The annual volcanic gas input into the atmosphere, in particular into the stratosphere: a global data set for the past 100 years

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Abstract

We compiled a global data set of volcanic degassing during both explosive and quiescent volcanic events. The data set comprises estimates of gas emissions of volcanoes from Europe (e.g. Etna), Asia (e.g. Merapi), the Americas (e.g. Fuego), Africa (e.g. Erta Ale) and ocean islands (e.g. Kilauea) over the past 100 yr. The set includes 50 monitored volcanoes and \( \sim 310 \) extrapolated explosively erupting volcanoes. Among the \( \sim 360 \) volcanoes, 75\% are located in the Northern and 25\% in the Southern Hemisphere. We have estimated the total annual global volcanic sulfur emission into the atmosphere to be on the order of \( 7.5-10.5 \times 10^{12} \) g/yr S (here as SO\(_2\)), amounting to 10-15\% of the annual anthropogenic sulfur output (\( \sim 70 \times 10^{12} \) g/yr S during the decade 1981-1990) and 7.5-10.5\% of the total global sulfur emission (e.g. biomass burning, anthropogenic, dimethylsulfide) with \( \sim 100 \times 10^{12} \) g/yr S. The estimates of other volcanic gases emitted (e.g. H\(_2\)S, HCl) are based on the assumption that the different gas components emitted by a volcano are in equilibrium with each other. Accordingly, the molar ratios of the gas species in high-temperature fumaroles are similar to molar ratios equilibrated at depth where the gas separates from the magma. Thus, we can use the directly measured SO\(_2\) fluxes and known molar ratios (e.g. H\(_2\)S/SO\(_2\)) for a semi-quantitative estimate of other gas components emitted (e.g. H\(_2\)S). The total annual emission of HCl is \( 1.2-170 \times 10^{12} \) g/yr, that of H\(_2\)S \( 1.5-37.1 \times 10^{12} \) g/yr, of HF \( 0.7-8.6 \times 10^{12} \) g/yr, of HBr \( 2.6-43.2 \times 10^{9} \) g/yr, and of OCS \( 9.4 \times 10^{7}-3.2 \times 10^{11} \) g/yr. We estimate an emission of \( 1.3 \times 10^{7}-4.4 \times 10^{10} \) g/yr for CS\(_2\). © 2002 Elsevier Science B.V. All rights reserved.

Keywords: sulfur dioxide; volcanic sulfuric gases; explosive degassing; stratospheric sulfur input

1. Introduction

Sulfate aerosols generated from volcanic emissions of sulfur-rich gas temporarily counteract some of the heating caused by elevated carbon dioxide concentrations in the atmosphere. Aerosols in the atmosphere cool the climate both directly due to backscattering of sunlight and indirectly through an increase in cloud reflectivity and residence time (e.g. Twomey, 1977). The fact that the emission of aerosol precursor gases from human activities appears to significantly affect the...
world climate necessitates a careful scrutiny of the primary natural climate-forcing processes among which the flux of volcanic gases into the atmosphere plays a major role. Here, we attempt to fill a gap that exists in our knowledge of global emission rates, especially with respect to sulfur, the major factor that influences both climate and ozone layer via the generation of long-lived stratospheric aerosol layers. Our assessment is based on a classification of volcanic activity associated with the gas flux data measured by the correlation spectrometer (COSPEC) and the total ozone mapping spectrometer (TOMS) and the annual frequency of the assigned eruption activity classified by the volcanic eruption index (VEI; Newhall and Self, 1982). Further, we used a modified volcanic sulfur dioxide index (VSI; Schnetzler et al., 1997) for historical eruptions prior to 1972, where no data of sulfur content in plumes measured by COSPEC and TOMS are available.

The amount of volcanic gases and particles arriving in the troposphere and/or stratosphere generally depends on both volcanological and meteorological boundary conditions:

- the chemical composition of the magma depending on the tectonic environment (e.g. subduction zone, continental rift zone);
- the strength and duration of an eruption;
- the location of the emitting volcano (altitude and latitude);
- atmospheric conditions such as temperature, humidity and wind profiles.

More than 90% of magmas erupted globally are of basaltic composition (Fig. 1). These magmas are characterized by low viscosity and relatively low gas content and are mostly erupted non-explosively. More than 90% of basaltic eruptions take place along the mid-oceanic ridges in deep water. A small fraction of basaltic volcanoes erupts subaerially such as Kilauea volcano on Hawaii, the most active volcano on Earth. Basaltic magmas are sulfur-rich. Eruptions of basaltic volcanoes add approximately 10–15% of the total globally erupted volcanic gases to the atmospheric gas budget. Volcanoes in island arcs and active continental margins are characterized by more silicic magmas and are the major source for the stratospheric volcanic gas input. Their eruption columns can rise up to 40 km and can, therefore, inject gases directly into the stratosphere. Once in the atmosphere, volcanic gases undergo chemical reactions. Sulfur dioxide and other volcanic sulfur-bearing gases transform to sulfate aerosols, which are able to cool the climate directly by backscattering incoming solar radiation and indirectly through increase in cloud reflectivity. Also, complex dynamic interactions lead to effects such

![Fig. 1. Global distribution of volcanoes in different tectonic settings.](image-url)
as ‘continental winter warming’ in the Northern Hemisphere following tropical eruptions (Graf et al., 1993). Reactions involving magmatic chlorine and bromine compounds strongly affect ozone concentrations in the stratosphere.

2. Volcanic gases and their reactivity in the global atmosphere

2.1. \( \text{SO}_2, \text{H}_2\text{S}, \text{HCl}, \text{HF}, \text{HBr}, \text{OCS} \) and \( \text{CS}_2 \)

Sulfate aerosol (\( \text{H}_2\text{SO}_4 \)) forms from sulfur dioxide (\( \text{SO}_2 \)) due to chemical reactions in the atmosphere. In the troposphere, however, volcanic sulfur dioxide will become rapidly oxidized to sulfate and washed out of the atmosphere in the presence of hydrometeors. These washout processes are not effective in the stratosphere. Since gaseous \( \text{H}_2\text{SO}_4 \) has a very low saturation pressure it can easily condens in the stratosphere and form aerosols of liquid hydrated sulfuric acid. Sulfate aerosols once formed have a residence time in the stratosphere of about 3 yr and can cause climate changes.

