Volcanic Particle Aggregation in Explosive Eruption Columns Part II: Numerical Experiments

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Abstract. Particle aggregation in eruption columns from explosive volcanoes is investigated by numerical experiments with the plume model ATHAM (Active Tracer High Resolution Atmospheric Model) employing the parameterizations described in part I of this paper.

The dominant role of hydrometeors in controlling many processes in the plume is revealed in this study. By far the highest portion of condensed water freezes to ice in the eruption column, because of the fast plume rise to regions, which are too cold for even supercooled liquid water to exist. The mass of tephra in the eruption column is about 100 times that of condensed hydrometeors during the eruption. However, aggregation of wet or icy volcanic ash by gravitational capture can cause efficient particle growth. Most of the particles occur as relatively dry icy-ash aggregates. Dry atmospheric conditions in the lower troposphere can lead to total evaporation and to the deposition of completely dry aggregates. As a consequence, no evidence of hydrometeor-ash interaction could be found in the fallout of the eruption, although it occurred to a significant degree in upper parts of the plume.

The collection efficiency for water-coated is much higher than for icy particles. However, in a series of experiments with modified ambient conditions, we do not find an effect of the amount of liquid water, because of the overwhelming dominance of ice. We did realize a high sensitivity to the amount of ice in the model domain, which is dependent on the ambient humidity. The aggregation efficiency is governed by the availability of hydrometeors acting as adhesives at the particles’ surface. Thus, in turn, is dependent on the ash morphology. The temperature dependence of the collection efficiency of frozen particles is an important parameter for aggregate growth. Particle microphysics is quite indifferent to a decrease of the threshold temperature, below which ash particles become active as ice nucleation nuclei, because of the quick temperature decrease well below this threshold. The initial size distribution of erupted particles has a major influence on ash aggregation and sedimentation patterns. Very little data exist particle properties within the eruption column, and our sensitivity studies proved an urgent need of further constraining ash porosity and surface properties at low temperatures by observations.

First experiments based on invariant electric fields and constant electric properties show a significant effect of electrostatic forces on the aggregation of small dry particles. These findings, however, await more detailed investigations.

In this study we employ simplified parameterizations, which we believe to cover the range of possible values. Our results provide insights in the sensitivity of the ash aggregation process to a number of parameters. These should now be further investigated in the laboratory and possibly in the field to reduce the uncertainty in our knowledge about particle microphysics within the eruption column.
1. Introduction

The goal of this paper is to examine the relevant parameters that determine the aggregation efficiency and the growth rate of volcanic particles within the eruption column. We do not try to simulate a specific eruption, but choose conditions typical for highly explosive events. We perform numerical experiments with the non-hydrostatic, non-steady plume model ATHAM (Active Tracer High Resolution Atmospheric Model), which is designed to simulate the processes within an explosive volcanic column in the atmosphere. We employ the parameterization of cloud and ash particle microphysics described in part I of this paper.

After a short introduction of the ATHAM model and its setup for this study in sections (2) and (3), we describe the results from the numerical simulations. In section (4.1) we explain the reference experiment, and present the general characteristics of tephra, hydrometeors, and the particle properties in the eruption cloud. We show the importance of considering when we compare the results of a simulation without particle aggregation to the reference simulation in section (4.2). The effect of particle morphology on the collection efficiency of wet or icy ash aggregation is investigated in section (4.3). The impact of electrostatic forces and that of the initial particle size distribution are examined in section (4.4) and in section (4.5), respectively. Finally, in section (4.6), we present simulations for different atmospheric background conditions.

