GEOPHYSICAL RESEARCH LETTERS, VOL. 16, NO. 11, PAGES 1293-1296, NOVEMBER 1989

# INFRARED RADIATIVE TRANSFER CALCULATIONS FOR VOLCANIC ASH CLOUDS

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Abstract. Radiative transfer calculations are performed for volcanic ash clouds assumed to consist of mixtures of ash particles, ice spheres, water droplets and ash particles coated with sulphuric acid. The results show that for nascent volcanic eruption clouds a *reverse* absorption effect is noticeable in the infrared window between 10 $\mu$m and 13 $\mu$m, for which absorption of infrared radiation increases with increasing wavelength. This effect may be of use in detecting hazardous volcanic clouds from space.

## Introduction

The study of volcanic ash clouds is important for at least two reasons. Firstly, because of their effects on the climate system and secondly because of the potential hazard they present to private and commercial aircraft. Various studies have investigated the effects of volcanic clouds on climate and demonstrated that the radiative properties of these clouds can have an impact on the earth's surface temperatures [e.g. Pollack *et al.*, 1976]. These studies address the problem of perturbations to the background stratospheric sulphate layer some time after an explosive volcanic eruption has injected large quantities of particles and gases into the atmosphere. The observed size distribution of these dispersed volcanic clouds consists largely of sulphuric acid coated particles of small mean radii ($\leq$ 0.1 $\mu$m). These layers are observed to be thin, have long residence times and have concentrations of 1-50 particles cm$^{-3}$, peaking in the stratosphere between roughly 20-35 km. On the other hand the nascent volcanic eruption cloud consists of large ash particles (0.1 $\mu$m to 100 $\mu$m) with short residence times and highly variable vertical concentration profiles. Such clouds present potentially disastrous situations for aircraft encountering them. Consequently their detection and discrimination from ice and water droplet clouds is of some practical importance.

In this paper monochromatic radiative transfer calculations are performed at two wavelengths appropriate for the Advanced Very High Resolution Radiometer (AVHRR) on board the NOAA polar orbiting satellites for idealised nascent and dispersed volcanic clouds and compared with results for normal ice and water droplet clouds.

## Radiative Transfer Model

We wish to calculate the upwelling infrared radiance that a space borne radiometer would measure from a plane parallel homogeneous cloud layer in an isothermal atmosphere overlying a non-reflecting lower boundary. The radiance $I_i$ measured by the radiometer in a narrow band centred at wavelength $\lambda_i$ ignoring scattering is approximately,

$$I_i \approx e^{-\tau_i} B(T_s) + (1 - e^{-\tau_i}) B(T_c), \qquad (1)$$

where $T_s$ is the temperature of the surface, $T_c$ is the temperature of the cloud top, $B$ is the Planck function and $\tau_i$ is the cloud optical depth along the line of sight. The cloud optical depth is related to the cloud thickness and microphysical structure by,

$$\tau_i = \beta_{ext}(\lambda_i) L \qquad (2)$$

where $\beta_{ext}$ is the volume extinction coefficient of the cloud of geometrical thickness $L$. For optically thick cloud the measured radiance $I_i \approx B(T_c)$. Conversely for a partially transparent cloud layer overlying a warm surface $B(T_s) \gg B(T_c)$ and $I_i \approx e^{-\tau_i} B(T_s)$. Suppose that the radiometer has a second channel centred at a different wavelength $\lambda_j$, $\lambda_j > \lambda_i$. Then depending on the extinctions at these two wavelengths, the measured radiance may be greater or smaller at $\lambda_j$. Defining the difference in Planck brightness temperatures at the two wavelengths to be,

$$\Delta T = T_i - T_j \qquad (3)$$

we note that provided $\beta_{ext}(\lambda_i) > \beta_{ext}(\lambda_j)$, $\Delta T$ is positive. This is generally the case for ice and water clouds. For volcanic clouds we will show that $\Delta T$ can be negative.