The halogens \( \text{HCl} \) and \( \text{HBr} \) effectively destroy ozone in the stratosphere in the form of the radicals Cl and Br. Mankin and Coffey (1984) documented a 40% increase in stratospheric chlorine between 20°N and 40°N following the eruption of El Chichón in 1982. A combined chlorine–bromine interaction can be regarded as a synergetic effect of Cl and Br, which is very effective in the polar stratosphere in destroying \( \text{O}_3 \) where oxygen atoms occur in low concentrations (Stolarski, 1979). HF is the dominant fluorine species in volcanic gases. The annual global industrial production of F as halocarbons amounted to \( 0.273 \times 10^{12} \) g between 1982 and 1984 (Symonds et al., 1988), on the same order of magnitude as that resulting from explosive volcanic eruptions. Fluorine compounds do not interact with stratospheric ozone, however.

Carbonyl sulfide (OCS) is a source of stratospheric background sulfate aerosol (Crutzen, 1976). This trace gas is the most abundant sulfur compound in the atmosphere, the OCS/\( \text{CO}_2 \) ratio being positively correlated with the emission temperature of fumarolic gas (Belviso et al., 1986). Much of OCS is transported into the stratosphere because it is chemically nearly inert in the troposphere. The photochemical dissociation of OCS in the stratosphere is another stratospheric source of \( \text{SO}_2 \). \( \text{CS}_2 \) undergoes chemical reactions in the stratosphere similar to those of OCS. Hydrogen sulfide (\( \text{H}_2\text{S} \)) is a water-soluble species. Its dominant tropospheric removal process involves an OH radical reaction.

2.2. Residence times of volcanic gases in troposphere and stratosphere

The residence time of water-soluble sulfuric gases (in particular \( \text{SO}_2 \)) emitted into the atmosphere depends on the height of an eruption column. It is, therefore, crucial whether or not volcanic gases are injected directly into the stratosphere or remain in the troposphere. The main sources for the stratospheric sulfur input are highly explosive volcanic eruptions with a VEI \( \geq 3 \) (Table 1). Some eruptions, e.g. the eruption of El Chichón in 1982, which emitted \( 7 \times 10^{12} \) g \( \text{SO}_2 \), can easily increase the stratospheric \( \text{SO}_2 \) budget by one order of magnitude (Bluth et al., 1992). OCS is a dominant stratospheric source for sulfate aerosols during periods lacking significant volcanic activity. The concentration of OCS in the troposphere is almost independent of altitude and latitude and this gas can remain in the atmosphere for up to 1 yr because of its stability and inertness towards water (Crutzen, 1976; Schult, 1991). In the stratosphere, OCS can remain for up to 10 yr (Chin and Davis, 1995). The reservoir of OCS in the troposphere is estimated to be eight times that in the stratosphere. Therefore, the OCS has a long-term effect on the Earth’s atmospheric chemistry.

3. Method

3.1. Instrumental methods

It is extremely difficult to determine gas and particle concentrations in a volcanic plume because of the plume’s opaqueness and the risk of
directly observing and sampling a plume. Volcanic emissions can be studied remotely by airborne and ground-based instruments (COSPEC) apart from satellite observations (TOMS). COSPEC is important in measuring sulfur dioxide injected into the troposphere during quiescent degassing of volcanic eruptions with low VEI. TOMS measures SO$_2$ primarily in the stratosphere during explosive eruptions. We used SO$_2$ gas data measured by both TOMS and COSPEC, both of them working only in daylight because of extinction of SO$_2$ in a specific range of the sunlight wavelengths (Kaye et al., 1998). The nightly gas emission needs to be extrapolated from the gas emission data monitored in daylight.

The principal problem with the TOMS data is that not every eruption is covered. Moreover, the minimum amount of SO$_2$ emitted during an eruption that can be measured by TOMS is about 5–15 kilotons (kt = 10$^9$ g) SO$_2$ (Bluth et al., 1992). Thus, high SO$_2$ emissions such as the one emitted from Mt. Pinatubo in June 1991 (≈ 20×10$^{12}$ g; Bluth et al., 1992) are best monitored by TOMS.

Large amounts of sulfur are released by quiescent degassing, especially from basaltic volcanoes and fumarole fields. But the energy level of quiescent degassing is generally too low to allow sulfur to directly enter the stratosphere. A rapid conversion of SO$_2$ to sulfate in a moist atmosphere can also hinder further rise of the emitted SO$_2$ gas. Sulfur from fumarolic sources can be measured more accurately with COSPEC than with TOMS. Finally, TOMS instruments were not originally designed to directly measure sulfur dioxide. Instead, SO$_2$ quantities are inferred from UV reflectance data based on an algorithm and the unique conditions surrounding each volcanic eruption. We included data directly measured by both instruments, TOMS and COSPEC, in our estimates of gas emissions. Many weak eruptions measured by COSPEC are irrelevant in assessing stratospheric gas loading. COSPEC measures the concentration of SO$_2$ in either a vertical column or horizontal (bent-over or wind-blown) plume by scanning across the plume perpendicular to the transport direction. Errors occur when physical barriers restrict perpendicular plume observations or when the relative locations of the instrument and plume are not precisely known. Another source of error is the difficulty in determining the rising rates for vertical plumes or transport velocity for horizontal plumes. The inherent uncertainty of COSPEC data is due to differences in monitoring techniques and the fact that gas plumes are monitored at varying distances from a crater and during different stages of an eruption. The uncertainty in COSPEC results at different distances also reflects rapid conversion rates (Oppenheimer et al., 1998). It is, therefore, not surprising that data on SO$_2$ concentrations obtained in the ascending plume by COSPEC differ significantly from those measured in the stratosphere by TOMS.

3.2. Classification of volcanic activity and gas emission

Our data set comprises 360 out of ≈ 1500 explosively degassing subaerial volcanoes, of which 50 were monitored directly by TOMS and/or COSPEC; the sulfur emission of the remaining 310 volcanoes was extrapolated (Fig. 1). The method
to arrive at a global annual data set of the past 100 yr of subaerial volcanic gas emission is simple. We studied every publication potentially related to directly monitored volcanic gas emissions emitted by either silent or explosive degassing. There are literature data of directly measured SO$_2$ from $\sim$50 volcanoes. We then compared volcanic parameters such as magma composition, tectonic setting, stage of eruption when the gas emission of each monitored volcano was measured, with 310 unmonitored active subaerial volcanoes. We studied all activity reports by Simkin and Siebert (1994) and in the Web pages of the Smithsonian Institution (http://www.volcano.si.edu/gvp/) to collect a data base only of eruptive activities for the 310 volcanoes not remotely monitored. The energy of volcanic activity is generally classified by the VEI (Table 1) of Newhall and Self (1982). The VEI is based on the height of the eruption column and the volume of material ejected. It is an open-end scale from VEI 0 for small explosive eruptions (Hawaiian) to VEI 8 for the largest known historic eruption (Ultraplinian). A positive relationship between average sulfur dioxide emission and VEI has been found by Stoiber et al. (1987).