2. Plume Model ATHAM

The non-steady state, non-hydrostatic numerical model ATHAM (Active Tracer High Resolution Atmospheric Model) [Oberhuber et al., 1998; Herzog, 1998; Graf et al., 1999] was especially designed to simulate the dispersal of a volcanic eruption column resulting from an explosive volcanic event. The simulation time is some hours covering spatial scales of some hundreds of kilometers. ATHAM describes the spatial-temporal evolution of the gas-particle-mixture. For this study, we developed and implemented a module for the description of the microphysics of hydrometeors and ash particles and the interaction between them, which leads to particle growth, see part I of this paper. For a complete description of the model concept and equations see Oberhuber et al. (1998), Herzog (1998) Herzog et al. (2003).

The plume model ATHAM explicitly predicts the temporal and spatial development, the only prescribed quantities are the parameters of the volcanic eruption and the conditions in the ambient atmosphere. The model formulations do not vary with the distance from the vent: the same set of equations for the dynamics, the thermodynamics and the microphysics is calculated at each grid point at every time step. The ATHAM model has been successfully used to simulate the evolution of a volcanic cloud in the atmosphere on a time scale of about an hour (Graf et al., 1999; Textor et al., 2003b,a; Herzog et al., 2003). In addition, ATHAM has been used for the simulation of biomass burning events (Trentmann et al., 2002, 2003). The numerical experiments
presented here refer to processes in the eruption column during the eruption and shortly afterwards. We focus on the exploration of the fate of the fine grain-size fraction in the atmosphere. There are several reasons for this. First, fine ash smaller than 1 mm accounts of over 90% of the particle mass in an eruption column (Sparks et al., 1997). Second, fine ash only is relevant for the mesoscale effects of the eruption, and on the amount of material injected into the stratosphere. We are not concerned here by in the large blocks and bombs which fall ballistically to the ground, and by the coarse material, because these do not influence the larger scale plume dispersion.

In this study the following processes are resolved within the concept of ATHAM:

- The dynamic part solves the Navier-Stokes equations for the gas-particle-mixture and treats the transport of active tracers including particle sedimentation. (Oberhuber et al., 1998; Herzog, 1998).

- The turbulence closure scheme delivers the turbulent exchange coefficients for each dynamic quantity, thereby describing the entrainment of ambient air into the plume (Herzog, 1998; Oberhuber et al., 1998; Herzog et al., 2003).

- The cloud microphysics describes condensation of water vapor and formation of precipitation. All phases of water are included: vapor, liquid and solid. The feedback of the thermal energy changes on the dynamics is considered (Graf et al. (1999); Herzog et al. (1998), and part 1 of this paper).

- The ash module describes particle growth and coagulation based on microphysical interactions between hydrometeors and ash part 1 of this paper).

3. Model Setup

In this study we employ the two dimensional version of the model which is formulated in cylindrical coordinates, for the sake of saving computer memory and time. The effects of a mean lateral wind cannot be considered, the resulting flow pattern in the atmosphere initially at rest is only influenced by the dynamics induced by the eruption column. However, the dilution of the mixture by entrainment of surrounding air corresponds to a three dimensional simulation with an atmosphere initially at rest. The results from our experiments are suitable to investigate the principal features of a volcanic eruption column. We did not try to simulate a specific volcano, but choose conditions typical for highly explosive eruptions.

The experiments are performed on a stretched lattice with $150 \times 80$ grid points. The model domain is 300 km in the horizontal and 50 km in the vertical direction. In the center of the model domain, where the volcano is situated, we use a spatial resolution of 250 m in the horizontal and 300 m in the vertical, at the lateral boundaries it is about 5 km. This grid choice allows for the plume dispersal without disturbances from the model boundaries during our simulation time of 90 min, with restricted computer resources.
The high-pressure regime in the conduit and at the vent, the shock-like expansion of the erupting mixture, and the small scale processes in the vicinity of the crater cannot be resolved in the concept of ATHAM. We focus on processes occurring in the eruption column, in the range of some tens of meters to some tens of kilometers. The numerical experiments begin just after the earliest mixing of the erupting gas-particle-mixture with the atmosphere, when the equilibration with the atmospheric pressure has already taken place. The input of volcanic material during the eruption is specified by the vertical velocity, temperature and composition of the ejecta at three grid points which represent the base of the eruption column.

