## **REMOVAL PROCESSES OF VOLCANIC ASH PARTICLES FROM THE ATMOSPHERE**

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## **INTRODUCTION**

The use of satellite techniques provides valuable information for mapping ash hazards, as well as the means to study and predict the fates of volcanic clouds. We have used ultraviolet (TOMS - Total Ozone Mapping Spectrometer) and infrared (e.g., AVHRR - Advanced Very High Resolution Radiometer; HIRS2 – High Resolution Infrared Radiation Sounder/2: GOES -Geostationary Operational Environmental Satellite; MODIS – Moderate Resolution Imaging Spectroradiometer) satellite sensors to examine the solid, liquid and gas species in numerous volcanic clouds over the past 25 years. Each sensor provides a somewhat different perspective of volcanic clouds, depending on their spatial, spectral and temporal resolutions. Thus, when combined these techniques provide important constraints on the interactions and fates of species within the clouds. Quantitative infrared techniques can provide information on how the size, size distribution, and total mass of fine ash particles evolve as a cloud mixes with the atmosphere. Here we present a review of past relevant observations and research, and the results of ongoing satellite studies.

## **VOLCANIC CLOUD PARTICULATES**

Volcanic clouds are typically composed of a variety of particulates, derived from:

-volcanogenic sources (silicate particles, erupted gases and liquids)

-the atmosphere (water/ice, dust, sea salt, gases)

-products from volcano-atmosphere reactions (aerosols, coatings, adsorbed species on existing particles)

#### (1) Particle compositions

-silicates: glassy pyroclasts and minerals, which represent the crystalline fraction of the magma. Their shapes are highly angular, and in fact can be composed of shards, bubble fragments, as well as microcrystals. The silicate particles reflect the magmatic origins, ranging from rhyolitic (high silica) to basaltic (low silica).

-non-silicates: the most common type is sulfate aerosols. Many other minor and rare phases have been observed (Rose et al., 1982).

#### (2) Shapes and Sizes:

A thorough examination of particle size, geometry, and composition by Riley et al. (2003) revealed:

-extremely high surface areas relative to volume, due to roughness and vesicularity (up to 2 orders of magnitude greater areas than calculated using shape alone).

-aspect ratios of 1.5 - 2.6 (long-to-short 2D dimensions, compared to 1 for a perfect sphere). See Figure 1.

Erupted materials are composed of a full spectrum of particle sizes, which quickly becomes differentiated through gravitational settling and physical interactions in the clouds. Based on satellite studies, about 1-2% of the total ash mass erupted is 1-12 microns (\_m) in radius (Rose et al., 2001), which is the fraction which can be most easily tracked by satellite.



**Figure 1.** SEM photos of (l-r) Fuego, Guatemala basaltic ash; Mt. Spurr, Alaska andesitic ash; bubble wall shards from the rhyolitic ash of the Ash Hollow Member, Nebraska (Riley et al., 2003). Scale bars are 1000\_m for Fuego, 100\_m for Mt. Spurr and Ash Hollow Member.

# THREE STAGES OF VOLCANIC CLOUD EVOLUTION

Rose et al. (2001) developed three general stages based on satellite observation of evolving clouds from the Mt. Spurr (Alaska) eruptions in 1992:

## (1) High energy and growth

During the eruption and for the first few hours afterward, the clouds resemble thunderstorms, and are often opaque to IR sensors. Most of the coarse (>50 microns diameter) material falls out near the vent source (Figure 2).



**Figure 2.** This simple calculation (after Bonadonna et al., 1998) shows that most large (>50 micron diameter) particles will fall out of a volcanic plume within the first day at rates of around 0.8 km/hr; on the other hand, very small particles (<1 micron) could persist in the atmosphere for years. "Particles" may be composed of a single fragment; aggregates of various sizes; particles with coating of water or ice; particle with adsorbed gas or liquid. Clearly, the shape and density of a particle or group of particles will affect how it falls (laminar or turbulent), and how fast it will fall, but this calculation gives a rough estimate, assuming laminar flow and high latitude atmospheric conditions.

### (2) Rapid physical and chemical changes

Lasting approximately one day, the cloud expands in areal extent, but the optical depth and fine-particle size concentrations decrease rapidly (by 1 or more orders of magnitude). Fine ash is rapidly removed from the cloud, most likely by aggregation or as icy ash balls (see next section).

#### (3) Drifting aircraft hazard

This stage lasts 3-5 days, during which the cloud can move thousands of kilometers. Ash

concentrations and optical depths decrease very slowly, and ash masses slowly decrease to below sensor detection limits.