We extrapolated the gas data from the 50 monitored volcanoes to the 310 unmonitored volcanoes to calculate a global annual volcanic gas emission based on TOMS and COSPEC data between 1972 and 2000. Remote sensing is carried out from a distance and requires looking through the atmosphere at a gas emission plume. TOMS data were used for very explosive eruptions; the algorithm to quantify SO$_2$ was modernized by Kaye et al. (1998). We did not extrapolate gas data measured by TOMS to historic explosive eruptions because the sources of error are too manifold. When using TOMS data, three main types of error sources have to be taken into account to quantify volcanic SO$_2$ emissions: (1) the measurement error of scan pixels; (2) the cloud tonnage error; and (3) the error in extrapolating back to an original emission (Symonds et al., 1995). Implicit in TOMS tonnage estimates are a number of assumptions and corrections, which introduce large uncertainties in our evaluation of the global annual volcanic SO$_2$ emission. CO-SPEC instruments can operate in an airplane, from a car or a boat, and also from a fixed point or a tripod (Rose et al., 1985). An important factor that influences the quality of COSPEC data is the necessity of measuring the wind speed that is moving the plume, or the rise speed of the plume. The uncertainty in the plume movement speed is usually the largest error source in individual COSPEC flux measurements. The high uncertainties (usually 20–50%) associated with COSPEC-determined fluxes are largely due to imperfect wind speed measurements (Symonds et al., 1995). Our data set thus includes major differences in the quality of SO$_2$ data measured with COSPEC on quiet and mildly explosive degassing. We further assessed the stage of an eruption at which SO$_2$ emission was measured. We extrapolated SO$_2$ emissions for other volcanoes based on similar parameters to monitored volcanoes:

- stage of activity (silent to explosive);
- tectonic setting (subduction zone, rift zone and ocean island);
- magma composition (basaltic to highly differentiated).

For example, we extrapolated SO$_2$ emission rates of Asama volcano (Japan) measured during its quiet phase to other Japanese volcanoes showing a similar stage of activity and magma composition. We divided volcanoes of a particular tectonic setting such as subduction zone into separate provinces, e.g. Japanese, Kamchatkan and so on. The same procedure was carried out for the different activity stages of a volcano.

We calculated the annual global volcanic SO$_2$ emission of silent and explosive degassing for the time period of the early 1970s until today for all 360 volcanoes. Because continuously acquired data are rare, we calculated the annual SO$_2$ emission based on 10-yr intervals. For this purpose, we divided literature data into 10-yr intervals beginning with the 1970s when the direct measurement of SO$_2$ by COSPEC started. We also calculated a SO$_2$ emission based on the original VSI (Schnetzler et al., 1997) and multiplied the values of the original VSI with a factor of approximately 2 to match the values of measured SO$_2$ emission. The VSI is based on the quantity of volcanic SO$_2$ produced by explosive eruptions and is scaled in
different degrees of SO₂ emission (in kt) based on the VEI scale. Schnetzler et al. (1997) divided volcanoes into subduction zone-related volcanoes and other volcanoes. We accepted this division for the modified VSI scale (Table 2). Schnetzler et al. (1997) developed the VSI using the relationship of the average sulfur dioxide emission and the VEI of volcanic eruptions for their index. We modified the VSI by the factor 2 because it significantly underestimated the quantity of degassed SO₂. In some extraordinary sulfur-rich eruptions such as El Chichón (1982) the modified VSI still slightly underestimates the SO₂ emission. The index is quite appropriate, however, for average sulfur-rich eruptions. We related the eruption frequency to VEI from 1900 to 1972 for the time series analyses prior to 1972 and determined the annual eruptive activity for each VEI category. Multiplying the new VSI value (average quantity of SO₂ degassed during an explosive eruption scaled with VEI) by the number of annual eruptions results in a total annual global volcanic SO₂ emission from explosive eruptions for a certain year.

The annual global volcanic SO₂ emission from 1972 to 2000 was first calculated with the original VSI by Schnetzler et al. (1997). The result was then compared with our estimate of global volcanic SO₂ emission based on 50 directly monitored volcanoes and the 310 extrapolated volcanoes. We modified the VSI by a factor of 2 for calculating the SO₂ emissions by volcanic eruptions prior to the period of monitoring by COSPEC and TOMS because the original VSI calculates a minimum quantity of the SO₂ emission based on the 50 directly monitored volcanoes. The modified VSI is very useful to quantify the sulfur yield of volcanic eruptions and is used as a base for our minimum estimate of the quantitative volatile input into the stratosphere for historic eruptions, especially from 1900 to 1971. It is a minimum value because the VSI does not cover quiescent degassing and is used only for explosively degassing volcanoes before remote sensing techniques became operative.

3.3. Classification of emissions of gases such as HCl, H₂S, OCS

Our calculation of the emission of the gases HCl, HF, OCS is based on mole ratios and the assumption that the gas composition in the emitted volcanic plume depends on:
- magma composition and melting process;
- separation of gases during magma ascent;
- processes inside the volcano edifice.

The relative concentrations of gases in high-temperature fumaroles are taken to be similar to those existing at depth when the gases separate from the magma (Giggenbach, 1996). This is based on the assumption that the difference in pressure has only a small effect on the gas equilibrium in the magma during ascent. Earlier studies indicate that high-temperature volcanic gases approach a state of thermodynamic equilibrium early on (e.g. Ellis, 1957; Heald et al., 1963).

The equilibrium compositions apparently represent the last equilibrium of the high-temperature volcanic gases before the gases stopped reacting because of rapid cooling. Most gas analyses prior to 1975 show evidence of severe disequilibrium (Gerlach, 1980). Modern improved collection procedures and analytical methods showed only mild to moderate gas disequilibria (Gerlach, 1993). The concentration of gases such as HCl, H₂S, HF, OCS can be estimated quantitatively from directly measured SO₂ fluxes related to known molar ratios, e.g. HCl/SO₂, HF/SO₂, CO₂/SO₂, OCS/CO₂, sampled in high-temperature fumaroles (Table 3).
Table 3
Gas contents and ratios in high-temperature fumaroles from different macro-tectonic environments in mol%. Equilibrium compositions and temperatures and low-pressure (1 atm) volcanic gases from subduction zone-related, rift zone-related and ocean island-related volcanoes. The concentrations are always average values from several samples taken at a volcano; _, not measured.

