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Phil. Trans. R. Soc. Lond. A 2000 **358**, 1585-1606
doi: 10.1098/rsta.2000.0605

THE ROYAL SOCIETY

Integrating retrievals of volcanic cloud characteristics from satellite remote sensors: a summary

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Volcanic eruptions are events that rapidly and suddenly disperse gases and fine particles into the atmosphere, a process most conveniently studied from the synoptic satellite perspective, where remote sensing offers a practical tool for spatial and temporal measurements. Meteorological satellites offer approximately 20 years of archived data, which can be analysed for measurements of masses of SO_2 and fine volcanic ash in spatial two-dimensional arrays and integrated with other meteorological data. The satellite data offer a tool to study volcano–atmosphere interactions in a quantitative way. They provide information of unique value for understanding the fate and transport of fine silicates with significant health hazards and for addressing the problem of volcanic cloud hazards to jet aircraft. Studies of satellite data have demonstrated the following.

(1) Volcanic clouds from convergent plate boundary volcanoes contain large and variable excesses of SO_2.

(2) The second day of atmospheric residence for volcanic clouds has significantly higher SO_2 than the first, suggesting that early volcanic H_2S may be converting to SO_2.

(3) Complete conversion of SO_2 to sulphate in the stratosphere occurs at an e-folding rate of approximately 120 days. SO_2 loss from stratospheric volcanic clouds occurs at an e-folding rate of approximately 35 days, and the SO_2 loss rate for volcanic clouds which only barely reach the stratosphere is rapid (e-folding only a few days). The latter limits the stratospheric aerosol build-up from smaller eruptions.

(4) Fine volcanic ash (with diameters of less than *ca.* 25 μm) in drifting volcanic clouds retrieved after 10 h or more appear to represent a small fraction (less than 2% of the total mass) of the total mass of magma erupted, and also a small fraction (less than 20%) of the total mass of fine ash erupted. This is probably explained by the fact that the total mass is greatly reduced by aggregation processes within the volcanic cloud.

(5) The amounts of fine ash decrease faster in volcanic clouds of larger eruptions, supporting the self-removal processes suggested by Pinto *et al.* in 1989.

(6) Evidence for a strong role of ice in the fallout and aggregation of volcanic cloud ash is considerable.

(7) In many cases, volcanic clouds separate into higher SO_2-rich portions and lower ash-rich portions. The two portions follow different trajectories and the lower, ash-rich portions are affected by interactions with moist tropospheric air.

Keywords: volcanic ash; sulphur dioxide; ice; aggregation; fallout

1. Introduction

Volcanologists study active processes at the volcano and deposits of eruptions that are mostly proximal, typically within tens, and rarely as far as hundreds, of kilometres from the volcano. Meteorologists and atmospheric physicists and chemists study clouds and cloud processes on scales ranging from microscopic to global. This is a paper about volcanic clouds, a study that draws from both of these groups of scientists. The paper aims to integrate satellite remote sensing measurements on volcanic clouds and to explain some of the main scientific results that have been obtained to date.

Clouds are suspensions of particles in the atmosphere. Meteorological clouds contain particles that are mainly liquid or solid H_2O (hydrometeors), which are smaller than *ca.* 100 μm in diameter and that fall through the atmosphere in a laminar regime at velocities of less than *ca.* 0.1 m s^{-1}. Larger meteorological particles fall much faster, in the turbulent regime, and are called precipitation (Rogers & Yau 1989; Houze 1993). Because of the slow fall speeds of their particles, clouds can persist in the atmosphere for periods of hours to weeks or longer, although they are often dynamic.

Volcanic clouds are much rarer features than meteorological clouds. They are initiated by explosive eruptions that release volcanic gases and hot silicate fragments called pyroclasts and form vertical buoyant columns or plumes that rise to heights of up to 50 km as heat is transferred from the hot pyroclasts to entrained air from the surrounding atmosphere (Sparks *et al.* 1997; Gilbert & Sparks 1998). Large amounts of lower tropospheric air are entrained in these plumes (Woods 1993; Glaze & Baloga 1996; Glaze *et al.* 1997), and this air typically contains water vapour, which saturates the rising air as it rises and condenses, forming hydrometeors. Large pyroclasts fall out of the eruption column margins quickly (Ernst *et al.* 1996): lapilli (pyroclasts greater than 2 mm in diameter) fall back to Earth within less than *ca.* 30 min (Walker *et al.* 1971; Wilson & Huang 1979; Lane *et al.* 1993). Ash (pyroclasts less than 2 mm in diameter) particles fall out more slowly, and fine ash (less than 50 μm in diameter) falls out in the laminar flow regime (Rose 1993; Bonadonna *et al.* 1998) at slow velocities like the particles in meteorological clouds. So, like meteorological clouds, volcanic clouds can persist in the atmosphere for days to weeks or longer. Typically, they detach from plumes before or after the eruption stops and drift in response to the three-dimensional wind patterns (Servranckx *et al.* 1999).

Table 1. *Direct sampling studies of small particles in volcanic clouds*

volcano	date	cloud description	reference
Fuego, Guatemala	1978, 1980	small basaltic ash clouds	1, 2, 3
Santiaguito, Guatemala	1978, 1980	small dacitic ash clouds	1, 2, 3
Pacaya, Guatemala	1978, 1980	gas emission plume	1, 2, 3
Mt St Helens, USA	1980, 1981	small ash clouds, gas plumes	4
Poas, Costa Rica	1982	plume through crater lake	5
Arenal, Costa Rica	1982	gas emission plume	5
Colima, Mexico	1982	gas emission plume	5
El Chichón, Mexico	1982	stratospheric halite and sulphate	6
Erebus, Antarctica	1985	gas emission plume	7
White Island, New Zealand	1985	gas emission plume	8
Augustine, Alaska	1986	small andesitic ash clouds	9

1. Cadle *et al*. (1979); 2. Lazrus *et al*. (1979); 3. Rose *et al*. (1980); 4. Rose *et al*. (1982); 5. Casadevall *et al*. (1984); 6. Woods *et al*. (1985); 7. Chuan *et al*. (1986); 8. Rose *et al*. (1986); 9. Rose *et al*. (1988).