The radiative transfer equation for the model cloud including scattering may be written [Chandrasekhar, 1960],

$$\mu \frac{\partial I}{\partial \tau}(\tau, \mu) = I(\tau, \mu) - (1 - \varpi_o) B(T) - \frac{\varpi_o}{2} \int_{-1}^{1} P(\mu; \mu') I(\tau, \mu') d\mu' \qquad (4)$$

where $\tau$ is the optical depth, $\mu$ is the cosine of the zenith angle, $\varpi_o$ is the single scattering albedo and $P$ is the axially-symmetric phase function. The cloud layer has an optical depth $\tau_1$ and we are only concerned with the upwelling radiance in the direction $\mu$ at the cloud top ($\tau = 0$). The boundary conditions are no downward radiance incident on the cloud top,

$$I(0, -\mu) = 0, \qquad (5)$$

and the upward radiance incident on the cloud base is due only to that emitted from the ground,

$$I(\tau_1, +\mu) = B(T_s). \qquad (6)$$

The plus and minus signs signify upward and downward directions respectively. The radiative transfer equation is solved using the discrete ordinates method [Chandrasekhar, 1960; Liou, 1973; Stamnes and Swanson, 1981]. It can be shown that the solution to (4) may be written

$$I(\tau, \mu_i) = \sum_j L_j W_j(\mu_i) exp(-k_j \tau) + B(T) \qquad (7)$$

with $-n \leq i, j \leq n$, where $2n$ is the number of discrete radiation streams. The eigenvalues, $k_j$ and eigenvectors $W_j$ are determined by using an algebraic eigenvalue equation solver as suggested by Stamnes and Swanson (1981). The constants of integration, $L_j$ are determined from the boundary conditions.



Paper number 89GL03201.
0094-8276/89/89GL-03201$03.00

In all of the results to be presented 16 radiation streams have been used in the calculations. The single scattering albedo and phase functions are determined from a Mie scattering program [Evans, 1987] and input into the model. Given the complex refractive index ($m$) the program computes an extinction efficiency factor ($\hat{Q}_{ext}$), a scattering efficiency factor ($\hat{Q}_{sca}$) and the efficiency for absorption ($\hat{Q}_{abs} = \hat{Q}_{ext} - \hat{Q}_{sca}$) for the particle size distribution specified and optionally computes the phase function. For polydispersions !e factors are defined by,

$$\hat{Q}_f = \frac{\int_o^\infty \pi r^2 Q_f(\frac{2\pi r}{\lambda}, m)\frac{dn(r)}{dr}dr}{\int_o^\infty \pi r^2 \frac{dn(r)}{dr}dr} \quad (8)$$

where $Q_f$ is the Mie efficiency factor for extinction, scattering or absorption and $n(r)$ is the size distribution of particles with radius $r$ in units of number of particles per unit volume. In these calculations the log-normal size distribution and the modified gamma distribution are used to compare results for ice spheres, water droplets and the volcanic substances.

## The Nature of Volcanic Clouds

Perhaps the best observational data on eruption clouds has come from studies of the Mt. St. Helens ash clouds. Summaries of the results are contained in two NASA publications [Deepak, 1982 and Newell and Deepak, 1982] and a series of articles appearing in the Science Journal [Science, Vol. 211, 1981]. As might be expected the results indicate highly inhomogeneous structure consisting of sulphur gases, ash particles and large amounts of water vapour. Large ash particles with bimodal and trimodal distributions were observed for the ash particles. The large particles had mode maxima in the range 1$\mu$m to 50 $\mu$m (radii).

The size distributions reported by Farlow *et al.* (1981) for the small particles indicate log-normal type distributions with mean particle radii varying between 0.5 to 1.0 $\mu$m. Chuan *et al.* (1981) and Hobbs *et al.* (1981) made size spectra measurements for various parts of ash clouds and found particle concentrations of 100-200 $cm^{-3}$ with radii of 0.5 to 5.0 $\mu$m. Similarly Mossop (1964) found high concentrations in the range 0.5 to 1.0 $\mu$m for volcanic dust from the Mt. Agung eruptions.

After a few days the larger particles fall out and chemical reactions transform the sulphur gases into sulphuric acid droplets and acid coated ash particles. Since it is likely that the variability in constituents for different eruption clouds will be large, we attempt only to indicate the main effects on radiative transfer in the 10$\mu$m- 13$\mu$m window by performing calculations for six major constitients:- volcanic pumice, quartz, sulphuric acid, acid coated particles, water droplets and ice particles.

