}Be, ^{35}S, and ^{22}Na, Health Physics, 22, 387-392, 1972.

Bekki, S., and J. A. Pyle, A two-dimensional modeling study of the volcanic eruption of Mount Pinatubo, Journal of Geophysical Research, 99, 18,861-18,869, 1994.

Bekki, S., J. A. Pyle, W. Zhong, R. Toumi, J. D. Haigh und D. M. Pyle, The role of microphysical and chemical processes in prolonging the climate forcing of the Toba eruption, Geophysical Research Letters, 23, 2669-2672, 1996.

Berresheim, H., and W. Jaeschke, The contribution of volcanoes to the global atmospheric sulfur budget, Journal of Geophysical Research, 88, 3732-3740, 1983.

Bluth, G. J. S. , Doiron, S. D., Schnetzler, C. C., Krueger, A. J., and L. S. Walter, Global tracking of the SO_2 clouds from the June, 1991 Mount Pinatubo eruptions, Geophysical Research Letters, 19, 151-154, 1992.

Bluth, G. J. S., C. C. Schnetzler, A. J. Krueger, and L. S. Walter, The contribution of explosive volcanism to global atmospheric sulphur dioxide concentrations, Nature, 366, 327-329, 1993.

Bluth, G. J. S., C. J. Scott, I. E. Sprod, C. C. Schnetzler, A. J. Krueger, and L. S. Walter, Explosive SO_2 emissions from the 1992 eruptions of Mount Spurr, Alaska. U.S. Geological Survey Bulletin, 2139, 37-45, 1995.

Bluth, G. J. S., W. I. Rose, I. E. Sprod, and A. J. Krueger, Stratospheric loading of sulfur from explosive volcanic eruptions, Journal of Geology, 105, 671-684, 1997.

Bösenberg, J., M. Alpers, C. Böckmann, H. Jäger, V. Mathias, T. Trickl, U. Wandinger, and M. Wiegner, A lidar network for the establishment of an aerosol climatology, Proceedings of the 19th International Laser Radar Conference, (Annapolis, July 6-10), NASA/CP-1998-207671/PT1, 23-24,1998.

Bureau H., H. Keppler, and N. Metrich, Volcanic degassing of bromine and iodine: experimental fluid/melt partitioning data and applications to stratospheric chemistry, Earth and Planetary Science Letters, 183 (1-2): 51-60, 2000.

Cadle, R. D., Volcanic emissions of halides and sulfur compounds to the troposphere and stratosphere, Journal of Geophysical Research, 80, 1650-1652, 1975.

Cadle, R. D., A comparison of volcanic with other fluxes of atmospheric trace gas constituents, Reviews of Geophysics and Space Physics, 18, 746-752, 1980.

Charlson, R. J., J. Langner, H. Rodhe, C. B. Leovy, and S. G. Warren, Perturbation of the Northern Hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols, Tellus, 43, 152-163, 1991.

Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A., Coakley, Jr., J. E. Hansen, and D. J. Hoffman, Climate forcing by anthropogenic aerosols, Science, 255, 423-430, 1992.

Chazette, P. C. David, J. Lefrere, S. Godin, J. Pelon, and G. Megie, Comparative lidar study of the optical, geometrical and dynamical properties of stratospheric postvolcanic aerosols, following the eruptions of El Chichón and Mount Pinatubo, Journal of Geophysical Research, 101, 23195-23207,1995.

Chin M., and D. D. Davis, Global sources and sinks of OCS and CS_2 and their distributions, Global Biogeochemical Cycles 7 (2), 321-337, 1993.

Crowley, T. J., T. A. Criste, and N. R. Smith, Reassessment of Crete (Greenland) ice core acidity/volcanism link to climate change, Geophysical Research Letters, 20, 209-212, 1993.

Crowley, T. J., and K.-Y. Kim, Modeling the temperature response to forced climate change over the last six centuries, Geophysical Research Letters, 26, 1901-1904, 1999.
Deshler, T., D. J. Hofmann, B. J. Johnson, and W. R. Rozier, Balloonborne measurements of the Pinatubo aerosol size distribution and volatility at Laramie, Wyoming during the summer of 1991, Geophysical Research Letters, 19, 199-202, 1992

Deshler, T., B. J. Johnson, and W. R. Rozier, Balloonborne measurements of Pinatubo aerosol during 1991 and 1992 at 41°N: vertical profiles, size distribution, and volatility, Geophysical Research Letters, 20, 1435-1438,1993.