| Volcano                | Tectonic environment | Magma type  | H₂O | H₂ | CO₂ | CO  | SO₂ | H₂S | HCl | HF | CO₂/H₂O | SO₂/H₂ | SO₃/H₂ | SO₄/H₂ | HCl/HF | H₂O/H₂ | OCS/H₂ | CO₂/H₂ |
|------------------------|----------------------|-------------|-----|----|-----|-----|-----|-----|-----|----|---------|---------|--------|--------|--------|--------|--------|--------|--------|
| Etna, Italy            | S                    | Nevada      | 1000 | 19.6 | 22.2 | 25.2 | 3.4  | -   | 0.88 | -  | 7.31    | 4       | -      | -      | -      | -      | -      | -      |
| Iwo Shima, Japan       | S                    | Andesite   | 1000 | 98.1 | 0.3  | 0.4  | 0.05 | -   | 0.75 | -  | 8       | -       | -      | -      | -      | -      | -      | -      | -      |
| Usu, Japan             | S                    | Andesite-arc | 640 | 99.3 | 0.06 | 0.85 | 0.29 | 0.26 | 0.36 | -  | 16.7    | 0.66    | 0.89   | -      | 381.9  | -      | -      | -      | -      |
| Shosashinzan, Japan    | S                    | Andesite   | 667 | 96.5 | 0.31 | 2.37 | 0.004 | 0.21 | 0.53 | 0.12 | 0.02    | 11.3    | 10.5   | 1.75   | 0.39   | 6.0    | 311.3  | -      |
| Mt. St. Helens, U.S.A. | S                    | Daicite    | 640 | 98.5 | 2.4  | 9.1  | 0.7  | 1.4  | 0.9  | -  | 12.5    | 0.8     | 0.63   | -      | 40.4   | -      | -      | -      | -      |
| St. Augustine, Alaska  | S                    | Andesite-dacite | 715 | 96.2 | 0.5  | 2.31 | 0.02 | 0.12 | 0.02 | 0.08 | 4.03    | 0.00    | 1.96   | 0.85   | -      | -      | -      | -      | -      |
| Tolbachik, Kamchatka   | S                    | Basalt     | 1050 | 88.17 | 1.89 | 2.48 | 0.98 | 0.14 | 0.65 | -  | 31      | 0.11    | 0.12   | 1230   | 48.7   | -      | -      | -      | -      |
| Klyuchevskoy, Kamchatka| S                    | Basalt     | 1075 | 94.79 | 1.15 | 2.30 | 0.1  | 1.72 | 0.23 | -  | 0.06    | 84.0    | 0.92   | -      | 81.4   | -      | -      | -      | -      |
| White Island, New Zealand | S                | Andesite  | 640 | 93.2 | 4.1  | -    | 1.1  | 0.05 | 0.9  | -  | 3.7     | 1.21    | 2.2    | -      | -      | -      | -      | -      |
| Kilauea, Hawaii        | S                    | Basalt     | 640 | 96.2 | 2.6  | 16.1 | -    | 10.2 | 5.8  | 2.5 | -      | 1.6     | 2.1    | 1.5    | -      | 384    | -      | -      | -      |
| Momotombo, Nicaragua   | S                    | Tholeiitic-basalt | 751 | 97.4 | 0.46 | 1.45 | 0.006 | 0.41 | 2.38 | 2.01 | 0.25    | 3.54    | 1.64   | 0.15   | 17.8   | 11.2   | 216.4  | -      |
| Poas, Costa Rica       | S                    | Tholeiitic-basalt | 1002 | 96  | 0.58 | 0.82 | 0.008 | 1.65 | 0.06 | 0.76 | 0.09   | 0.5     | 18.1   | 2.15   | 27.2   | 8.44   | 165.5  | -      |
| Merapi, Indonesia      | S                    | Andesite   | 819 | -    | -    | -    | -    | -    | -    | -  | -      | -       | -      | -      | -      | -      | -      | -      | -      |
| Guntung Merapi         | S                    | Andesite   | 733 | 92.5 | 1.07 | 5.0  | 0.08 | 0.64 | 0.59 | 0.04 | -      | 7.81    | 16     | 1.08   | 39.4   | -      | -      | -      | -      |
| Nyiragongo, Africa     | R                    | Melilite-melilitine | 956 | 44.7 | 1.4  | 4.7  | 2.5  | 2.2  | 1.5  | -  | 21.8    | 1.38    | -      | -      | 0.1    | -      | -      | -      | -      |
| Erta' Ale, Africa      | R                    | Tholeiitic-basalt | 1076 | 40.4 | 14.9 | 29.9 | -    | 18.3 | 0.9  | -  | 16      | 1.6     | 10.2   | -      | 2.7    | 1.99   | 0.04   | -      | -      | -      |
| Anoukroba,Africa       | R                    | Tholeiitic-basalt | 1070 | 77.4 | 1.7  | 3.7  | 0.2  | 14.2 | 1.7  | -  | 0.26    | 8.38    | -      | -      | 45.3   | 3.24   | 0.04   | -      | -      | -      |
| Surtsey, Iceland       | R                    | Alkaline-basalt | 1126 | 92.0 | 1.65 | 1.94 | 0.07 | 3.27 | 0.07 | -  | 0.59    | 27.3    | -      | 55.8   | -      | -      | -      | -      |
| Kiluaea, Hawaii        | OI                   | Tholeiitic-basalt | 1148 | 48.8 | 0.68 | 34.47 | 1.05 | 14.61| 0.11 | 0.16 | 0.02   | 2.36    | 70.1   | 97.3   | 32.8   | 0.8    | 71.8   | -      |

Refs.: 1 = Gerlach (1979); 2 = Iwasaki (1965); 3 = Giggenbach et al. (1986); 4 = Matsuo et al. (1982); 5 = Gerlach and Casadevall (1986); 6 = Kodosky et al. (1991); 7 = Menyailov and Nikitina (1980); 8 = Taran et al. (1991); 9 = Giggenbach (1974); 10 = Bernard (1985); 11 = Delorme (1983); 12 = Belviso et al. (1986); 13 = LeGuern et al. (1982); 14 = Gerlach (1980); 15 = Allard (1983).
4. Annual global volcanic gas emissions emitted explosively