We choose gas a mass fraction \( q_{a,\text{base}} \) of 7.34 wt.\%, where water vapor contributes about 82 \%. The particle density is set to \( \rho_{\text{ash}} = 1800 \text{kg/m}^3 \). The temperature of the expanding mixture does not significantly change because of the high concentration of particles, which have a large heat capacity. Thus, we can use temperature data obtained from thermochemical calculations to initialize the model. Decompression of the erupting mixture typically takes place within a vertical distance of some jet radii above the vent. Hence, we can neglect the topography of the volcanic crater and use a flat surface. At the end of the decompression phase, the vertical velocity at the base of the eruption column \( w_{\text{base}} \) can be at most twice the speed of sound, if the apex angle of the crater is wide enough (Woods and Bower, 1995).

We use the specific heat capacities for air and water vapor, and get for the speed of sound of the mixture \( c_s \approx 210 \text{ m/s} \). For the initial vertical velocity we set \( w_{\text{base}} = 400 \text{ m/s} \). Within the first 60 seconds, the eruption velocity is increased to its maximum value. It is kept constant for the following 27 min. During the last 3 min of the eruption, the vent exit velocity is reduced to zero again. We continue the experiment for additional 60 min in order to investigate the post-eruptive development of the eruption cloud.

The radius of the base of the eruption column \( \text{rad}_{\text{base}} \) depends on the mass eruption rate \( M \) in [kg/s], which is equal to:

\[
M = \pi \text{rad}_{\text{base}}^2 \rho_{\text{mix}} w_{\text{base}}
\]

where \( \rho_{\text{mix}} \) the density of the gas-particle mixture. It can be obtained from the volume ratio of particles and gas:

\[
\rho_{\text{mix}} = \frac{q_a + q_{\text{ash}}}{q_a/\rho_a + q_{\text{ash}}/\rho_{\text{ash}}}
\]

with \( q \) indicating the mass mixing ratios and the indices \( \text{ash} \) and \( a \) referring to ash and gas (air), respectively. The gas density is taken from the ideal gas law. With a plume base radius of \( \text{rad}_{\text{base}} = 375 \text{ m} \), and with \( w_{\text{base}} = 400 \text{ m/s} \) we get a mass eruption rate of \( M \approx 4.5 \cdot 10^8 \text{ kg/s} \), which is typical for a highly explosive event (e.g., Sparks et al., 1997) REF OK??.
The initial conditions at the base of the eruption column summarized below are the same for all experiments presented here.

- height: 600 m above sea level, plume base diameter: 750 m;
- vertical velocity: 400 m/s, temperature: 1100 K;
- gas mass fraction: 7.34 wt.%, where water vapor contributed 81.74 %;
- particle mass fraction: 92.66 wt.%;
- particle density: $\rho = 1800 \text{ kg/m}^3$;
- density of the gas-particle-mixture: $\sim 2.6 \text{ kg/m}^3$;
- mass eruption rate $\sim 4.5 \cdot 10^8 \text{ kg/s}$.

The particle size distribution of the reference experiment has been fitted to Mt. St. Helens (1980) data (see part I of this paper), with 40% small (2.5 $\mu$m in radius) and 60% large particles (50 $\mu$m in radius). The ambient conditions for pressure, temperature and humidity correspond to standard atmospheric profiles for the subtropics taken from (McClatchey et al., 1972), with the tropopause at about 17 km as shown in figure (1).

**Figure 1:** Atmospheric profiles for temperature and relative humidity in the reference experiment McClatchey et al. (1972). The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines.
4. Results

4.1. General Characteristics of the Eruption Column

In the reference experiment, the plume penetrates the tropopause at 17 km about 5 min after the eruption has started. The average vertical velocity in the core of the column is greater than 100 m/s. The eruption column consisting of the hydrometeor-ash aggregates is shown 30 min after the eruption onset in figure 2.