Deshler, T., G. B. Liley, G. Bodeker, W. A. Matthews, and D. J. Hofmann, Stratospheric aerosol following Pinatubo, comparison of the north and south mid latitudes using in situ measurements, Advances Space Research, 20, 2057-2061, 1997.

Facchini, M. C., S. Fuzzi, M. Kessel, W. Wobrock, W. Jaeschke, B. G. Arends, J. J. Möls, A. Berner, I. Solly, C. Kruisz, G. Reischl, S. Pahl, A. Hallberg, J. A. Ogren, H. Fierlinger-Oberlinninger, A. Marzorati, and D. Schell, The chemistry of sulfur and nitrogen species in a fog system: A multiphase approach, Tellus, 44B, 505-521, 1992.

Fiocco, G., M. Cacciani, A. G. di Sarra, D. Fuà, P. Colagrande, G. De Benedetti, P. Di Girolano, and R. Viola, The evolution of the Pinatubo stratospheric aerosol layer observed by lidar at South Pole, Rome, Thule: A summary of results, in: The Mount Pinatubo Eruption: Effects on The Atmosphere and Climate, NATO ASI Series, 42, 17-32, 1996.

Franklin, B., Meteorological imaginations and conjectures, Manchester Literary and Philosophical Society Memoirs and Proceedings 2, 122, 1784, reprinted in Weatherwise 35, 262, 1982.

Free, M., and Robock, A.: Global warming in the context of the Little Ice Age, Journal of Geophysical Research, 104, 19,057-19,070, 1999.

Friend, J. P., The global sulfur cycle, in Chemistry of the Lower Atmosphere, edited by S. I. Rasool, 177-201, Plenum, New York, 1973.

Fung, C. S., P. K. Misra, R. Bloxam, and S. Wong, A numerical experiment on the relative importance of H_2O_2 and O_3 in aqueous conversion of SO_2 to SO_4^{2-} Atmospheric Environment, 25A, 411-423, 1991.

Gallagher, M. W., R. M. Downer, , T. W. Choularton, M. J. Gay, I. Stromberg, C. S. Mill, M/ Radojevic, B. J. Tyler, B. J. Bandy, S.A. Penkett, T. J. Davies, G. J. Dollard, and B. M. R. Jones, Case studies of the oxidation of sulphur dioxide in a hill cap cloud using ground and aircraft based measurements. Journal of Geophysical Research, 95, 18517-18537, 1990.

Gerlach, T.M., and Casadevall, T.J., Fumarole emissions at Mount St. Helens Volcano, June 1980 to October 1981: Degassing of a magma-hydrothermal system: Journal of Volcanology and Geothermal Research, 28, 141-160, 1986.

Gerlach, T.M., Present-day CO_2 emissions from volcanoes: EOS, 72, 249, 254-255, 1991.

Gerlach, T. M., H. R. Westrich, and R. B. Symonds, Preeruption vapor in magma of the climactic Mount Pinatubo eruption: Source of the giant stratospheric sulfur dioxide cloud, in Newhall, C. G., and R. S. Punongbayan, eds., Fire and mud: Eruptions and lahars of Mount Pinatubo, Philippines: Philippine Institute of Volcanology and Seismology, Quezon City, and University of Washington Press, Seattle, 415-433, 1996.

Graf, H-F., I. Kirchner, A. Robock, and I. Schult, Pinatubo eruption winter climate effects: Model versus observations, Climate Dynamics, 9, 81-93, 1993.

Graf, H.-F., J. Perlwitz, and I. Kirchner, Northern Hemisphere tropospheric mid-latitude circulation after violent volcanic eruptions Contributions to Atmospheric Physics [Beitrage zur Physik der Atmosphäre.], 67(1), 3-13, 1994.

Graf, H.-F., J. Feichter, and B. Langmann, Volcanic sulfur emissions: Estimates of source strength and its contribution to the global sulfate distribution, Journal of Geophysical Research, 102, 10727-10738, 1997.

Graf, H.-F., B. Langmann, and J. Feichter, The contribution of Earth degassing to the atmospheric sulfur budget, Chemical Geology 147, 131-145, 1998.