## Results

### Optical constants

The complex refractive indices for the six major components of eruption clouds and the sources of the information are given in Table 1 for 10.8 $\mu$m and 11.9 $\mu$m wavelengths. Some of the samples (e.g. volcanic pumice) were collected during ashfall and therefore may be more representative of the optical properties expected in an actual eruption cloud. Sulphuric acid particles are assumed here to consist of 75% $H_2SO_4$ aqueous solution in water and the ash particles contain a high percentage ($\geq$ 50%) of $SiO_2$.

### Mie Scattering Calculations

Table 2 summarises the results obtained for the log-normal size distribution using the optical constants shown in Table 1. These results show that the volcanic and quartz substances have similar characteristics and that for particle sizes less than a wavelength the absorption at 10.8 $\mu$m is greater than that at 11.9 $\mu$m. This is the opposite effect obtained for ice and water. The effect is more easily seen in Figure 1 where $\hat{Q}_{ext}$ is plotted as a function of particle radius for the modified-$\gamma$ distribution.

TABLE 1. Refractive indices ($n_r$ -real part; $k$ - imaginary part) for six constituents of volcanic eruption clouds. The last column gives a reference for the information with the following meaning:- HQ=Hale and Querry (1973), W=Warren (1984), PW=Palmer and Williams (1975), IP=Ivlev and Popova (1973) and V=Volz (1973).

| Substance | 10.8 $\mu$m | | 11.9 $\mu$m | | Source |
|---|---|---|---|---|---|
| | $n_r$ | $k$ | $n_r$ | $k$ | |
| Water | 1.153 | 0.097 | 1.111 | 0.199 | HQ |
| Ice | 1.091 | 0.168 | 1.258 | 0.409 | W |
| 75% $H_2SO_4$ | 1.678 | 0.301 | 1.844 | 0.194 | PW |
| Quartz | 1.940 | 0.350 | 1.780 | 0.190 | IP [1] |
| Pumice | 1.790 | 0.370 | 1.800 | 0.180 | V |

[1] Interpolated from their Figure 4.

TABLE 2. Mie scattering results for three volcanic cloud constituents and two sulphuric acid coated spherical ash particles using the log-normal size distribution. The coated particles consist of a percentage $H_2SO_4$ (by volume) covering a volcanic pumice particle.