![Figure 2: Eruption cloud of hydrometeor-ash aggregates, shown for the reference experiment after 30 min. The scale gives mass mixing ratio in [g/kg \text{tot. mass}].](image)

The height of neutral buoyancy (HNB) occurs at about 23 km, 6 km above the tropopause. The ash fraction of hydrometeor-ash aggregates is typically greater than 80% by mass. This reflects the fact that the hydrometeor mass accounts only for some percents of the total particle mass after 30 min. Hence, figure (2) corresponds roughly to the plume of pure volcanic ash. Figure (3) depicts the vertical distribution, \textit{i.e.}, the horizontal integral of ash at each height level, at different times during the course of the simulation. Larger aggregated particles settle into the troposphere, forming a deep ash layer. This is indicated in figure (3) by the lower broad peak in the

\footnote{All fractions given in this paper are mass fractions if not otherwise noted.}
distributions, which moves downward with time from the HNB, which was reached after 5 min, to about 5 km after 90 min (i.e., one hour after the end of the eruption). The narrower, stationary peak at about 23 km stems from small particles (about 2.5 μm in radius), which are suspended within the umbrella region. This portion of the cloud spreads horizontally in the stratosphere. After 90 min, the plume has a horizontal extend of more than 250 km, and only about 10% of the erupted ash can be found in the stratosphere.

**Figure 3.** Vertical distributions (horizontal integral in mass per height level, [kg/m]) of ash particles at t = 5, 10, 30, 60 and 90 min, shown for the reference experiment. The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines.

The eruption clouds of liquid water plus ice after 30 and 90 min, as well as the related vertical distributions of each hydrometeor class are plotted in Figures (4) and (5), respectively.

**Figure 4.** Eruption cloud of all hydrometeors (mass mixing ratio in [g/kg]) (left panel). Related vertical distribu-
tions of each hydrometeor class (horizontal integral in mass per height level, [kg/m]) The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines. (right panel). Shown for the reference experiment after 30 min.

**Figure 5.** Eruption cloud of all hydrometeors (mass mixing ratio in [g/kg]) (left panel). Related vertical distributions of each hydrometeor class (horizontal integral in mass per height level, [kg/m]) The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines. (right panel). Use key from figure (4). The horizontal extent of the hydrometeor cloud in the stratosphere is about 250 km. Shown for the reference experiment after 90 min (60 min after the end of the eruption)

Liquid water persists only in the core of the plume, with the greater fraction of the hydrometeors in the eruption cloud taking the form of ice particles (>99% by mass after 30 min). This predominance of ice in eruption clouds is caused by the fast plume rise to temperatures too low for even supercooled water to exist. The temperature field at 30 min of eruption is shown in figure (6). Numerical simulations with the ATHAM model including a parameterization for freezing temperature depression caused by dissolved gases confirmed this result (Textor et al., 2003a). The prevalence of ice in volcanic plumes is supported by observations, as discussed in this paper.

**Figure 6.** Temperature in the eruption cloud, shown for the reference experiment after 30 min. The scale gives temperature in [°C].

At the beginning of the eruption, more than 95% of the ice is in the small particle class. Water vapor deposition in the rising eruption column occurs primarily on small particles, which provides most of the ash surface. Small particles grow by aggregation thus transferring the ice with them to the larger particle class, which contains about half of the ice in the model domain from 10 min of eruption on. The small particles spread
horizontally at the HNB, where water vapor deposition produces relatively ice-rich particles consisting of up to 25 % ice. At the same time, the larger ice-ash aggregates continue to settle down. The low relative humidity in the lower troposphere in our scenario (see figure 1), causes sublimation of ice above melting level. Below an altitude of approximately 8 km, liquid water or ice is almost entirely evaporated, see figure (5), and almost no rain reaches the ground. Thus, in a deposit of an eruption under conditions like in our experiment, no hydrometeors would be detected in the deposits, although they play a significant role during the evolution in the eruption column.