Graf, H.-F., M. Herzog, J. M. Oberhuber, and C. Textor, The effect of environmental conditions on volcanic plume rise, Journal of Geophysical Research, 104(D20) , 24,309 ,1999.

Graf, H.-F., and C. Timmreck, A general climate model simulation of the aerosol radiative effects of the Laacher See eruption (10,900 BC), Journal of Geophysical Research 106, 14,747- 14,756, 2001.

Granat, L., H. Rodhe, and R. O. Hallberg, The global sulphur cycle, in Nitrogen, Phosphorous and Sulphur-Global Cycles, edited by B. H. Svensson, and R. Söderlund, SCOPE Report 7, Ecological Bulletin, 22, 89-134, 1976.

Grant, W. B., E. V. Browell, J. Fishman. V. G. Brackett. R. E. Veiga, D. Nganga. A. Minga, B. Cros. C. F. Butler. M. A. Fenn, C. S. Long, and L. L Stowe, Aerosol-associated changes in tropical stratospheric ozone following the eruption of Mount Pinatubo, Journal of Geophysical Research, 99, 8197-8211, 1994.

Granier, C., and G. Brasseur, Impact of heterogeneous chemistry on model predictions of ozone changes, Journal of Geophysical Research, 97, 18015-18033, 1992.

Grgi, I., Hudnik, V., Bizjak, M., and J. Levec, Aqueous S(IV) oxidation-I. Catalytic effects of some metal ions, Atmospheric Environment, 25A, 1591-1597, 1991.

Groisman, P. Y., Regional climate consequences of volcanic eruptions (in Russian), Meteorol. Hydrol., 4, 39-45, 1985.

Halmer, M. M., H.-U. Schmincke, and H.-F. Graf , The annual volcanic gas input into the atmosphere, in particular into the stratosphere: a global data set for the past 100 years, Journal of Volcanology and Geothermal Research, 115, 511-528, 2002.

Hammer, C.U., H.B. Clausen, and W. Dansgaard, Greenland ice sheet evidence of post-glacial volcanism and its climate impact, Nature, 288, 230-235,1980.

Hansen, A. D. A., W. H. Benner, and T. Novakov, Sulfur dioxide oxidation in laboratory clouds, Atmospheric Environment, 25A, 2521-2530, 1991.

Herzog, M., H.-F. Graf, C. Textor, and J. M. Oberhuber, The effect of phase changes of water on the development of volcanic plumes, Journal of Volcanology and Geothermal Research, 87, 55-74, 1998.

Hitchman, M. H., M. McKay, and C. R. Trepte, A climatology of stratospheric aerosol, Journal of Geophysical Research, 99, 20,689-20,700, 1994.

Hofmann, D. J., J. M. Rosen, R. Reiter, and H. Jager, Lidar- and balloon-borne particle counter comparisons following recent volcanic eruptions, Journal of Geophysical Research, 88, 3777-3782, 1983.

Hofmann, D. J., and S. Solomon, Ozone destruction through heterogeneous chemistry following the eruption of El Chichón, Journal of Geophysical Research, 94 , 5029-5041, 1989.

Hofmann, D. J., S.J. Oltmans, J.M. Harris, W.D. Komhyr, J.A. Lathrop, T. DeFoor, and D. Kuniyuki, Ozonesonde measurements at Hilo, Hawaii following the eruption of Pinatubo, Geophysical Research Letters, 20, 1555-1558, 1993.

Jäger, H. Uchino, O. Nagai, T. Fujimoto, T. Freudenthaler, and V. Homburg, F., Ground-based remote sensing of the decay of the Pinatubo eruption cloud at three Northern Hemisphere sites Geophysical Research Letters, 22(5), 607-610, 1995.

Joos, F., and U. Baltensperger, A field study on chemistry, S(IV) oxidation rates and vertical transport during fog conditions. Atmospheric Environment, 25A, 217-230, 1991.

Kellogg, W. W., Cadle, R. D., Allen, E. R., Lazrus, A. L., and E. A. Martell, The sulfur cycle. Science, 175, 587-596, 1972.

Kent, G. S., and McCormick, M. P., SAGE and SAM-II measurements of global stratospheric aerosol optical depth and mass loading, Journal of Geophysical Research, 89(D4): 5303-5314, 1984.