4.1. Sulfur dioxide

About 30–50 out of a total of more than 360 active subaerial volcanoes have erupted explosively each year between 1900 and 2000 (Simkin and Siebert, 1994; the Web pages of the Smithsonian Institution). The annual SO2 fluxes along the major tectonic environments including their geographic locations (Table 4) emphasize the importance of subduction zone-related volcanoes. Only a few active subaerial volcanoes are located in rift zones and on ocean islands. Some of them, such as Kilauea, emit large amounts of SO2 and degas at relatively consistent rates. In contrast, forceful degassing during explosive eruptions (e.g. Mt. Pinatubo) is notoriously sporadic and more difficult to quantify. The total annual volcanic gas emission of sulfur dioxide is therefore not well-constrained. We calculated $15–21 \times 10^{12} \text{g SO}_2$ as a total annual global volcanic SO2 input into the atmosphere from both silent and explosive degassing volcanoes (Table 4), corresponding to $7.5–10.5 \times 10^{12} \text{g/yr S}$ (S here as SO2) for the time period 1972 to 2000, the numbers representing an average value for volcanic gas emission over almost 30 yr. To split up the emissions into single years we calculated an annual SO2 emission

Table 4
Annual global volcanic SO2 emissions into the atmosphere, divided into tectonic and geographic environments, 1972–2000

<table>
<thead>
<tr>
<th>Geographic location</th>
<th>Tectonic environment</th>
<th>SO2 emission/yr (10^{12} g/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hawaii</td>
<td>Ocean island</td>
<td>0.7–0.8</td>
</tr>
<tr>
<td>Africa</td>
<td>Rift</td>
<td>1.6–2.1</td>
</tr>
<tr>
<td>Antarctica</td>
<td>Rift</td>
<td>0.01–0.02</td>
</tr>
<tr>
<td>Iceland</td>
<td>Rift</td>
<td>0.8–1.0</td>
</tr>
<tr>
<td>Europe (Italy)</td>
<td>Subduction zone</td>
<td>2.9–3.2</td>
</tr>
<tr>
<td>Japan</td>
<td>Subduction zone</td>
<td>1.9–2.7</td>
</tr>
<tr>
<td>North America</td>
<td>Subduction zone</td>
<td>0.1–0.2</td>
</tr>
<tr>
<td>Central America</td>
<td>Subduction zone</td>
<td>0.6–0.8</td>
</tr>
<tr>
<td>South America</td>
<td>Subduction zone</td>
<td>2.1–3.6</td>
</tr>
<tr>
<td>Kamchatka</td>
<td>Subduction zone</td>
<td>1.2–2.1</td>
</tr>
<tr>
<td>Alaska</td>
<td>Subduction zone</td>
<td>0.6–1.0</td>
</tr>
<tr>
<td>Indonesia</td>
<td>Subduction zone</td>
<td>2.1–2.6</td>
</tr>
<tr>
<td>New Zealand</td>
<td>Subduction zone</td>
<td>0.1–0.4</td>
</tr>
<tr>
<td>Philippines and southeast Asia (without Pinatubo)</td>
<td>Subduction zone</td>
<td>0.3–0.4</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>≈15–21</td>
</tr>
</tbody>
</table>
from sporadic explosive eruptions (based on COSPEC and TOMS data) to the year the eruption occurred in. For example, the emissions from Indonesian volcanoes are probably underestimated due to incomplete monitoring (Table 4). Most Indonesian data are from the very active Merapi volcano, which is a low-SO2 emitter. In contrast, volcanoes in Kamchatka and Alaska are major sulfur sources, dominantly by silent degassing.

We estimated that 40% of the total annual volcanic SO2 emission into the atmosphere is caused by permanent quiescent SO2 degassing based on the volcanoes directly monitored by COSPEC. The amount of SO2 emission by quiescent degassing was estimated based on the active volcanoes erupting between 1972 and 2000. Hence the data of measured SO2 emissions during non-eruptive stages were extrapolated for the volcanoes showing quiet degassing activity. The remaining 60% of the volcanic SO2 emissions are produced by explosive volcanic eruptions. Accepting this partitioning and our estimate of 15.0–21.0 \( \times 10^{12} \) g/yr SO2 (= 7.5–10.5 \( \times 10^{12} \) g/yr S) as a total volcanic emission for the period between 1972 and 2000, explosive degassing produces 9.0–12.6 \( \times 10^{12} \) g/yr SO2 (= 60%), quiescent degassing 6.0–8.4 \( \times 10^{12} \) g/yr SO2 (= 40%).

The differences in the estimates of SO2 emission estimated from 1972 to 2000 (Table 5) illustrates the dilemma of achieving a reliable estimate of the annual global volcanic SO2 emissions. These estimates range from 1.5 to 50 \( \times 10^{12} \) g/yr SO2. One group includes the very low estimates from Kellogg et al. (1972) (1.5 \( \times 10^{12} \) g/yr SO2), Friend (1973) (4.0 \( \times 10^{12} \) g/yr SO2), Cadle (1975) (7.0 \( \times 10^{12} \) g/yr SO2) and Granat et al. (1976) (6.0 \( \times 10^{12} \) g/yr SO2), which are one order of magnitude lower than the estimates from other authors. A second group includes estimates of SO2 emissions from Stoiber and Jepsen (1973) (10.0 \( \times 10^{12} \) g/yr SO2), LeGuern (1982) (10.0 \( \times 10^{12} \) g/yr SO2), Berresheim and Jaeschke (1983) (15.2 \( \times 10^{12} \) g/yr SO2), Stoiber et al. (1987) (18 \( \times 10^{12} \) g/yr SO2), Andres and Kasgnoc (1997) (13.0 \( \times 10^{12} \) g/yr SO2) and our estimate of 15.0–21.0 \( \times 10^{12} \) g/yr SO2. A third group shows the highest amounts of annual SO2 emission including Bartels (1972) (34.0 \( \times 10^{12} \) g/yr SO2), Naughton et al. (1975) (47.0 \( \times 10^{12} \) g/yr SO2), Lambert et al. (1988) (50.0 \( \times 10^{12} \) g/yr SO2) and Graf et al. (1997) (28.0 \( \pm 12 \) \( \times 10^{12} \) g/yr SO2).

Our estimate of 15–21 \( \times 10^{12} \) g/yr SO2 is believed to be more realistic than previous budgets, because we included more than 360 active subaerial volcanoes and the fluxes of 50 directly monitored volcanoes. Our results for the annual global volcanic sulfur emission are based on a running average of 10 yr (early 1970s to the end of the 1990s), since it is generally difficult to precisely evaluate volatile fluxes from active volcanoes over a long time period. The use of such running average of 10 yr is long enough to remove much of the inter-annual variability, but is short enough to reflect long-term degassing trends.