2. What volcanic clouds are made of

Volcanic clouds contain a variety of components including

(1) volcanogenic products from the eruption, i.e. volcanic gases, pyroclasts and aerosol particles derived from reactions of volcanogenic and atmospheric materials; and

(2) products from the ambient atmosphere, such as H_2O and gaseous species and various particles from the land and sea, including wind-blown silicates, sea salt and others.

The volcanogenic components make the clouds distinctive, and they can be tracked by satellite sensors for periods that range from minutes to weeks (Bluth *et al*. 1997; Schneider *et al*. 1995). During this time, the volcanogenic particles mix and interact with meteorological and hydrospheric particles.

Volcanogenic particles in volcanic clouds consist of fine pyroclasts, salts and acids in aerosol form. Direct sampling of volcanogenic particles has been accomplished by balloon studies (see, for example, Rietmeijer 1993) and a variety of research aircraft. Volcanogenic particles in volcanic clouds have been examined in a number of studies (table 1) where a research aircraft with a particle-collection system was flown through the clouds. Because of safety, only plumes and relatively small volcanic clouds have been directly sampled in this way. The particles consist of two main types.

1. *Silicate pyroclasts representing fragments of the magma.* These are glassy pyroclasts and minerals, which represent the crystalline fraction of the magma. Their shape is angular, and basaltic and andesitic eruptions give rise to particles that have moderate aspect ratios (Riley *et al*. 1999), while rhyolitic eruptions can generate an abundance of glassy pyroclasts with a platy geometry and extreme aspect ratios (Rose & Chesner 1987). The diameters of silicate pyroclasts generated during explosive eruptions range from metres to micrometres. Those in volcanic clouds are smaller, generally less than *ca.* 50 μm. The mass proportions of silicate particles with diameters less than *ca.* 1 μm are very small (Rose *et al*. 1980).

Table 2. *Sources of data for remote sensing retrievals used in the study of volcanic clouds*

sensor	TOMS	AVHRR	GOES
wavelengths	312–380 nm	10–12.5 μm	10–12.5 μm
orbit	polar	polar	geostationary
sensing target	ash, SO_2	ash	ash
archive	1979–present	1981–present	1996–present
scenes per day	1	4–8	48

2. *Non-silicate particles that are related to reactions among the constituents of the volcanic gases.* These particles are generally smaller than the silicates, usually less than 1 μm in diameter. The most common composition for these is sulphate, especially H_2SO_4, which forms as submicrometre spherical droplets that also contain H_2O (typically *ca.* 25% by volume; see Zhao *et al.* (1995)). A total of at least 28 different phases have also been observed (see table 3 in Rose *et al.* (1982) for a partial list) including native sulphur, sulphates, haloids, metallic oxides, and such exotic species as silver sulphide and even native gold (Meeker *et al.* 1991). Overall, the analogy between the observed phases and fumarolic incrustations and sublimates at gas vents (Stoiber & Rose 1974; Bernard 1985; Symonds *et al.* 1987) suggests that these phases originate from reactions among the volcanic gases, sometimes involving the atmosphere and volcanic silicates.

Besides these two broad types, a wide variety of other, unexplained materials has been observed in volcanic clouds. They largely consist of phases that are amorphous and have uncertain compositions (Chuan *et al.* 1987). Many or most of these particles are likely to be non-volcanic in origin, and represent accidental material of surficial or extraterrestrial origin.

Direct sampling and analysis of gases in volcanic clouds has been done only rarely (see, for example, Cadle *et al.* 1979), although analysis of CO_2 has been done much more extensively during CO_2 flux-determination surveys (Harris *et al.* 1981; Gerlach *et al.* 1999). Other information about gases has been collected from extensive airborne remote sensing of volcanic plumes using the correlation spectrometer (COSPEC) instrument. These results show that the volcanic gases in volcanic clouds are mixed and highly diluted by the ambient atmosphere, and the concentrations of volcanic SO_2 and CO_2 are less than a few ppmv (McGee 1992).

3. Methods of studying volcanic clouds

During the late 1970s and 1980s, a number of studies of volcanic clouds were made using direct sampling methods (table 1). These gave us direct data about the particles and gases. During this time recognition of the hazards to aircraft from volcanic cloud particles widened (Rose 1986; Bernard & Rose 1990), and remote sensing methods advanced. Sampling volcanic clouds directly with an instrumented aircraft is impractical because it involves risk to aircraft and because such aircraft are highly scheduled for research purposes and cannot be conveniently refitted and reprogrammed for rare, ephemeral events like volcanic clouds (Riehle *et al.* 1994). Remote sensing has developed into a very convenient study tool, because satellites designed to monitor and measure weather phenomena and global atmospheric change have the capability to

Table 3. *Data retrieved from volcanic-cloud satellite sensors, 1999*

sensor	retrieval data[a]	resolution[b]	reference
TOMS	SO_2 mass	40 km	Krueger *et al.* (1995)
TOMS AI	optical depth (UV)	40 km	Krotkov *et al.* (1999)
AVHRR and GOES	optical depth (IR); particle radius, particle mass	*ca.* 4 km	Wen & Rose (1994)

[a]In addition to two-dimensional position data.
[b]Sensor dependent Nimbus = 50 km.

map and measure many features of volcanic clouds, and have a very valuable synoptic perspective. Repetitive geometric visual observations of eruption clouds using the geostationary GOES (Sarna-Wojcicki *et al.* 1981; Holasek & Self 1995) and the GMS meteorological satellite (Sawada 1987; Holasek *et al.* 1996) have shown that satellites can map volcanic clouds of a wide variety of scales in two dimensions. This paper focuses on quantitative data retrieved from satellites concerning burdens of particles and SO_2 and sizes of particles and integrating such data with cloud mapping. The main sources of retrieval data we have used to study volcanic clouds are listed in table 2.