| Substance | 10.8 $\mu$m | | | 11.9 $\mu$m | | |
|---|---|---|---|---|---|---|
| | $\varpi_o$ | $g$ | $Q_{ext}$ | $\varpi_o$ | $g$ | $Q_{ext}$ |
| $r_o$=0.2 $\mu$m, $\sigma$=0.5 $\mu$m | | | | | | |
| Pumice | 0.005 | 0.005 | 0.084 | 0.005 | 0.004 | 0.035 |
| Quartz | 0.007 | 0.005 | 0.070 | 0.005 | 0.004 | 0.038 |
| 75% $H_2SO_4$ | 0.004 | 0.005 | 0.075 | 0.005 | 0.004 | 0.037 |
| 90% coating | 0.004 | 0.005 | 0.077 | 0.006 | 0.004 | 0.038 |
| 10% coating | 0.005 | 0.005 | 0.085 | 0.005 | 0.004 | 0.036 |
| $r_o$=1.0 $\mu$m, $\sigma$=0.5 $\mu$m | | | | | | |
| Pumice | 0.266 | 0.131 | 0.779 | 0.329 | 0.097 | 0.360 |
| Quartz | 0.333 | 0.142 | 0.801 | 0.307 | 0.096 | 0.369 |
| 75% $H_2SO_4$ | 0.247 | 0.124 | 0.646 | 0.336 | 0.099 | 0.386 |
| 90% coating | 0.250 | 0.119 | 0.644 | 0.321 | 0.092 | 0.353 |
| 10%!ating | 0.266 | 0.125 | 0.747 | 0.316 | 0.090 | 0.335 |
| $r_o$=2.0 $\mu$m, $\sigma$=0.5 $\mu$m | | | | | | |
| Pumice | 0.448 | 0.422 | 2.066 | 0.572 | 0.352 | 1.517 |
| Quartz | 0.502 | 0.427 | 2.331 | 0.551 | 0.350 | 1.488 |
| 75% $H_2SO_4$ | 0.441 | 0.418 | 1.769 | 0.573 | 0.355 | 1.636 |
| 90% coating | 0.452 | 0.455 | 1.945 | 0.583 | 0.399 | 1.818 |
| 10% coating | 0.457 | 0.458 | 2.194 | 0.583 | 0.396 | 1.713 |
| $r_o$=3.0 $\mu$m, $\sigma$=0.5 $\mu$m | | | | | | |
| Pumice | 0.493 | 0.634 | 2.790 | 0.626 | 0.577 | 2.744 |
| Quartz | 0.521 | 0.623 | 3.042 | 0.610 | 0.579 | 2.675 |
| 75% $H_2SO_4$ | 0.502 | 0.639 | 2.577 | 0.618 | 0.576 | 2.873 |
| 90% coating | 0.501 | 0.659 | 2.662 | 0.625 | 0.597 | 2.993 |
| 10% coating | 0.497 | 0.654 | 2.830 | 0.630 | 0.599 | 2.895 |
| $r_o$=5.0 $\mu$m, $\sigma$=0.5 $\mu$m | | | | | | |
| Pumice | 0.486 | 0.775 | 2.860 | 0.588 | 0.712 | 3.197 |
| Quartz | 0.487 | 0.760 | 2.900 | 0.580 | 0.719 | 3.165 |
| 75% $H_2SO_4$ | 0.502 | 0.783 | 2.865 | 0.571 | 0.711 | 3.184 |
| 90% coating | 0.495 | 0.795 | 2.843 | 0.567 | 0.726 | 3.182 |
| 10% coating | 0.484 | 0.788 | 2.833 | 0.579 | 0.727 | 3.207 |

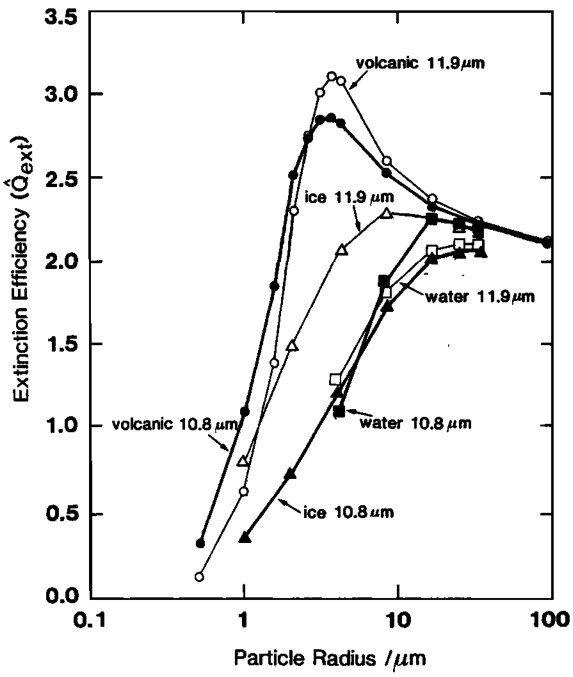


Fig. 1. Extinction efficiency ($\hat{Q}_{ext}$) for volcanic pumice, ice and water at two wavelengths as a function of particle size for the modifed-$\gamma$ size distribution.

## Radiative Transfer Calculations

Using the scattering parameters, calculations were performed at 10.8 $\mu$m and 11.9 $\mu$m for a cloud layer of varying optical thickness. The boundary conditions were applied for a cloud with cloud top temperature, $T_c$=220 K (representing a height of approximately 12 km) and surface temperature $T_s$=300 K. For each value of the optical thickness, corresponding brightness temperatures were obtained by inverting the Planck function at wavelengths 10.8 $\mu$m and 11.9 $\mu$m. As the cloud optical thickness increases from $\tau_1=0$ to $\tau_1=\infty$ the brightness temperature decreases from the lower boundary value $T_s$=300 K to the cloud top value of $T_c$=220 K.