Kinnison, D. E., K.E. Grant, P.S. Connell, D.A. Rotman, and D. J. Wuebbles, The chemical and radiative effects of the Mount Pinatubo eruption, Journal of Geophysical Research, 99, 25,705-25,731, 1994.

Kjellström, E., A three-dimensional global model study of Carbonyl Sulfide in the troposphere and in the lower stratosphere, Journal of Atmospheric Chemistry,151-172,29, 1998.

Kodera K., M. Chiba, H. Koide, A. Kitoh, and Y. Nikaidou, Interannual variability of the winter stratosphere and troposphere in the Northern Hemisphere, Journal of the Meteorological Society of Japan, 74 (3), 365-382,1996.

Krueger, A. J., L. S. Walter, P. K. Bhartia, C. C. Schnetzler, N. A. Krotkov, I. Sprod, and G. J. S. Bluth, Volcanic sulfur dioxide measurements from the Total Ozone Mapping Spectrometer (TOMS) instruments, Journal of Geophysical Research, 100, 14057-14076, 1995.

Krueger, A. J., C. C. Schnetzler, and L. S. Walter, The December 1981 eruption of Nyamuragira Volcano (Zaire), and the origin of the "mystery cloud" of early 1982, Journal of Geophysical Research, 101, 15,191-15,196, 1996.

Lamb, H. H., Volcanic dust in the atmosphere; with a chronology and assessment of its meteorological significance, Philosophical Transactions of the Royal Society of London Series A-Mathematical and Physical Sciences, 266, 425-533, 1970.

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

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

Lambert, G., M.-F. Le Cloarec, and M. Pennisi, Volcanic output of SO_2 and trace metals: A new approach. Geochimica et Cosmochimica Acta, 52, 39-42, 1988.

Langmann, B., and H.-F. Graf, The chemistry of the polluted atmosphere over Europe: Simulations and sensitivity studies with a regional chemistry-transport-model, Atmospheric Environment 31, 3239-3257, 1997.

Le Guern, F., Les débits de CO_2 et de SO_2 volcaniques dans l'atmosphère. Bulletin of Volcanology, 45, 197-202, 1982.

Mankin, W. G., and M. T. Coffey, Increased stratospheric hydrogen chloride in the El Chichón cloud, Science, 226(4671), 170-172, 1984.

Mankin, W. G., M. T. Coffey, and A. Goldman, Airborne observations of SO_2, HCl, and O_3 in the stratospheric plume of the Pinatubo volcano in July 1991, Geophysical Research Letters, 19(2), 179-182, 1992.

Mauldin, L. E., III, N. H. Zaun, M. P. McCormick, J. H. Guy, and W. R. Vaughn, Stratospheric Aerosol and Gas Experiment II Instrument: A Function Description, Optical Engeneering, 24, 307-312, 1985.

McClelland L., T. Simkin, M. Summers, E. A. Nielsen, and T. C. Stein (eds), Global Volcanism 1975-1985: The First Decade of Reports from the Smithsonian Institution's Scientific Event Alert Network (SEAN). Prentice Hall and American Geophysical Union, Englewood Cliffs, 657 pp., 1989.

McCormick, M. P., P. Hamill, T. J. Pepin, W. P. Chu, T. J. Swissler, and L. R. Master, Satellite studies of the stratospheric aerosol, Bulletin of the American Meteorological Society, 60, 1038-1046, 1979.

McCormick, M. P., SAGE II: An overview, Advances in Space Research, 7, 219-226, 1987.

McCormick, M. P., L. W. Thomason, and C. R. Trepte, Atmospheric effects of the Mt Pinatubo eruption, Nature, 373, 399-404, 1995.

McElroy, M. B., R. J. Salawitch, and K. Minschwaner, The changing stratosphere, Planetary and Space Science, 40, 373–401,1992.

McKeen, S. A., S. C. Liu, and C. S. Kiang, On the chemistry of stratospheric SO_2 from volcanic eruptions, Journal of Geophysical Research, 89(D3), 4873-4881, 1984.

Menzies, R. T., and D. M. Tratt, Evidence of Seasonally Dependent Stratosphere-troposphere Exchange and Purging of Lower Stratospheric Aerosol from a Multiyear Lidar Data Set, Journal of Geophysical Research, 100, 3139-3148, 1995.

Michelangeli, D. V., M. Y. Allen, and L. Yuk, El Chichón volcanic aerosols: impact of radiative, thermal, and chemical perturbations, Journal of Geophysical Research, 94(D15), 18429-18443, 1989.