These sensors have been capable of volcanic-cloud sensing for variable periods: TOMS, 1979 to present; AVHRR, 1981 to present; GOES, 1996 to present (actually GOES has been operating for much longer, but only since 1996 has it used two thermal infrared (IR) bands, which allow retrievals). The result is that we have a substantial archive of volcanic cloud data, and this archive has been used to improve our capability of sensing and measuring volcanic clouds. The sensors collect from 1 to 48 datasets for any existing volcanic clouds each day. Because of their much higher frequency of data collection, the geostationary tools (currently only GOES) are most useful (Rose & Schneider 1996; Davies & Rose 1998). Unfortunately, only relatively small eruptions have been sensed by GOES, which has been in operation for only a few years. Also, NOAA has decided to suspend the two-band IR sensing on future GOES satellites, which means that the volcanic cloud-sensing option will be diminished on GOES after 2002. Fortunately, new generations of GMS, which is a geostationary satellite covering the Western Pacific, will have excellent two-band IR coverage of Earth's most volcanically active areas.

The current capability for volcanic-cloud sensing can be expressed in terms of the algorithm retrievals from the above sensors (table 3). Volcanic clouds can be mapped in two dimensions, based on both SO_2 and silicate particles. The masses of SO_2 and fine silicates (1–12 μm radius) can be estimated. The optical depth of the volcanic cloud at 10 μm and the effective radius of silicate particles is also retrievable.

4. Scientific results of volcanic-cloud studies

(*a*) SO_2 *results*

(i) *Excess sulphur from convergent plate boundary volcanoes*

Compilations of results of the mass retrievals of SO_2 (from the TOMS instrument algorithms) in volcanic clouds since 1979 were published by Bluth *et al.* (1993, 1997).

Table 4. SO_2 *and erupted magma masses from selected recent eruptions*

eruption	date	SO_2 MT[a]	magma MT	equivalent wt% S	S, melt (wt%)[b]
St Helens	May 1980	1	312[c]	0.2	0.0068
El Chichón	May 1982	7	338[d]	1.1	0.02
Ruiz	November 1985	0.66	15[e]	2.2	0.009–0.07
Pinatubo	June 1991	20	9600–13 800[f]	0.08–0.1	0.0075

[a]Bluth *et al*. (1992, 1997); [b]Scaillet *et al*. (1998); [c]Sarna-Wojcicki *et al*. (1981); [d]Varekamp *et al*. (1984); [e]Naranjo *et al*. (1986); [f]Scott *et al*. (1996).

These results show that non-arc eruptions release higher SO_2 masses than arc eruptions of the same volume, a result largely explained by the higher sulphur content of more primitive basaltic non-arc magmas (Devine *et al*. 1984). They also consistently demonstrate (see, for example, Gerlach *et al*. 1996) that convergent plate boundary volcanoes (CPBVs) release 1 to 2 orders of magnitude more SO_2 in eruptions than petrologists estimate from the volume of erupted magma and from melt-inclusion analyses (Palais & Sigurdsson 1989), which are thought to reflect the pre-eruption magma content of sulphur. This 'excess emission' of sulphur is also observed from ground-based remote sensing (COSPEC) measurements during open-vent activity at CPBVs (Casadevall *et al*. 1981; Andres *et al*. 1991). Recognition of excess sulphur from CPBVs has led to a re-evaluation of our understanding of where sulphur resides before eruption. The advocacy of a separate SO_2-rich gas phase that coexists with magma (in exsolution before eruption) has gained a consensus (Gerlach *et al*. 1996). This excess of sulphur has not been found in volcanoes from divergent or hot-spot sources (Andres *et al*. 1989), where the sulphur emission rates can be related simply to magma extrusion rates without an excess. S (and Cl) in CPBVs probably has its source in subducted slab sediments (Anderson 1974) and is perhaps supplied in excess at andesitic and dacitic CPBVs through magma mixing or incipient magma mixing. Petrological evidence for magma mixing in andesitic volcanoes is abundant (see, for example, Eichelberger 1975; Halsor & Rose 1991). Even when such evidence is not obvious, there may be evidence for a basaltic heat source transmitted to andesite, which could also supply excess gas (Murphy *et al*. 1999; Barclay *et al*. 1998). One important issue about excess sulphur releases is that they are highly variable, and they all represent proportional concentrations of magmatic sulphur (see column 4 in table 4) that are much higher (1–3 orders of magnitude) than would be expected from petrological data (see column 5 in table 4).

The 1982 El Chichón eruption released seven times more sulphur than the 1980 Mt St Helens event (Bluth *et al*. 1997), although its eruption volume was only slightly (*ca*. 10–20%) higher. El Chichón released *ca*. 35% as much sulphur as did the Pinatubo 1991 release, although Pinatubo was an order of magnitude or more greater in magma volume (Scott *et al*. 1996). The variability of excess sulphur has been shown by Scaillet *et al*. (1998) to be associated with redox controls, wherein reduced magmas with sulphides do not release large sulphur excesses. Excess volcanic gas information is related to volatile saturation and its significance remains undigested in our consideration of how volcanoes work, because how these excesses affect the nature of eruptions has not been explored. If excess gas information can

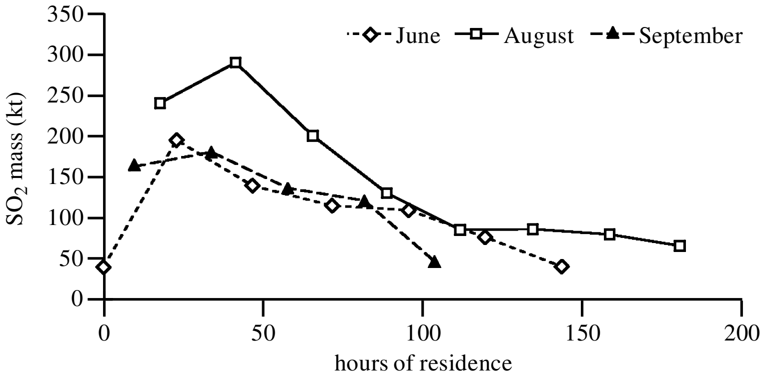

Figure 1. Masses of SO_2 in the 1992 Spurr eruptions (from Bluth *et al*. 1995). Plot resembles the trends shown for other eruptions, with SO_2 showing a mass increase in the second day (see text for discussion).

be understood, it offers a potential window into processes that happen in convergent plate boundary magma bodies (see, for example, Harris & Rose 1996).