Table 6 shows the total volcanic sulfur input into the atmosphere and into the stratosphere compared with the total anthropogenic sulfur emission. The decrease in anthropogenic sulfur emission from the 1980s to the 1990s is most likely due to the collapse of the East European economy and, to a smaller extent, stricter environmental controls. The increase in the emission rates from a minimum of 15 \( \times 10^{12} \) g/yr to a maximum of 21 \( \times 10^{12} \) g/yr over the past three decades (7.5–19.5 \( \times 10^{12} \) g/yr S) relative to the anthropogenic

Table 5

<table>
<thead>
<tr>
<th>Authors</th>
<th>SO2 (( 10^{12} ) g/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kellogg et al. (1972)</td>
<td>1.5</td>
</tr>
<tr>
<td>Bartels (1972)</td>
<td>34.0</td>
</tr>
<tr>
<td>Friend (1973)</td>
<td>4.0</td>
</tr>
<tr>
<td>Stoiber and Jepsen (1973)</td>
<td>10.0</td>
</tr>
<tr>
<td>Cadle (1975)</td>
<td>7.5</td>
</tr>
<tr>
<td>Naughton et al. (1975)</td>
<td>47.0</td>
</tr>
<tr>
<td>Granat et al. (1976)</td>
<td>6.0</td>
</tr>
<tr>
<td>Friend et al. (1982)</td>
<td>4.0</td>
</tr>
<tr>
<td>LeGuern (1982)</td>
<td>10.0</td>
</tr>
<tr>
<td>Berresheim and Jaeschke (1983)</td>
<td>15.0</td>
</tr>
<tr>
<td>Stoiber et al. (1987)</td>
<td>18.0</td>
</tr>
<tr>
<td>Lambert et al. (1988)</td>
<td>50.0</td>
</tr>
<tr>
<td>Andres and Kasgnoc (1997)</td>
<td>13.0</td>
</tr>
<tr>
<td>Graf et al. (1997)</td>
<td>28.0 ( \pm 12 )</td>
</tr>
<tr>
<td>This work (2001)</td>
<td>15.0–21.0</td>
</tr>
</tbody>
</table>
sulfur input is therefore due mainly to the decrease of the anthropogenic sulfur emission. During the 1990s, the volcanic emissions make up 25–30% of the total compared with the anthropogenic input (35×10^{12} g/yr S; Graedel and Crutzen, 1995), while amounting to 10–13% during the 1980s (70×10^{12} g/yr S; Oørn et al., 1996). The amount of emission given as a percentage of the annual volcanic sulfur emissions (10–13% in the 1980s and 25–30% in the 1990s) is due to the reduction of anthropogenic sulfur emissions. The slight increase in SO_2 starting with the 1950s is caused by an increase in monitoring of volcanic eruptions.

Volcanic SO_2 emissions from subduction zone volcanoes (dacitic/andesitic magma) comprise approximately 70–80% of the total volcanic sulfur dioxide emissions (Table 4), calculated by adding the SO_2 emissions from subduction zone-related volcanoes and comparison with the total volcanic SO_2 emission. The emissions of SO_2 and other volcanic aerosol precursor gases were obtained by multiplying the SO_2 based on the modified VSI with the gas ratios measured at high-temperature fumaroles based on the emitted volcanic SO_2 (Table 7).

### Table 6
Volcanic sulfur emission based on COSPEC/TOMS data compared with the anthropogenic sulfur emission (1972–2000)

<table>
<thead>
<tr>
<th>Time</th>
<th>Volcanic S (atmosphere) (10^{12} g/yr)</th>
<th>Volcanic S (stratosphere) (10^{12} g/yr)</th>
<th>Anthropogenic S (10^{12} g/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1972–1980</td>
<td>7.5–8.5</td>
<td>1.2–1.4</td>
<td>60.0^a</td>
</tr>
<tr>
<td>1981–1990</td>
<td>8.5–9.5</td>
<td>1.3–1.9</td>
<td>70.0^b</td>
</tr>
<tr>
<td>1991–2000</td>
<td>9.5–10.5</td>
<td>1.8–3.3</td>
<td>35.0^b</td>
</tr>
</tbody>
</table>

The volcanic S is here SO_2.

^a^ From Oørn et al. (1996).


4.2. Halogens (HCl, HF, HBr)

We distinguished the SO_2 emission into three tectonic sections of annual SO_2 emission for evaluating the quantity of the other volcanic gases such as H_2S, HCl, etc., based on their ratios with SO_2 taken from high-temperature fumaroles (Table 4).

We used an average value and disregarded the extremely Cl-rich volcano Mt. Augustine because taking the ratio of Mt. Augustine into the average SO_2/HCl ratio would increase the order of magnitude of the annual global HCl output by just one volcano. Augustine is definitely an exception regarding its very high-Cl-bearing eruptions.

We obtained an annual emission input of 1.2–170×10^{12} g/yr HCl for subduction zone-related volcanoes with a molar ratio of SO_2/HCl = 0.1–10. For all other volcanoes, the molar ratio for SO_2/HCl is 97, which gives an annual HCl emission of 3.2–4.0×10^{10} g. This sums up to a total range of 1.2–170×10^{12} g/yr HCl (Table 7). The highly water-soluble HCl is almost totally removed (99.99%) by washout during the ascent of the volcanic plume. Thus, only a minimum amount of the initial emission by explosive erup-
tions can finally reach the stratosphere \((0.1 - 12.0 \times 10^{12} \text{ g/yr HCl})\). For HF, the molar ratio of \(\text{SO}_2/\text{HF} = 2 - 18\) for subduction zone-related volcanoes and \(\text{SO}_2/\text{HF} = 70\) for other volcanoes. We find for the subduction zone \(0.7 - 8.5 \times 10^{12} \text{ g/yr HF}\) and for the others \(4.4 - 5.6 \times 10^{10} \text{ g/yr HF}\). This sums up to a total annual input of \(0.7 - 8.6 \times 10^{12} \text{ g for HF}\) (Table 7). We calculated a HF stratospheric input by volcanoes as \(0.03 - 0.04 \times 10^9 \text{ g/yr}\). Bromine to chlorine ratios measured in volcanic gases by Yung et al. (1980) are \(0.4 - 2.4 \times 10^{-3}\). We calculated an amount of \(2.6 - 43.2 \times 10^9 \text{ g/yr HBr}\) (Table 7). We also present our estimates of water-soluble gas species (in particular HCl) reaching the stratosphere by taking into account the washout effect for each gas species (Table 7).