The radiative transfer calculations show that the temperature difference ($\Delta T$) between the brightness temperatures at 10.8 $\mu$m and 11.9 $\mu$m at a particular optical depth is positive for ice and water clouds [Yamanouchi *et al.*, 1987], whereas for volcanic pumice, quartz and the acid droplets $\Delta T$ is negative provided the mean particle size is less than 3 $\mu$m or so. It would appear that $\Delta T$ is a sensitive function of the refractive index and the size distribution. The negative difference may be indicative therefore of volcanic type clouds.

## Volcanic Cloud Scenarios

### Nascent Eruption Cloud

Potentially the most hazardous situation for aircraft is an encounter with a recent eruption, for which the cloud contains large particulate matter mixed with eruption gases, ice and water droplets. Such a cloud in reality would be extremely inhomogeneous, with a composition changing rapidly with time. This complex structure is modelled here by assessing the effects of the major components separately and then assuming that each represents a non-interacting fraction of the total effect of the cloud. The components are assumed to be a fraction $f_i$ of ice particles and a fraction $f_a$ of ash particles. Since the water droplets behave in a similar manner to the ice particles and the quartz and acid coated particles behave like volcanic pumice it is expected that including these components as fractions would not change the results too much. The scenario is depicted inset in Figure 2 together with the results for the radiation stream closest to vertical and a mixture consisting of $f_i$=0.5 for modified-$\gamma$ size distributions with $r_0$=2 $\mu$m and 16 $\mu$m (solid line). The region of negative $\Delta T$ extends from the coldest to warmest parts of the cloud for 2 $\mu$m particles and only positive $\Delta T$ is found for 16 $\mu$m. Particle sizes increasing from 2 $\mu$m lead to progressively more positive $\Delta T$. Consequently for a distribution with a large mean particle size negative differences may not be observed.

### Stratospheric Eruption Cloud

In this scenario, illustrated in Figure 3, the cloud is assumed to have penetrated the tropopause and formed a dispersed layer of acid coated particles (10% $H_2SO_4$ by volume coating volcanic pumice) and ice particles. The acid particle size distribution is assumed to be log-normal with $r_o$=1.0 $\mu$m and standard deviation $\sigma$=0.5 $\mu$m. The ice particles follow a modified-$\gamma$ distribution with $r_o$=4 $\mu$m. Two fractions are shown:- $f_i$=0.7 (solid line) and $f_i$=0.5. In the case $f_i$=0.7 both positive and negative $\Delta T$ occur with the positive values over the warmer (optically thinner) parts of the cloud.

## Conclusions

Detection of volcanic eruption clouds particulary in the early stage of formation when their composition is most hazardous to aircraft, by measuring the upwelling radiances at 10.8 $\mu$m and 11.9 $\mu$m has been shown to be theoretically possible. The 'reverse' absorption effect observed for acid particles and some volcanic debris provides a signature that may be used to discriminate volcanic clouds from water/ice clouds. Because there are few measurements of the optical properties of volcanic clouds there is some uncertainty about the magnitude of the effects on the 10.8 $\mu$m and 11.9 $\mu$m brightness temperatures discussed here. Using AVHRR measurements, Prata (1989) observed negative temperature differences of 1-5 K for the eruption clouds of the Mt. Galunggung volcano in Indonesia. Global area coverage (GAC) data from the AVHRR was obtained from NOAA/NESDIS for a NOAA-7 orbit received at 1900 UT on 13 July 1982, just a few hours after an eruption of the volcano. Figure 4 shows contours of $\Delta T$ computed from the measured radiances