(ii) *Possible co-emission of* H_2S *and* SO_2 *in explosive eruptions*

TOMS measurements are made only once a day, but most of the eruptions studied show higher SO_2 on the second day than they do on the first (Bluth *et al*. (1995); and see also figure 1). In the Hudson eruption of 15 August 1991, studied in detail by Constantine *et al*. (2000), the second day SO_2 mass was three times larger than the first. This difference cannot be explained by the continuing emission of SO_2, because the first day's measurement occurred after the end of the eruption. It is unlikely to reflect an error in TOMS data analysis (Krueger *et al*. 1995), which would be far less than the observed difference. It is unlikely that the TOMS detector was saturated or suppressed by an interference from volcanic ash in the SO_2 signal, because simulations of this effect would probably result in an overestimate on the first day rather than an underestimate (Krueger *et al*. 1995). The favoured explanation is that the mass increase appears to be the co-emission and subsequent oxidation of H_2S. This possibility has been suggested by Bluth *et al*. (1995), although the nature of the TOMS data left much uncertainty. The conversion of H_2S to SO_2 in the atmosphere, although poorly constrained for volcanic cloud conditions, is thought to be roughly an order of magnitude faster than the conversion of SO_2 to H_2SO_4 aerosol (Graedel 1977). Therefore, it is consistent with the TOMS mass retrievals to find such an increase from one day to the next. Determination of an H_2S component could be important, as this could indicate hydrolysis of SO_2 to aqueous H_2S by a liquid-dominated magmatic system, as proposed by Doukas & Gerlach (1995) for the Mount Spurr, Alaska eruptions of 1992. They suggest that SO_2 'scrubbing' by this mechanism could prevent (predictive) pre-eruptive SO_2 degassing from being detected by routine monitoring. Also, because the proportions of H_2S and SO_2 in volcanic gases reflects the oxidation state, the presence of H_2S could be related to the sulphur excess (see above).

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(iii) *Rates of processes related to the fate of atmospheric* SO_2 *tested*

Bluth *et al*. (1997) applied a simple model to TOMS data to test the rates of conversion of SO_2 to sulphate aerosol. They used a rate based on the decrease of SO_2 in the Pinatubo volcanic cloud, largest in the TOMS archive and corresponding to an e-folding time of 35 days, which is similar to other satellite-derived data on SO_2 removal rates (30–40 days) (see table 2 in Bluth *et al*. (1997)). This rate is four times faster than the rate of sulphate build-up in the stratosphere following Pinatubo and El Chichón eruptions, however. This lag in build-up of sulphate (about 120 days e-folding) is thought to reflect multiple reactions in the conversion process. The conversion of SO_2 to sulphate is the result of a series of reactions, while the TOMS measurements of SO_2 only reflect the first of these, which is the destruction of SO_2. Thus, one of the later reactions must be rate limiting in this case. Rates of removal of SO_2 for smaller eruptions are much faster than 35 days e-folding, however, often being only a few days (Bluth *et al*. 1997). This faster rate of removal is also associated with very little stratospheric aerosol build-up, which suggests that eruption columns that do not rise much higher than the tropopause (with volcanic explosivity index not more than 4; see Newhall & Self (1982)) are subject to highly efficient self-removal processes (Pinto *et al*. 1989). Conversely, it is thought (Bekki *et al*. 1996) that gas-to-particle conversion may be considerably slower when there has been extremely high stratospheric loading (e.g. the Toba scenario), but as yet we have no satellite data to measure an eruption with this large a scale.

(*b*) *Ash results*

(i) *Masses of very fine particles*

The two-band IR algorithms used on GOES and AVHRR detectors only sense particles between *ca*. 1 and 12 μm in radius. This provides important data on those materials that have high surface area/mass and which can therefore potentially catalyse atmospheric reactions. It also provides an insight into the fate and transport of those volcanic particles with potentially the greatest impact on health, because recent reports have tended to emphasize the significance of particulate material with diameters less than 10 (PM10) or 2.5 μm (PM2.5). Health standards for PM10 and PM2.5 have been revised downward recently, reflecting the discovery that particularly small particles are not caught in the oesophagus and enter the lungs. Studies of dust hazards in Idaho (Norton & Gunter 1999) have demonstrated the effects of distal ashfall on health. They show that the PM10 and PM2.5 splits of fine particles on Idaho farms (600–900 km from Mt St Helens) are dominated by Mt St Helens ash a decade and more after the 1980 eruption.

Infrared satellite data offer a tool for studying the fate and transport of fine ash erupted in explosive eruptions. The satellite measurements appear to show that the masses of fine ash in volcanic clouds are less than a few per cent of the total mass of ash produced in the eruptions (table 5). This percentage is probably much less than the total percentage of fine particles in the eruption, because we know that large amounts of fines fall out as part of aggregated materials in ash blankets near the vent. For example, a fallout sample from Wells Bay, Alaska (only 300 km from the Crater Peak vent of Mount Spurr), which erupted in August 1992 (McGimsey, personal communication; Riley *et al*. 1999), contains *ca*. 25% of ash finer than 10 μm

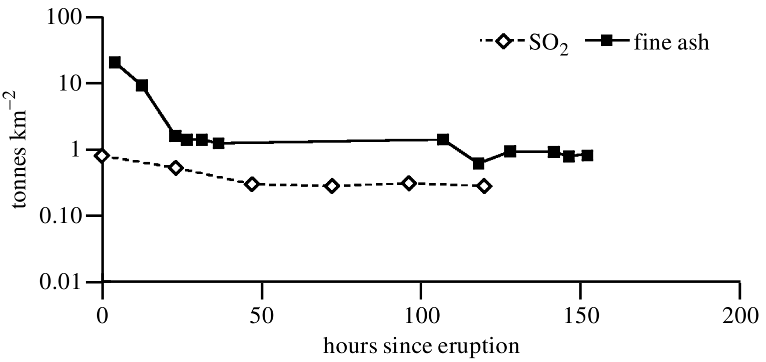

Figure 2. Evolution of volcanic cloud total mass/unit area (a measure of average volcanic cloud density) as measured by TOMS and AVHRR data for the June 1992 Spurr eruption (from Shannon 1997). The plot resembles the trends shown for other eruptions with rapidly decreasing ash density in the first 36 h, followed by similar slow decreases in both ash and SO_2 after that (see text for discussion).