Naughton J. J., V. A. Greenberg, and R. Goguel, Incrustations and Fumarolic Condensates at Kilauea Volcano, Hawaii - Field, Drill-Hole And Laboratory Observations, Journal of Volcanology and Geothermal Research, 1(2), 149-165, 1976.

Newhall,C., and S. Self, The Volcanic Explosivity Index (VEI): An estimate of explosive magnitude for historical volcanism, Journal of Geophysical Research, 87, 1281-1238, 1982.

Nober F., H.-F. Graf, and D. Rosenfeld, Sensitivity of the global circulation to the suppression of precipitation by anthropogenic aerosols, in print Global Planet. Change, 2002.

Oberhuber, J. M., M. Herzog, H.-F. Graf, and K. Schwanke, Volcanic plume simulation on large scales. , Journal of Volcanology and Geothermal Research, 87, 29-53, 1998.

Oppenheimer C., P. Francis, and J. Stix, Depletion rates of SO_2 in tropospheric volcanic plumes, Geophysical Research Letters, 25, 2671-2674, 1998.

Oppenheimer, C., Limited global change due to the largest known Quaternary eruption, Toba 74 kyr BP?, Quaternary Science Reviews, 21(14-15), 1593-1609, 2002.

Palais, J., Sigurdsson, H., Petrologic evidence of volatile emissions from major historic an pre-historic volcanic eruptions, In: Berger, A., Dickinson, R.E., Kidson, R.E., Understanding Climate Change, American Geophysical Union, Washington, DC, 31-53, 1989.

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

Pinto, J. R., R. P. Turco, and O. B. Toon, Self-Limiting Physical and Chemical Effects in Volcanic Eruption Clouds, Journal of Geophysical Research 94, 11165-11174, 1989.

Pitari, G., and V. Rizzi, An Estimate of the Chemical and Radiative Perturbation of Stratospheric Ozone Following the Eruption of Mt. Pinatubo, Journal of Atmospheric Sciences, 50, 3260-3276, 1993.

Prather, M., Catastrophic loss of stratospheric ozone in dense volcanic clouds, Journal of Geophysical Research, 97(D9), 10187-10191, 1992.

Pyle, D. M., P. D. Beattie, and G. J. S. Bluth, Sulphur emissions to the stratosphere from explosive volcanic eruptions. Bulletin of Volcanology, 57, 663-671, 1996.

Quiroz, R. S., Compact review of observational knowledge of blocking, with emphasis on the long-wave composition of blocks, IN: Annual Climate Diagnostics Workshop, 7th, Boulder, CO., Oct. 18-22, 1982, Proceedings, Wash., D.C., U.S. National Oceanic and Atmospheric Administration, March, 1983, p. 104-107, Climate Analysis Ctr., NMC/NWS/NOAA, Wash., D.C. M.G.A, 35, 8-254,1984.

Randel, W. J, F. Wu, J. M. Russell III, J. W. Waters, and L. Froidevaux, Ozone and temperature changes in the stratosphere following the eruption of Mount Pinatubo, Journal of Geophysical Research, 100, 16753-16764, 1995.

Rani, A., D. S. N. Prasad, P. V. S. Madnawat, and K. S. Gupta, The role of free fall atmospheric dust in catalysing autoxidation of aqueous sulphur dioxide, Atmospheric Environment, 26A, 67-673, 1992.

Read, W. G., L. Froidevaux, and J. W. Waters, Microwave Limb Sounder measurement of stratospheric SO_2 from the Mt. Pinatubo volcano, Geophysical Research Letters, 20, 1299-1302, 1993.

Robertson, A., J. Overpeck, D. Rind, E. Mosley-Thompson, G. Zielinski, J. Lean, D. Koch, J. Penner, I. Tegen, and R. Healy, Hypothesized climate forcing time series for the last 500 years, Journal of Geophysical Research, 106, 14783-14803, 2001.

Robock, A., A latitudinally dependent volcanic dust veil index, and its effect on climate simulations, Journal of Volcanology and Geothermal Research, 11, 67-80, 1981a.

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

Robock, A., and J. Mao, Winter warming from large volcanic eruptions, Geophysical Research Letters, 19, 2405-2408, 1992.