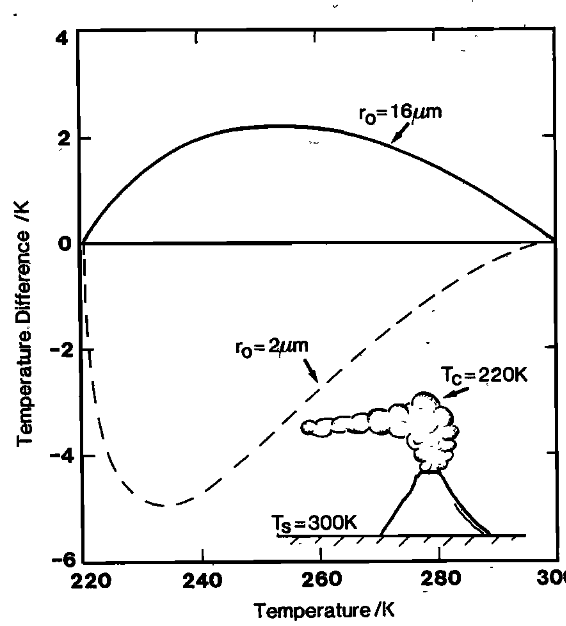


Fig. 2. Temperature difference ($T_{10.8} - T_{11.9}$) as a function of temperature at 10.8 $\mu$m for a recent volcanic cloud with cloud top temperature $T_c$=220 K and surface temperature $T_s$=300 K.

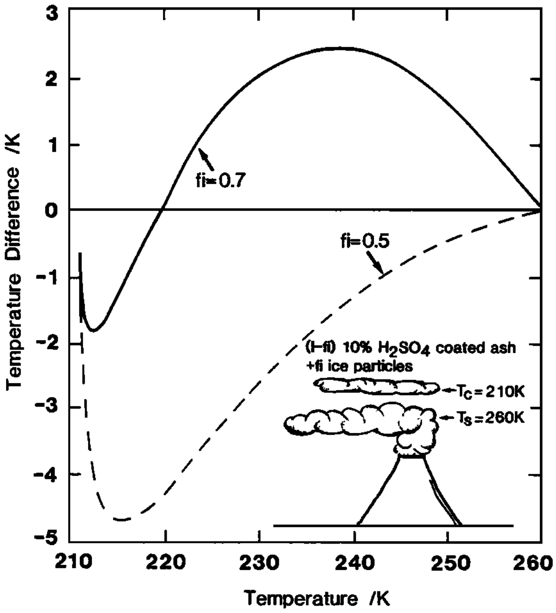


Fig. 3. As for Figure 2 but for a dispersed stratospheric cloud overlying optically thick volcanic cloud. The stratospheric cloud top temperature is $T_c$=210 K and the temperature of the thick cloud is $T_s$=260 K.

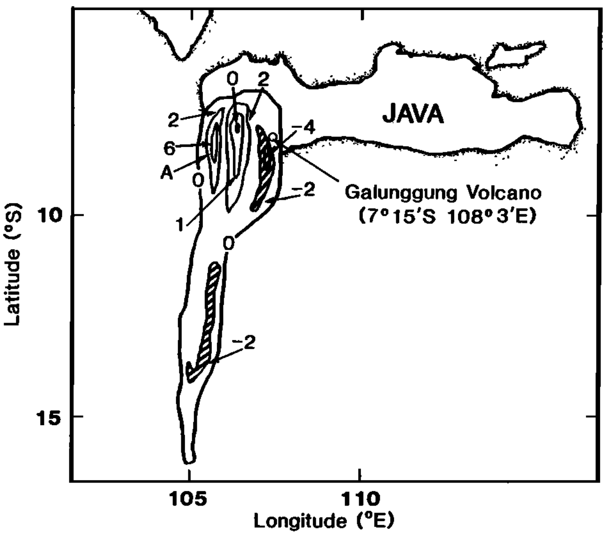


Fig. 4. Contours of the temperature difference ($\Delta T$) derived from observations of an eruption of the Mt. Galunggung volcano made by the NOAA-7 AVHRR.

for the eruption cloud. Negative $\Delta T$'s (shaded on the figure) occur in several locations near to the volcano and in a region some distance downwind from the main vent. Large postive differences also occur (region A on Figure 4); presumably where there is a high proportion of entrained water vapour and/or ice particle formation. Further observational evidence is required in order to verify these findings and a more complicated theoretical study utilising a realistic inhomogeneous volcanic cloud is warranted.

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(Received: April 3, 1989
Accepted: July 26, 1989)