Table 5. *Fine ash* (*radius* 1–12 µm) *masses in volcanic clouds from satellites*

volcano	date	total ash erupted ($\times 10^6$ t)	maximum fine ash ($\times 10^6$ t)	%	height (km)[a]
Spurr	June 1992	21.1[b]	0.44	2.1	14.5
Spurr	August 1992	21.3[b]	0.42	2.0	13.7
Spurr	September 1992	23.3[b]	0.61	2.6	13.9
El Chichón	April 1982	910[c]	6.5[c]	0.7	32
Láscar	April 1993	345[d]	4.8[e]	1.4	18
Hudson	August 1991	7600[f]	2.9[g]	0.04	18

[a]Maximum column height observed (above sea level).
[b]Neal *et al*. (1995); [c]Schneider *et al*. (1999); [d]Viramonte (1995); [e]Shocker *et al*. (2000); [f]Scasso *et al*. (1994); [g]Constantine *et al*. (2000).

in diameter (PM10), and *ca.* 1.5% of ash finer than 2.5 µm in diameter (PM2.5). Also the masses of fine ash, as measured by satellite, decrease rapidly in the first 1.5 days and then decrease much more slowly (figure 2). From this we conclude that as much as 75–90% of fine ash falls out of volcanic clouds in the first 36 h, and is deposited in ash blankets and, sometimes, within regions of secondary thickness maxima of ash blankets. Only a small fraction of this fine ash continues with the volcanic cloud.

(ii) *Ash fallout patterns described*

Fallout of silicates from volcanic clouds can be quantified with remote sensing data. The fallout of large particles (greater than 1 mm in diameter) in the turbulent-flow regimes can be observed by ground-based weather radar systems, which are optimized for large particles (precipitation) (Harris *et al*. 1981; Harris & Rose 1983; Rose *et al*. 1995). The radar data depict the rapid fall of materials that deposit in the proximal ash blankets, and *ca.* 30 min after eruption the C-band radar signal returns to background, because only particles with diameters less than *ca.* 2 mm are still airborne (Rose *et al.* 1995). During the first three hours or so after the eruption stops, the volcanic cloud may contain enough particles to be optically opaque in the IR (Schneider *et al.* 1995). The mass measurements of fine particles in volcanic clouds determined from IR data decrease rapidly for *ca.* 36 h after eruption (figure 2), even though these particles will not fall out rapidly as single particles (note that a simple spherical silicate ash particle with a diameter of *ca.* 25 μm would fall out from 14 km in 36 h). This is viewed as direct evidence for aggregation, and the rates of mass decrease are greater for large eruptions (see figure 4).

A key observation from table 5 and figure 4 is the apparent negative correlation between eruption intensity and the wt% of fine ash retrieved in the cloud: lower mass fractions of fine ash remain in clouds from big eruptions. This can be rationalized as follows.

(1) More intense eruption columns characterized by higher upward velocities are more efficient at re-entraining particles than low-intensity eruptions columns (Ernst *et al.* 1996).

(2) More intense eruptions also have higher eruption rates, so that the volume of fragmented ash is higher.

(3) More fragmentation results in more electric charge generated in the volcanic conduit by fracto-emission (Lane *et al.* 1993).

(4) Higher columns also entrain more moist air (Glaze *et al.* 1997) and experience higher temperature gradients leading to the formation of hydrometeors (Rose *et al.* 1995; Herzog *et al.* 1998), resulting in further charge generation by processes analogous to those that generate electric charges in thunderstorms (Black & Hallett 1998).

(5) As a result of items (1)–(4), more intense eruption columns result in more efficient particle removal by ash aggregation and premature fallout of aggregates as icy pyroclasts (Rose *et al.* 1995).

This is viewed as support for self-removal processes as envisioned by Pinto *et al.* (1989), and demonstrates that aggregation probably happens in the coarse proximal fallout as well as farther from the vent.

After *ca.* 36 h, the apparent ash mass loading of volcanic clouds does not change greatly, and the clouds may drift for several more days in the prevailing winds at various levels. The fine ash that remains in the cloud is only a small fraction (less than 20%) of the maximum detected, and the burden of fine particles, which has fallen to about one-fifth of its maximum after 36 h, follows a trend that is similar to SO_2 from then on (figure 2). Studies of the trajectory of the June 1992 Spurr clouds by Shannon (1997) has shown that the lower parts of these clouds thin greatly and become undetectable after ash falls, presumably because ash particles are scavenged by hydrometeors (see also Rose *et al.* 1995; Textor 1999). Thus, the final fate of the remaining fine ash is closely related to the local weather of the upper troposphere (see also Bursik 1998).

(iii) *Meteorology and volcanic clouds*

Remote sensing of the Rabaul eruption clouds of 1994 showed that the stratospheric cloud contained much ice (up to 200–300 Mt), a result explained by the

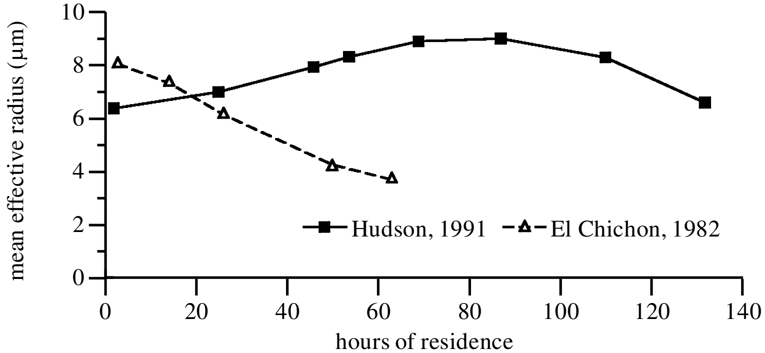

Figure 3. Mean effective radius of volcanic clouds of El Chichón in 1982 (Schneider *et al*. 1999) and Hudson in 1991 (Constantine *et al*. 2000) as a function of time, measured using AVHRR data. The sizes of El Chichón silicates decrease with time, while those for the Hudson do not (see text for discussion).

strong interaction of sea water with the erupted magma (Rose *et al*. 1995). This study heightened the awareness of meteorological processes in volcanic clouds, and new model simulations have demonstrated that ice is likely to be abundant at levels above *ca.* 6 km in eruption clouds, even when there is no seawater interaction (Herzog *et al*. 1998). This prediction is consistent with experimental data (Rogers & Yau 1989) on heterogeneous nucleation of ice on volcanic ash showing nucleation from temperatures of −13 °C (*ca.* 5 km in height). Recycling of icy particles within the eruption column on a time-scale of less than 1 h can allow ice–ice nucleation at even lower levels (Lane-Serff 1995). In the absence of external ground or seawater involvement, ice originates within the eruption column because of entrainment of moist lower tropospheric air that rises, condenses and freezes (Woods 1993; Glaze & Baloga 1996; Glaze *et al*. 1997).