Figures (7a), (7b), and (7c) display the particle radii in the dominant large particle class (i.e., rain or graupel) at 30, 60 and 90 min, respectively. In our model formulation, it is not possible for hydrometeors and ash particles to coexist in the same place. Hence, the displayed radius refers to particles consisting of either pure hydrometeor, pure volcanic ash or mixed hydrometeor-ash aggregates.
Figure 7: Radii of particles in the large mode of frozen particles at locations with mass mixing ratios > 5g/kg. Shown at a: 30, b: 60, and c: 90 min for the reference experiment.

Particle growth continues during lateral transport in the umbrella cloud, thus, the biggest particles with radii larger than 120 $\mu$m can be found at the leading edges of the plume, see figures (7).

The parameterizations for the interaction of volcanic particles and hydrometeors in this study is based on the assumption that the ash particles are always active as cloud condensation nuclei for liquid as well as for ice clouds. Although this assumption is probably true (see discussion in part I of this paper) we tested the influence of decreased ice nucleation capacity in a sensitivity experiments where we allowed the sublimation of water only for temperatures lower than $-15^\circ$C. The high temperature gradient within the rising volcanic eruption column leads a quick reduction of temperatures well below $-15^\circ$C. Therefore, the microphysics in a volcanic plume is not very sensitive to changes in ice nucleation capacity with temperature and our results are unaffected. In our model, we included additional processes for the nucleation of frozen and liquid hydrometeors on non-volcanic particles in the backgeround atmosphere. A sensitivity study neceleging these processes did not change our findings.

In contrast to the parameterization of usual meteorological clouds, we considered in this study the intra-modal aggregation of large cold particles. The life times of these particles are relatively long in a volcanic cloud, because they are smaller than precipitating particles in meteorological clouds, and because of the great height of the volcanic umbrella cloud. We tested the effect of neglecting this process in a sensitivity study. Intra-modal particle aggregation leads to enhanced particle growth, and the precipitating tropospheric ash layer can thus be found about 3 km higher than in the reference experiment, but the amount of ash in the stratosphere is not changed. Although, the effect of intra-modal aggregation of large cold particles on the plume development of an explosive volcanic eruption is not significant, this experiment indicates the strong need of more detailed information on the ash aggregation process.
In the reference experiment, the aggregates’ terminal fall velocities increase with height from 3 to 7 m/s due to the decrease of friction at greater altitudes. With an average fall velocity of 4 m/s these aggregates need about 90 min to fall from the top of the umbrella cloud to the ground. In this study, the particles have been assumed to be solid spheres. The effect of particle shape on the fall velocity is only considered by applying a drag coefficient as explained in Herzog et al. (1998). These simplifications might lead to an overestimation of the fall speed, however, they seem to be justified compared to the general lack of information about the particle’s shape and size distribution in the plume. We will however improve the representation of ash particle shape in a consecutive study.

Observations of the ash deposits showed millimeter sized volcanic aggregates as reported in (e.g., Sparks et al., 1997). In our simulation, aggregates do not reach this size. We will demonstrate, why ash growth is inefficient under the conditions of our reference experiment. In the following we will show the general importance of the ash aggregation process, and investigate the factors that determine its efficiency.

4.2. Neglecting Ash Particle Aggregation

The relevance of ash particle aggregation is investigated in this section. We compare the reference study (REF) with an experiment tagged DRY, in which we completely neglect the interaction of ash particles and hydrometeors. Here, water vapor deposition on ash and aggregation of coated particles is not considered, and ash is transported in the model, but does not take part in any microphysical process. The parameterizations presented in part I of this paper are applied only for pure hydrometeors. This experiment corresponds to an eruption taking place in an arid climatic zone with a smaller amount of condensed water available in the plume. Figure (8) depicts the vertical ash distributions at different times during the simulation. See also figure (3) for comparison with the reference experiment (note that the x-range is twice as large in DRY).
**Figure 8.** Vertical distributions of ash particles at $t = 5$, 10, 30, 60 and 90 min, shown for the experiment \textit{DRY}. The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines. The scale gives [kg/m].