Robock, A., and M. P. Free, Ice cores as an index of global volcanism from 1850 to the present, Journal of Geophysical Research, 100, 11,549-11,567, 1995.

Robock, A., and M. P. Free, The volcanic record in ice cores for the past 2000 years, Climatic Variations and Forcing Mechanisms of the Last 2000 Years, edited by Philip D. Jones, Raymond S. Bradley, and Jean Jouzel, Springer-Verlag, Berlin, 533-546, 1996.

Robock, A., Volcanic eruptions and climate, Reviews of Geophysics, 38, 191-219, 2000.

Rose, W. I., and C. A. Chesner, Worldwide dispersal of ash and gases from the earth largest known eruption: Toba, Sumatra, 75 ka. Palaeogeography Palaeoclimatology Palaeoecology, 89, 269-275, 1990.

Rose, W. I., D. J. Delene, D.J. Schneider, G. J. S. Bluth, A. J. Krueger, I. Sprod, C. McKee, H. Davies, and G.G.J. Ernst, Ice in the 1994 Rabaul Eruption Cloud: Implications for volcano hazard and atmospheric effects, Nature, 375, 477-479, 1995.

Rose, W. I., G. J. S. Bluth, and G. G. J. Ernst, Integrating retrievals of volcanic cloud characteristics from satellite remote sensors - a summary, Philosophical Transactions of Royal Society, Series A, 358 (1770), 1585-1606, 2000.

Rosen, J. M., N. Kjome, R. L. McKenzie, and J. B. Liley, Decay of Mount Pinatubo aerosol at mid-latitudes in northern and southern hemispheres, Journal of Geophysical Research, 99, 25733-25739, 1994.

Rosenfeld, J. E., Considine, D. B., Meade, P. E., Bacmeister, J. T., Jackmon, C. H., and M. R. Schoeberl, Stratospheric effects of Mount Pinatubo aerosol studied with a coupled two-dimensional model. Journal of Geophysical Research, 102, 3649-3670, 1997.

Russell, P. B., J. M. Livingston, R. F. Pueschel, J. B. Pollack, S. Brooks, P. Hamill, J. Hughes, L. Thomason, L. Stowe, T. Deshler, and E. Dutton, Global to microscale evolution of the Pinatubo volcanic aerosol, derived from diverse measurements and analyses, Journal of Geophysical Research, 101, 18745-18763, 1996.

Sassen, K., Evidence for liquid-phase cirrus cloud formation from volcanic aerosol: Climate implications, Science, 257, 516-519, 1992.

Sassen, K., D. O'C. Starr, G. G. Mace, M. R. Poellot, S. H. Melfi, W. L. Eberhard, J. D. Spinhirne, E. W. Eloranta, D. E. Hagen, and J. Hallett, The 5-6 December 1991 FIRE IFO II jet stream cirrus case study: possible influences on volcanic aerosols, Journal of Atmospheric Sciences, 52, 97-123 1995.

Sato, M., J.E. Hansen, M. P. McCormick, and J. B. Pollack, Stratospheric aerosol optical depths 1850-1990, Journal of Geophysical Research, 98, 22,987-22994,1993.

Scaillet, B., B. Clemente, B. W. Evans, and M. Pichavant, Redox controls of sulfur degassing in silicic magmas, Journal of Geophysical Research, 103, 23,937-23,949, 1998.

Schimel, D., D. Alves, I. Enting, M. Heimann, F. Joos, D. Raynaud, T. Wigley, M. Prather, R. Derwent, D. Ehhalt, P. Fraser, E. Sauhueza, X. Zhou, P., Jonas, R. Charlson, H. Rodhe, S. Sadasivan, K. P. Shine, Y. Fouquart, V. Ramaswamy, S. Solomon, J. Srinivasan, D. Albritton, R. Derwent, I. Isaksen, M. Lal, and D. Wuebbles, Radiative forcing of climate change, In: Climate Change 1995: The Science of Climate Change, J. T. Houghton, L. G. M. Filho, B. A. Callander, N. Harris, A. Kattenberg, and K. Maskell (eds), 65-131, Cambridge University Press, Cambridge, UK, 1996.

Schmincke, H.-U.,Transfer von festen, flüssigen und gasförmigen Stoffen aus Vulkanen in die Atmosphäre, Zeitschrift für Umweltchemie und Ökotoxikologie, 5, 27-44, 1993.