The simulation models of eruptions that include microphysical processes (Herzog *et al*. 1998; Textor 1999) show that icy hydrometeors will form from fine ash nuclei in volcanic clouds, but these hydrometeors may be aggregates with an ash content of above 80 wt% (Textor 1999). This may explain why volcanic clouds usually show silicate IR signals with negative brightness temperature difference (BTD) (10–11 μm BTD) because of the high wt% of silicate even in the icy ashballs. However, the effective radii of many drifting volcanic clouds are relatively large and increase with time, especially when there are abundant high meteorological clouds present, as was the case for the Hudson eruption (Constantine *et al*. 2000; figure 3). We interpret this to be the result of the formation of hybrid ash–ice hydrometeors (icy ashballs), which still have the spectral signal of ash. Thus, we can infer that ice possibly plays an important role in fallout, particularly in the first 36 h in the life of the volcanic cloud.

Significantly, in the model studies that consider tropical atmospheric conditions (Herzog *et al*. 1998), the ice often disappears (melts and evaporates) before the ash reaches the ground, a fact that explains why direct evidence for ice in ash blankets is elusive. There is the potential that ice can be preserved on the ground in some conditions (i.e. phreatoplinian eruptions at high latitudes). This was illustrated during the late November 1963 eruptions at Surtsey, Iceland, where the fallout of icy

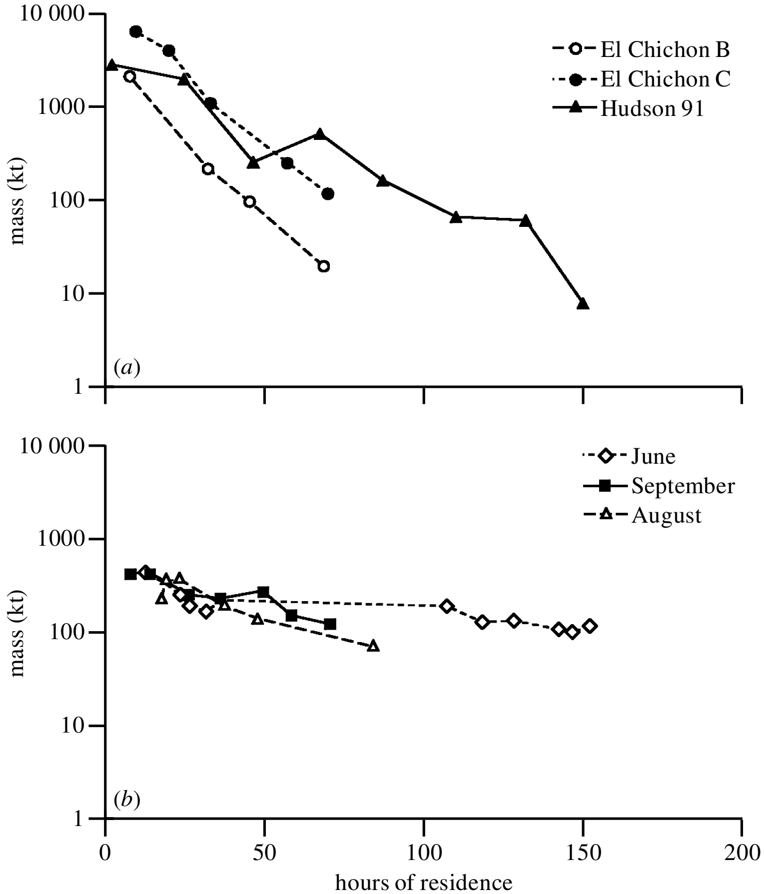

Figure 4. Two plots comparing the rates of total retrieved mass decreases for larger volcanic clouds (El Chichón and Hudson, see above) with smaller ones (three 1992 Spurr clouds, see below). The smaller clouds decrease at a much slower rate (see text for discussion).

pyroclasts onto local ships was described as hail showers with a grain of ash within each hailstone (Thorarinsson 1966).

Understanding the meteorology is essential in interpreting the data, because the occurrence of high clouds, such as those that form above the Andes in the roaring forties (Constantine *et al*. 2000), can create difficulties for the IR remote sensing measurements. The spectral signal of ice in volcanic clouds is dominant when there is a hydrospheric source of water in addition to the atmospheric entrainment of water (Rose *et al*. 1995). In addition to the Rabaul example (above), the pyroclastic flow eruptions of Soufrière Hills, Montserrat frequently reach the sea and exchange much of their heat with seawater. The rise of this water vapour creates meteorological clouds that interact and rise together with the volcanic ash clouds and increase the potential for hydrometeor formation and turbulent convection (Mayberry *et al*. 2000). This incorporation of large amounts of hydrospheric water into the volcanic cloud may be quite important because there are many volcanoes near the ocean or lakes, and the latent heat contributions can change the character of the volcanic cloud

Table 6. *Examples of gas–ash separations of volcanic clouds*

eruption	date	SO_2 height (km)	ash height (km)	reference
El Chichón	April 1982	22–26	19–21	1
Hudson	August 1991	14–18	10–14	2
Láscar	April 1993	> 18	12–18	3
Soufrière Hills	December 1997	*ca.* 15	4–14	4

1, Schneider *et al.* (1999); 2, Constantine *et al.* (1999); 3, Shocker *et al.* (2000); 4, Mayberry *et al.* (2000).

greatly. When hydrometeors become dominant, the eruption cloud may resemble a giant thunderstorm, or, in the extreme cases of megaeruptions into a lake or the sea (e.g. Toba, 75 ka or Krakatau, 1883), the results could generate a hurricane or even a hypercane (the runaway hurricane of Emmanuel *et al.* (1995)).