### 4.3. Other sulfuric gases \((\text{H}_2\text{S, OCS, CS}_2)\)

The total annual emission of \(\text{H}_2\text{S}\) ranges from \(1.5\) to \(37.1 \times 10^{12} \text{ g/yr}\), whereby \(1.2 - 34.0 \times 10^{12} \text{ g/yr}\) are emitted from subduction zone-related volcanoes, \(0.2 - 3.1 \times 10^{12} \text{ g/yr}\) from rift zone volcanoes and only \(5.4 - 6.2 \times 10^9 \text{ g/yr}\) from ocean island volcanoes. The molar ratio of \(\text{SO}_2/\text{H}_2\text{S}\) varies between \(0.5\) and \(10\) for subduction zone-related volcanoes; volcanoes along rift zones show a similar molar ratio of \(1 - 10\). The molar ratio for oceanic island volcanoes is exceptionally high with \(130\) for \(\text{SO}_2/\text{H}_2\text{S}\). The stratospheric loading of \(\text{H}_2\text{S}\) is estimated as \(0.1 - 5.0 \times 10^{12} \text{ g/yr}\) with a calculated washout of \(25\%\) during the ascent of a volcanic plume. We estimated an annual volcanic emission of \(9.5 \times 10^5 - 8.5 \times 10^9 \text{ g/yr}\) for subduction zone-related volcanoes and \(9.3 \times 10^7 - 3.1 \times 10^{11} \text{ g/yr}\) for all other volcanoes for carbonyl sulfide (OCS; Table 7). The molar ratio of OCS/\(\text{SO}_2\) varies between \(6.1 \times 10^{-4}\) and \(7.9 \times 10^{-8}\) for subduction zone-related volcanoes and between \(3.4 \times 10^{-5}\) and \(7.9 \times 10^{-2}\) for all other volcanoes. OCS is rarely measured even though it contributes significantly to the aerosol layer in the stratosphere. CS\(_2\) concentrations are generally smaller by a factor of \(3 - 10\) than those of OCS (Rasmussen et al., 1982). Thus, the annual volcanic input reaches \(1.3 \times 10^7 - 4.4 \times 10^{10} \text{ g/yr CS}_2\) (Table 7), based on the assumption that every explosive eruption is emitting CS\(_2\). This is still uncertain, however, since CS\(_2\) has only been detected in a few eruptions (e.g. Mt. St. Helens, Soufrière; e.g. Rasmussen et al., 1982). The widely ranging estimates especially of HBr, OCS and CS\(_2\) can be explained by large uncertainties in the ratios and because of the small number of observational data from active volcanoes. All of the calculated estimates of gas emissions are averages over many years.

### 5. Quantitative volcanic gas input into the stratosphere

#### 5.1. Estimates of \(\text{SO}_2, \text{HCl}, \text{HF}, \text{HBr, OCS, CS}_2\) and \(\text{H}_2\text{S}\) over the past 100 yr

An empirical analysis was carried out with recorded volcano eruptions between 1900 and 2000 based on the annual eruption frequency and on the assumption that every single eruption is a standardized outburst of ashes and gases in a vertical plume. Thus, we considered every single eruption to eject a plume into the atmosphere, rejecting flank eruptions and variable wind conditions which could blow the rising plumes sideways. We subdivided all recorded explosive eruptions into ‘injecting into the upper-atmosphere (tropopause/stratosphere)’ eruptions (\(\text{VEI} \geq 3\)) and into ‘injecting only into the lower-atmosphere (troposphere)’ eruptions (\(\text{VEI} < 3\)) (Fig. 2). Neither the effect of latitude and altitude of the volcano location were considered in this estimate, nor the specific and/or the relative humidity of the atmosphere surrounding a volcanic plume during the eruption. More than two thirds of all annual eruptions have an energy level of \(\text{VEI} < 3\) and are mainly important because of their emission of inert gases (e.g. OCS, CS\(_2\)) which can reach the tropopause or stratosphere by thermal convection in the lower atmosphere. Eruptions with \(\text{VEI} \geq 3\) are less frequent but are able to inject their gases directly into the tropopause region or even into the stratosphere. We only used \(\text{VEI} \geq 3\) eruptions for estimating \(\text{SO}_2\) emission into the stratosphere. About \(25\%\) of all eruptions reach the stratosphere.
The SO$_2$ data set between 1900 and 1971 is based on the modified VSI and was calculated for the years 1972–2000 using the collected data based on COSPEC/TOMS. A possible stratospheric input of SO$_2$ and HCl is based on an assumption of Turco et al. (1985) that the composition of a gas mixture in an eruption cloud changes after a few days because of alteration.

Fig. 2. Frequency of volcanic eruptions from 1900 to 2000, divided based on the VEI into eruptions which can reach the troposphere (VEI < 3) or the stratosphere (VEI ≥ 3). (VEI is defined on the height of the eruption column and the volume of the eruption ejecta.)

Fig. 3. Minimum and maximum sulfur (S here as SO$_2$) emissions from explosive volcanoes without quiescent degassing from 1900 to 2000, based on a modified VSI for historical eruptions from 1900 to 1971 and on directly measured volcanic SO$_2$ flux from 1972 to 2000.
Fig. 4. Estimated volcanic sulfur emission during the 20th century (S here as SO₂). Comparison of the total atmospheric S input (a) with the hypothetical total S input into the stratosphere (b) and after subtracting the scavenging effect from the hypothetical total S input into the stratosphere (c). The shaded areas indicate the deviation factor of the S emission caused by the monitoring instruments and calculation.
processes. The residual gas mixture in the altered plume is responsible for the amount remaining in the stratosphere.

The gases \( \text{SO}_2 \), \( \text{H}_2\text{S} \), \( \text{HCl} \), \( \text{HF} \), \( \text{HBr} \), \( \text{OCS} \), \( \text{CS}_2 \) finally reach the stratosphere after subtracting the washout effect in the atmosphere of the different gases. We assumed that the gases OCS and \( \text{CS}_2 \) undergo no washout during their residence in the atmosphere. We have subtracted specific washout effects only for the water-soluble gases such as \( \text{HCl} \) (Table 7).

Fig. 3 shows a nearly steady annual \( \text{SO}_2 \) emission over the past 100 yr. Two peaks, spaced 10 yr apart at the beginning of the 20th century and two to three peaks in the 1980s and 1990s reflect increased volcanic activity. The two major peaks are spaced \( \sim 80 \) yr apart, similar to the ca. 70-yr periods of increased volcanic activity suggested by Simkin and Siebert (1984). Annual \( \text{SO}_2 \) emission appears to increase from the 1950s to the year 2000. Fig. 4 shows the actual \( \text{SO}_2 \) input into the stratosphere under the assumption that approximately 25% of the \( \text{SO}_2 \) emission by explosive eruptions is already washed out by atmospheric water or scavenged in hydrometeors during the rise of a volcanic plume (Textor, 1999). The \( \text{SO}_2 \) emission prior to 1972 was calculated using a modified VSI considering the VEI of a volcanic eruption. Uncertain factors for our evaluation are the time and the location where the gas release occurs.