Schnetzler, C. C., G. J. S. Bluth, A. J. Krueger, and L. S. Walter, A proposed volcanic sulfur dioxide index (VSI), Journal of Geophysical Research, 102(B9), 20087-20091, 1997.

Seinfeld, J. H., and S. N. Pandis, Atmospheric Chemistry and Physics, From Air Pollution to Climate Change, John Wiley & Sons, New York, 1998.

Sheridan, P. J., R. C. Schnell, D. J. Hofmann, and T. Deshler, Electron microscope studies of Mt. Pinatubo aerosol layers over Laramie, Wyoming, during summer 1991, Geophysical Research Letters, 19, 203-206, 1992.

Simkin T. L. Siebert, L. McClelland, D. Bridge, and C. Newhall, J. Latter, Volcanoes of the World: a regional directory, gazetteer, and chronology of volcanism during the last 10 00 years, Hutchinson Ross Pub Co, Stroudsburg, 1981.

Simkin, T., Terrestrial volcanism in space and time, Annual Review of Earth and Planetary Sciences, 21, 427-452, 1993.

Simkin T., and L. Siebert , Volcanoes of the World: a Regional Directory, Gazetteer, and Chronology of Volcanism During the Last 10,000 Years, (second edition), Geoscience Press, Tucson, 1994.

Solomon, S., R. W. Portmann, R. R. Garcia, L. W. Thomason, L. R. Poole, and M. P. McCormick, The role of aerosol variations in anthropogenic ozone depletion at northern midlatitudes, Journal of Geophysical Research, 101, 6713-6727, 1996.

Solomon, S., R. W. Portmann, R. R. Garcia, W. Randel, F. Wu, R. Nagatani, J. Gleason, L. Thomason, L. R. Poole, and M. P. McCormick, Ozone depletion at mid-latitudes: coupling of volcanic aerosols and temperature variability to anthropogenic chlorine, Geophysical Research Letters, 25(11), 1871-1874, 1998.

Song, N., D. O'C. Starr, D. J. Wuebbles, A. Williams, and S. M. Larson, Volcanic aerosols and interannual variation of high clouds, Geophysical Research Letters, 23(19), 2657-2660, 1996.

Stenchikov, G. L., I. Kirchner, A. Robock, H.-F. Graf, J. C. Antuña, R. G. Grainger, A. Lambert, and L. Thomason, Radiative forcing from the 1991 Mount Pinatubo volcanic eruption, Journal of Geophysical Research, 103, 13,837-13,857, 1998.

Stevermer, A., I. Petropavlovskikh, J. Rosen, and J. DeLuisi, Development of global stratospheric aerosol climatology: Optical properties and applications for UV, Journal of Geophysical Research, 105, 22763-22776, 2000.

Stoiber, R. E., and A. Jepsen, Sulfur dioxide contributions to the atmosphere by volcanoes, Science, 182, 577-578, 1973.

Stoiber, R. E., S. N Williams, and B. Huebert, Annual contribution of sulfur dioxide to the atmosphere by volcanoes. Journal of Volcanology and Geothermal Research, 33, 1-8, 1987.

Stothers, R. B., Major optical depth perturbations to the stratosphere from volcanic eruptions: Pyrheliometric period 1881-1960, Journal of Geophysical Research, 101, 3901-3920,1996.

Symonds, R. B., W. I. Rose, and M. H. Reed, Contribution of Cl- and F-bearing gases to the atmosphere by volcanoes, Nature, 334, 415-418, 1988.

Symonds R. B., W. I. Rose, G. J. S. Bluth, and T. M. Gerlach, Volcanic gas studies: methods, results and applications, in: Volatiles in Magma, published by Mineral Society of America, M. R. Carrol and J. R. Holloway (eds), Reviews in Mineralogy, 30, 1-66, 1994.

Tabazadeh, A., and R. P. Turco, Stratospheric chlorine injection by volcanic eruptions: HCl scavenging and implications for ozone, Science, 260, 1082-1086, 1993.

Textor, C., Sachs, P.M., Graf, H.-F., and T. Hansteen, The 12,900 yr BP Laacher See eruption: estimation of volatile yields and simulation of their fate in the plume, In: Volcanic Degassing, Geological Society Special Publication, Clive Oppenheimer, David Pyle and Jenni Barclay (eds), in press 2002.