(*c*) *Integrated data*

(i) *Separation of gas and ash*

Use of both IR and UV sensors in studies of the same volcanic clouds enables more holistic studies. One highlight of the studies of this type (see, for example, Rose *et al.* 1995; Schneider *et al.* 1999; Shannon 1997; Constantine *et al.* 2000; Shocker *et al.* 2000; Mayberry *et al.* 2000) is that many eruptions exhibit separation of SO_2 and volcanic ash, typically almost immediately after eruption. The SO_2 ends up higher in the atmosphere than the ash and is then moved by higher-level winds in a different direction and at different velocities than the ash (table 6). Of the comprehensive data studied to date, only the relatively small Spurr eruptions do not show this separation.

In all cases where separation is observed, the SO_2 and ash-rich clouds are apparently already well separated vertically on the first satellite image available. The effect of windshear is to make the separation of the two clouds evident without any further vertical splitting apparent for either cloud as time goes by (Schneider *et al.* 1999). The causes of this separation and its implications are mostly unexplored. Since the SO_2 comes largely from an exsolved gas phase before eruption (see above), it may be that it separates largely from the ash during eruption, and it may even be released selectively before much of the ash. This scenario is consistent with measurements of electrical potential field during explosive activity at Sakurajima Volcano, Japan (Lane & Gilbert 1992). At Sakurajima, a gas-rich cloud precedes an ash-rich cloud during the vulcanian eruptions documented. It may be that the sedimentation of ash within the umbrella cloud that forms in the upper parts of the drifting volcanic cloud dynamically triggers separation in a fluid dynamic process (Holasek *et al.* 1996). However, we realize that the experiments of Holasek *et al.* (1996) are inconclusive because the simulated process of particle–fluid separation would be expected on the same time-scale that the effects of the tank walls induce a return flow in the tank and force the breakdown of particle–fluid separation. The process envisioned by Holasek *et al.* (1996), however, is also suggested by tropical volcanic-cloud simulations that incorporate hydrometeors (Herzog *et al.* 1998), and the presence of hydrometeors is expected to enhance the process by accelerating the sedimentation of ash. A third

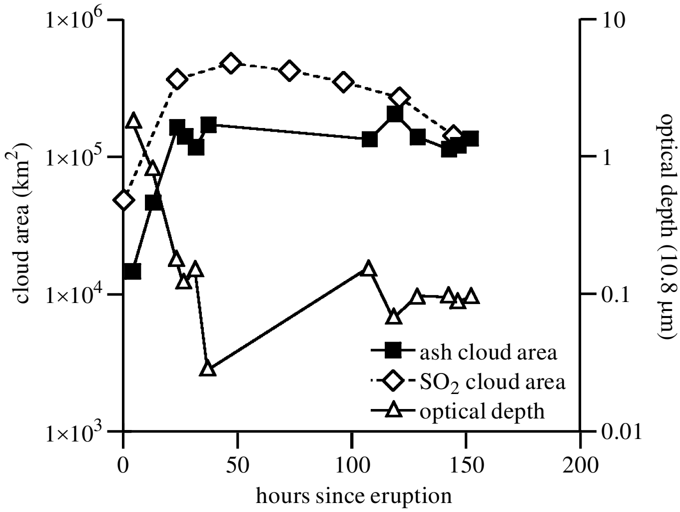

Figure 5. Volcanic-cloud-area and optical-depth measurements plotted against time for the June 1992 Spurr event (from Shannon 1997). Ash-cloud area and optical depth measured from AVHRR and SO_2 area measured from TOMS data (see text for discussion).

process that could explain the separation is scavenging or absorption of SO_2 by ice in the cloud, especially at levels below the top. This process was suggested for the Rabaul eruption by Rose *et al*. (1995) and is consistent with most of the other cases in table 6. It differs from the other explanations because it involves removal of SO_2 rather than dynamic separation. We are optimistic that we can test these hypotheses by continuing our remote sensing and experimental studies.

(ii) *Morphology and dynamics of volcanic clouds*

Figure 5 shows an example of area measurements of volcanic clouds. Data on cloud shapes, widths and distances are also readily available along with changes with time. The possibilities for studies of the sizes and shapes of volcanic-cloud sequences to define the cloud-spreading dynamics has already been demonstrated by Sparks *et al*. (1986, 1997) and Volon (1997). A discussion of some of the research issues and processes of volcanic plume or cloud-shape changes (e.g. draughting and bending) was given by Bursik (1998), while preliminary studies of plume bifurcation have been made by Ernst *et al*. (1994) and Rose *et al*. (1995). Photoinclimetric studies of satellite data have been used to extract three-dimensional patterns of dispersion and movement (Glaze *et al*. 1999), while Shannon (1997) and Mayberry *et al*. (2000) have combined the UV and IR satellite data (table 3) with trajectory analysis to develop three-dimensional patterns. Evidence for thermal disequilibrium for eruption clouds has also been documented from satellite data, with implications for eruption column models (Woods & Bursik 1991; Woods & Self 1992). Geometrically corrected satellite images are used to derive the lateral spreading of umbrella clouds (Sparks *et al*. 1986) as well as cloud radial velocities (see, for example, Volon 1997), and then these datasets are compared with theoretical-model predictions (Bursik *et al*. 1992; Woods *et al*. 1995; Sparks *et al*. 1997). Comparison between theory and lateral spreading data for the 18 May 1980 Mt St Helens cloud demonstrated that the cloud spreading was dominated by gravity flow, advection of the cloud by wind, and outfalling particles within the first few hours of spreading, with atmospheric diffusion playing a very minor role (Sparks *et al.* 1997). This is in direct contradiction with the suggestion of Heffter (1996) that advection–diffusion models could adequately describe this stage of spreading. However, such advection–diffusion models can be useful to describe the second stage of spreading, after the turbulence level in the volcanic cloud becomes dominated by atmospheric turbulence (see Bursik 1998).

5. Potential improvements

As satellite detectors evolve and improve, as the algorithms used to retrieve data develop, and as more users learn to interpret the data, we expect that our ability to study volcanic clouds will improve. The following is a list of some of the major improvements that are now within reach.