#### PARTICLE REMOVAL RATES

Table 1 compares the removal rates of ice and ash for several different eruptions. The measurement periods vary owing to instrument

Table 1. Ash and ice removal rates measuredby satellite

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	Period	type	Removal	(nrs)		
	(hrs after		Rate			
	eruption)		(kt/hr)			
El	5 - 68	ash	34	13	AVHRR	а
Chichón,						
Mexico						
(1982a)						
El	7 - 70	ash	99	15	AVHRR	а
Chichón.						
Mexico						
(1982b)						
Pinatubo	5 - 111	ach	182	24	HIRS/2	h
Philippines	5-111	asii	402	24	11110/2	U
(1001)						
(1991)	7 111		010	20		1
Pinatubo,	5 - 111	ice	819	30	HIRS/2	b
Philippines						
(1991)						
Pinatubo,	6 - 104	ash	363	27	AVHRR	b
Phillipines						
(1991)						
Pinatubo,	6 - 104	ice	648	27	AVHRR	b
Philippines						
(1991)						
Hudson	2 - 132	ash	21.8	30	AVHRR	c
Chile	2 152	usii	21.0	50	21 V 111(1)	Ũ
(1991)						
(1991)	12 152	<b>1</b> -	2.2	1.4.2	AVIIDD	L
Spull,	15 - 152	asn	2.3	145	Ανπκκ	a
USA (Jun						
1992)						
Spurr,	14 - 84	ash	3.7	43	AVHRR	d
USA (Aug						
1992)						
Spurr,	8 - 70	ash	4.9	52	AVHRR	d
USA (Sept						
1992)						
Hekla	6 - 24	ice	48	8	AVHRR	d
Iceland				-		
(2000)						
Cleveland	6 - 20	ash	16	10	GOES +	P
	0-20	asii	1.0	10		C
(2001)					MODIS	
(2001)						

a) Schneider et al., 1999; b) Guo et al., 2004b; c) Constantine et al., 2000; d) Rose et al., 2001; e) Gu, 2004.

temporal resolution, and the period reflects the time between when ash decrease is measured to the end of sensor detection. While this represents only a small portion of volcanic eruptions, it appears that the large events (El Chichón, Pinatubo, Hudson) exhibit fast removal rates of ice or ash and consequently much shorter residence times than the smaller events (Spurr, Hekla). However, note the exception from the Cleveland event, which was small, yet underwent rapid removal. In contrast to ash cloud decrease (e-folding, which is the time for the cloud to reduce to 1/e of its original mass), sulfur dioxide gas e-folding rates are on the order of 2 - 25 days, rather than hours (e.g., Figure 3).



**Figure 3.** Mass removal patterns of ash, ice and sulfur dioxide in the 1991 Pinatubo volcanic cloud (Guo et al., 2004a; 2004b).

#### **REMOVAL PROCESSES**

#### (1) Ash/ice hydrometeors

Satellite studies find that the radii of suspended particles can *increase* over the first 36 hours after eruption, which we interpret as indicative of ice formation on ash particles (Rose et al., 2000). Modeling studies suggest that these aggregates may be over 80% ash by weight (e.g., Herzog et al., 1998; Textor, 1999).

## (2) Particle aggregation (sticking)

The rate of fine ash removal during the first 1-2 days shows a rapid decrease (Figure 3), which cannot be explained through discrete particle settling rates (Figure 2). These fine particles must be removed by either adsorption onto larger

particles or aggregation as a result of particle collisions.

## REMOVAL RATES OF DIFFERENT CLOUD SPECIES

Note that in Figure 3 the ice and ash have similar removal rates, which are much faster than gas removal. Sulfur dioxide removal is largely a function of its chemical conversion rates to sulfate aerosol, rather than any kind of gravitational process. The resulting formation of sulfate aerosol is essentially the inverse of the gas decay. Other eruption clouds have shown similar patterns where fine ash removal is much faster than the sulfur dioxide gas (Table 1). The similarity of ice and ash removal rates, together with slow fallout calculated for discrete particles of this size, strongly suggest that both of these species undergo aggregation which drives their relatively rapid removal from the atmosphere.

## DOES ASH IN LARGE ERUPTIONS FALL OUT FASTER?

For Pinatubo, about 90% of the 1-15 micron sized ash fell out within the first 4 days of eruption (Figure 3). Approximately 99% of the fine ash was removed within 6 days. We have observed that several large eruptions have a significantly faster removal rate than smaller eruptions. How might this occur?

(1) The more intense eruption columns typically involve greater upward velocities. These stronger events are therefore more efficient at re-entraining particles in the rising ash and gas plumes than low-intensity eruption columns (Ernst et al., 1996).

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

(3) More fragmentation, and a higher ash volume, results in more electric charge generated in the volcanic plume, producing more electrostatic "sticking" of particles.

(4) Higher columns entrain more moist air and experience higher temperature gradients leading to

the formation of hydrometeors, resulting in further charge generation by processes similar to electric charge formation in thunderstorms.

(5) The combination of processes magnified by the more intense eruption columns produce more efficient particle removal by ash aggregation, ice coating and rapid removal as icy pyroclasts.

#### CONCLUSIONS

Satellite sensors have the ability to detect and quantify the 1-12 micron radius size fraction of drifting volcanic ash clouds. Studies of removal rates and processes for a range of volcanic eruptions reveal that ash and ice particles fall out at much faster rates than do the co-emitted sulfur dioxide gas. The rapid fallout of fine particulates is best explained by aggregation processes, and in some cases, the formation of ice on ash particles.

## ACKNOWLEDGEMENTS

Our satellite-based studies over the past several years have been supported by NASA's Solid Earth and Natural Hazards program, NASA Headquarters, and the TOMS Science Team; and the National Science Foundation's Petrology and Geochemistry program.

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