Textor, C., Graf, H.-F., Herzog, M., and J. M. Oberhuber, Injection of Gases into the Stratosphere by Explosive Volcanic Eruptions, accepted for publication by Journal of Geophysical Research, 2003.

Thomason, L. W., A diagnostic aerosol size distribution inferred from SAGE II measurements, Journal of Geophysical Research, 96, 22501-22508, 1991.

Thomason, L. W., L. R. Poole, and T. Deshler, A global climatology of stratospheric aerosol surface area density deduced from stratospheric aerosol and gas experiment II measurements: 1984-1994, Journal of Geophysical Research,102, 8967-8976, 1997.

Tie, X., G. P. Brasseur, B. Briegleb, and C. Granier, Two-dimensional simulation of Pinatubo aerosol and its effect on stratospheric ozone, Journal of Geophysical Research, 99, 20,545-20,562, 1994.

Timmreck, C., and H.-F. Graf, A microphysical model to simulate the development of stratospheric aerosol in a GCM. Meteorologische Zeitschrift, 9, 263-282, 2000.

Toohey, D. W., A critical review of stratospheric chemistry research in the U.S.: 1991-1994, U.S. National Report to the International Union of Geodesy and Geophysics 1991- 1994, Reviews of Geophysics, 33, 759-773, 1995.

Turco, R. P., Toon, O. B., Whitten, R. C., Hamill, P., and R. G. Keese, The 1980 eruption of Mount St. Helens: Physical and chemical processes in the stratospheric cloud, Journal of Geophysical Research, 88, 5299-5319, 1983.

Twomey, S., Pollution and planetary albedo. Atmospheric Environment, 8, 1251-1265, 1974.
Uchino, O., et al. Observation of the Pinatubo volcanic cloud by lidar network in Japan, Journal of the Meteorological Society of Japan, 71, 285-295, 1992.

Varekamp, J.C., J.F. Luhr, and K.L. Prestegaard, The 1982 eruptions of El Chichón Volcano (Chiapas, México): Character of the eruptions, ash-fall deposits, and gasphase, and Geothermal Research, 23, 39-68, 1984.

Veiga, R. E., SAGE II measurements of volcanic aerosols, Technical Digest Series, 5, Optical Society of America, Washington, D.C., 467-470, 1993.

Wallace, L., and W. Livingston, The effect of the Pinatubo cloud on hydrogen chloride and hydrogen fluoride, Geophysical Research Letters, 19(12), 1209, 1992.

Weisenstein, D. K., G. K. Yue, M. K. W. Ko, N.-D. Sze, J. M. Rodriguez, and C. J. Scott, A two-dimensional model of sulfur species and aerosols, Journal of Geophysical Research, 102, 13019-13053, 1997.

Westrich, H.R., and T.M. Gerlach, Magmatic gas source for the stratospheric SO2 cloud from the June 15, 1991 eruption of Mount Pinatubo, Geology, 20, 867-870, 1992.

Wilson, L., Explosive volcanic eruptions – III. Plinian eruption columns, Geophysical Journal of the Royal Astronomical Society, 45, 543-556, 1976.

Woods, D. C., R. L. Chuan, and W. I. Rose, Halite particles injected into the stratosphere by the 1982 El Chichón eruption, Science, 230, 170-172,1985.

Woods, A. W., The fluid dynamics and thermodynamics of volcanic eruption columns, Bulletin of Volcanology, 50, 169-193,1988.

Woods, A. W., Moist convection and the injection of volcanic ash into the atmosphere, Journal of Geophysical Research, 98, 17627-17636,1993.

Zielinski, G. A., Stratospheric loading and optical depth estimates of explosive volcanism over the last 2100 years derived from the GISP2 Greenland ice core, Journal of Geophysical Research, 100, 20937-20955, 1995.

Zielinski, G. A., P.A. Mayewski, L.D. Meeker, S. Whitlow, and M. Twickler, A 110,000 year record of explosive volcanism from the GISP2 (Greenland) ice core, Quaternary Research 45, 109-118, 1996.

Zielinski, G. A., Use of paleo-records in determining variability within the volcanism-climate system, Quaternary Science Reviews, 19, 417-438, 2000.

Zhao, J., R. P. Turco, and O. B. Toon, A model simulation of Pinatubo volcanic aerosols in the stratosphere, Journal of Geophysical Research, 100, 7315-7328, 1995.