(*a*) *Cross validation of methods*

We have only very occasionally the opportunity to study TOMS and GOES or AVHRR data collected at exactly the same time. One such example is one of the Spurr volcanic clouds studied by Krotkov *et al.* (1999). This comparison helps us improve the algorithms used in retrievals and also to understand the environmental variables that sometimes prevent full retrieval of IR data. Validation of volcanic-cloud retrievals is crucially important, especially for the hazards considerations regarding public heath and fine particles and ash hazards to aircraft. Although we hope for direct sampling studies for validation (see, for example, Riehle *et al.* 1994), it is logistically difficult to arrange them.

(*b*) *Geostationary positioning of detectors*

The availability of GOES two-band IR data clearly demonstrates the advantage of the stare mode in the study of rapidly changing volcanic clouds (Rose & Schneider 1996; Davies & Rose 1998). Unfortunately, this capability will be lost in 2002 and the TOMS has never been placed on a geostationary platform. A NASA proposal (VOLCAM) to demonstrate the use of a geostationary platform with both UV and IR detectors is currently being negotiated among US agencies for possible funding, which could result in favourable positioning of one or both of the detectors on a geostationary platform in about 2003. This would result in much better temporal resolution of integrative satellite data on volcanic clouds, and a huge increase in data for detailed studies.

(*c*) *Volcanic-cloud-height determinations*

Currently, we use trajectory analysis to constrain the heights of volcanic clouds but do not have an independent way of determining the altitude of the cloud top. The TOMS instrument can potentially determine cloud top pressure by using the 'ring effect' (filling-in of solar Fraunhofer lines by rotational Raman scattering). The algorithm compares radiance differences between cloud and clear areas within the Fraunhofer lines to those in a flat spectral region. Joiner & Bhartia (1995) have demonstrated that this technique can make reliable estimates of effective cloud top pressure using the Ca, H and K lines in the solar spectrum. Future designs of TOMS instruments, such as the one planned for VOLCAM, will initiate this ability, which will help our three-dimensional understanding of volcanic clouds.

(*d*) *Improvements in the IR detection of volcanic clouds*

Unlike the TOMS detector, which has not shown any false alarms, the two-band IR detectors can give erroneous or misleading results, especially when the atmospheric conditions are not ideal. High clouds beneath the volcanic cloud (Constantine *et al*. 1999) reduce the thermal contrast between the volcanic cloud and the subsurface radiator. In the first few hours of volcanic clouds, high optical depth prevents retrieval of particle size and mass data (Schneider *et al*. 1995). When the lower atmosphere is moist, as is the case for most tropical eruptions, such as Montserrat, the high water vapour absorption of IR wavelengths produces a shift of BTDs unless the differential absorption effects are corrected for. This latter problem has been targeted for correction and can be effectively minimized if information about the water vapour content of the first 3 km of atmosphere is known (Yu & Rose 2000). We expect that two-band algorithms can improve these atmospheric corrections markedly.

In the very near future there will be new capability for volcanic clouds as part of EOS AM-1, launched in December 1999. The moderate resolution imaging spectrometer (MODIS) detector, with improved spatial resolution (250–1000 m) and multispectral IR bands, including coverage of the critical 10–12.5 μm region, will offer great opportunities for improvements in ash retrievals. It will also enable sensing of SO_2 (Realmuto *et al*. 1997) and sulphate aerosols (Yu & Rose 2000). ASTER (advanced spaceborne thermal emission and reflection radiometer) is another EOS instrument with even better spatial (15–90 m) and spectral resolution in the IR than MODIS, and it will allow very-high-resolution coverage of a small number of volcanic clouds. Although more vulnerable to environmental conditions that adversely affect accurate detection and retrievals, IR data are available during the night-time, when UV detectors will not work. We expect that IR will always be a significant part of volcanic-cloud monitoring.

(*e*) *Early sulphate in volcanic clouds*

Using a multispectral IR method, Yu & Rose (2000) have shown that SO_2 is partly converted to sulphate aerosol during the first 24 h of atmospheric residence. This conclusion is also consistent with the observation of scavenged sulphate on fresh volcanic ash (Rose 1977) and with the observational data on volcanic clouds summarized above and in table 1. The rate of sulphate formation is consistent with the rate of SO_2 mass decreases in volcanic clouds such as El Chichón (Bluth *et al*. 1997), and suggests that the catalysis of sulphate formation resulting from the presence of large amounts of volcanic ash early in volcanic clouds is minimal. The presence of sulphate aerosols is potentially vital for the formation of aggregates, because, by analogy with reactions associated with fumarolic incrustations (Stoiber & Rose 1974; Symonds *et al*. 1987), they can react with the silicate surfaces and form an effective chemical cement that enhances aggregation. Alternatively, chemical reaction is not required to bind particles: mere evaporation of sulphate/chloride solution films are expected to precipitate salts that can bind the particles (Gilbert & Lane 1994).

(f) Interaction of ash and gases

We have already discussed the accumulation of sulphate aerosols on ash particles and the role of ice that forms on ash surfaces and scavenges gases such as SO_2 and HCl (Tabazedeh & Turco 1993), but direct adsorption of gases such as SO_2 on ash particles may also be important (Oskarsson 1980). We have begun studies of this process in laboratory experiments (Gu *et al.* 1999). Like many of the processes that occur in volcanic clouds, we can evaluate and study them better if we can get improved data frequency, particularly in the UV.

(g) Widespread use of satellite data on volcanic clouds

The widespread use of volcanic-cloud remote sensing is likely because of its potential to reduce the hazards of volcanic clouds to aircraft (Casadevall 1994). However, this application will require technology and training to be regionalized and placed in centres around the world near the volcano observatories, weather stations and airports affected. In recent years, this work has been proceeding in the regional volcanic ash advisory centres (VAACs).

This research is supported by the National Science Foundation (EAR 9725682) and by NASA. W.I.R. was supported by a Leverhulme fellowship at the University of Bristol during the preparation of this manuscript. G.J.S.B. was supportedm by SENH, NASA. Support of the USGS through the Alaska Volcano Observatory was also important. G.G.J.E. acknowledges support from the Nuffield Foundation (NAL award). Tom Casadevall helped improve the manuscript.

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