Fig. 5 shows the final \( \text{HCl} \) input into the stratosphere after subtracting the amount scavenged by atmospheric water. One \( \text{HCl} \) molecule out of 1000 may indeed reach the stratosphere (Turco et al., 1985; Textor, 1999). The \( \text{HCl} \) input into the atmosphere was calculated via the ratio \( \text{SO}_2/\text{HCl} \). Thus, the \( \text{HCl} \) curve follows the trend of the \( \text{SO}_2 \) curve. The total amount of emitted \( \text{HCl} \) appears to be extremely high (magnitudes of \( 10^{12} \) g/yr), but a comparably smaller amount actually reaches the stratosphere (\( \sim 10^9 \) g/yr). The \( \text{SO}_2/\text{HCl} \) ratio (Fig. 5) is a first approximation to the stratospheric \( \text{HCl} \) input. The gases \( \text{H}_2\text{S} \), \( \text{HBr} \), \( \text{HF} \) undergo a similar development. Only the strongly inert gases OCS, \( \text{CS}_2 \) are assumed to penetrate the troposphere without major loss by washout.

6. Discussion

In order to understand the impact of volcanism on the Earth’s atmosphere, the eruption frequency and outgassing rates for the entire spectrum of volcanic activity must be known. There is still a lack of continuous measurements on active volca-
noes in different tectonic environments despite 30 yr of monitoring volcanic degassing. Only few literature data exist, for example, on African volcanoes compared with data available for Italian volcanoes (in particular Etna). Thus, the major contribution of Etna volcano to the total global SO₂ emission of >10% estimated by Francis et al. (1996) decreased to 8–9% or less in our assessment; it is likely to decrease further once more volcanoes are monitored. Our evaluation of volcanic aerosol precursor gases must still be regarded as a statistical estimation on a very limited data basis, however. Although we increased the data base (monitored volcanoes by COSPEC/TOMS instruments) compared with the earlier estimates, there is still a large uncertainty in using those data. A major uncertainty is that both instruments for monitoring SO₂ emission (COSPEC and TOMS) are unable to detect degassing during the night. TOMS only monitors events with an SO₂ output >5–10 kt, but because TOMS is working from a satellite in orbit it can monitor constantly during major eruptions. COSPEC is very versatile for measurements during quiescent periods and small eruptions but must be placed close to an active volcano to produce useful results. Thus, degassing by small eruptions is detected neither by COSPEC nor by TOMS. Moreover, volcanic SO₂ emissions measured by both instruments show significant differences – up to 40%. Also, one has to consider that only 50 directly measured volcanoes exist in the world, that is ~15% out of the ~360 active subaerial volcanoes that are used in our estimate.

Another source of error is due to the irregular use of the COSPEC instrument. Quite often, SO₂ degassing is measured for short times only (days, even minutes), or just at fine weather conditions, etc. The sources of measurement errors are manifold. The lack of continuity in monitoring SO₂ emission caused a high uncertainty factor in our estimate because approximately 70–80% of the data used by us were produced with COSPEC. Only a few volcanoes such as Mt. Etna exist which have been monitored continuously. To improve estimates of volcanic gas emission, volcanoes which are monitored sporadically need to be measured more continuously. We also need to include a larger number of volcanoes in the remote sensing data, chosen on a statistical basis and not, for example, on the existence of roads directly leading to a volcanic vent (e.g. Masaya). The extrapolation of brief sporadic measurements of SO₂ degassing (days) and done by different monitoring institutions significantly influences our estimate because it is mostly impossible to attain raw data from the institutes.

The error in the extrapolated gas emissions increases proportionally to the declining number of data. An adequate, improved database can only be obtained by a significant increase in the number of monitored volcanoes. There is also a deplorable lack of standardized methods in using COSPEC measuring volcanic SO₂ flux. Future studies must focus on the development of routines in using COSPEC for monitoring SO₂ degassing. The COSPEC users should improve the accuracy of measurements of wind speed that is driving the volcanic plume because evaluating imprecise wind speeds was, and still is, one of the largest sources of error. Finally, we believe that the method we used for calculating the global annual volcanic gas emission is the most detailed estimate existing based on recently available gas data, although we had to use unreliable parameters such as VEI and the molar gas ratios.

Volcanic activity has a disproportionately large climatic impact despite low total SO₂ input compared to anthropogenic SO₂ sources because explosive volcanism has the ability to inject sulfur directly into the stratosphere. On the other hand, the effect of volcanic emission on ozone destruction caused by halogen gases (HCl, HBr) was overestimated in the past and the volcanoes were underestimated regarding their climate cooling effect induced by the injection of volcanic sulfur-rich gases directly into the stratosphere. This means that volcanoes contribute more effectively to maintain global mean temperature at the same level by sporadically compensating the global warming caused by anthropogenic gases.

7. Conclusion

We calculate total volcanic sulfur dioxide dur-
ing the last 30 yr as \(15 - 21 \times 10^{12} \text{ g/yr SO}_2\) (= 7.5 - 10.5 \( \times 10^{12} \) g/yr S). Explosive eruptive activity leads to \(9 - 12.6 \times 10^{12} \text{ g/yr SO}_2\) (= 4.5 - 6.3 \( \times 10^{12} \) g/yr S) producing approximately 60% of the volcanic \( \text{SO}_2\); quiescent degassing produces the remaining 40% (6.0 - 8.4 \( \times 10^{12} \) g/yr \( \text{SO}_2\) equals 3.0 - 4.2 \( \times 10^{12} \) g/yr S).

An apparent increase in volcanic activity over the past 30 yr (Fig. 3) is probably due to improved monitoring rather than to a real increase in volcanic activity. The differences between our estimate of volcanic gas emission compared with previous estimates primarily depends on an increased global data base. Our estimate considers the scavenging effect during the ascent of a plume, which is an important limiting factor not previously taken into account.

We achieved a higher accuracy with an increased amount of directly monitored volcanoes used in our estimate to almost 15% of the total subaerial volcanoes (explosively active during the past 100 yr) and by including significant parameters for a more precise extrapolation from directly measured \( \text{SO}_2\) emission on non-directly monitored volcanoes than were considered in the previous estimates. We compared magma composition, tectonic setting (divided into provinces) and stage of volcanic activity (silent to explosive) of the monitored volcanoes with the unmonitored volcanoes.

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References