In this experiment, sedimentation is much less distinct. At 90 min simulation time, the lower edge of the cloud consisting of 50 $\mu$m particles has reached about 10 km, whereas the aggregates in the experiment \textit{REF} have already reached the ground. In the experiment \textit{DRY}, a strong peak of small particle occurs in the stratosphere at about 23 km. Figure (9) shows the mass fraction of erupted ash above a certain height at 90 min simulation time. In the DRY experiment, about 40% remains above the tropopause, in contrast to about 13% in the reference study.

**Figure 9.** Mass fraction of erupted ash above a certain height at 90 min simulation time. The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines. Shown for the sensitivity experiment without particle aggregation.

Our experiments show that the consideration of aggregation significantly influences the residence time volcanic ash in the atmosphere, its injection into the stratosphere, and its deposition behavior. This is a conservative statement, because the aggregation efficiency of volcanic ash is more likely to be underestimated in our reference experiment.
4.3. Modification of the Collection Efficiency

The sensitivity of ash particle aggregation to variations of the collection coefficient $E$ is subject of this section. We modify $E$ but leave the ambient and volcanic conditions as in the reference experiment.

The collection efficiencies for mixed hydrometeor-ash particles in a volcanic cloud are unknown to date, but it is very probable that it depends on the availability of hydrometeor at the particle’s surface. Volcanic ash is often non-spherical in shape and highly porous depending on the magma type and on the style of eruption. Because of the limited knowledge about ash morphology, we do not attempt to calculate the amount of water or ice necessary to completely cover a particle. To our knowledge, there are no quantitative theoretical or experimental studies on the impact of these parameters on ash particle aggregation. Therefore, we performed sensitivity studies, which allow us to explore the effect of ash-hydrometeor-ratio on ash aggregation, and, thus, provide insights in the sensitivity of the aggregation process on the ash porosity. We employ simple parameterizations, which should cover the range of possible values, but which certainly await experimental validation.

As explained in part I of this paper, we approximate the collection efficiencies with the parameterizations commonly used in cloud microphysics $E_{sl,hydro}$ (see e.g., Pruppacher and Klett [1997]), and modify these parameterizations with simple functions, $f(x)$, reflecting the availability of hydrometeors at the particle surfaces:

$$ E_{sl,agg} = E_{sl,hydro} \cdot f(x) $$

(4)

$(x)$ is the hydrometeor mass fraction in the hydrometeor-ash particles.

$$ x = \frac{q_x}{q_{ax}} $$

(5)

For two different colliding particle classes, $x$ is determined by the particles class with the higher hydrometeor fraction. To ensure that the collection efficiency for mixed particles does not exceed that of pure hydrometeors, we limit $x <= 1$.

We assume a stable layer of water or ice around an ash particle for hydrometeor mass fractions of 50% or larger, so that the application of the collection efficiencies for pure hydrometeors can be employed. Hence, for a solid, spherical ash particle of 10 μm in radius, the hydrometeor layer around the ash core would have a diameter of $\Delta r = 2.6 \mu m$. We vary the collection efficiency for $x < 1$ as given below in equations (6) and illustrated in
Functions \( f \) given in equation (6), which modify the collection efficiencies, versus the hydrometeor fraction \( x \) given in equation (5). In the reference study, \( REF \), the efficiency is linearly dependent on the hydrometeor fraction of the colliding particles, as given in \( f_{REF} \). In next two experiments, named \( LIN1 \) and \( LIN2 \), we consider the possibility that less water or ice might be sufficient to completely cover the particle. We presume that hydrometeor fractions of 33 \% and about 10 \%, i.e., a layer of \( \Delta r_{LIN1} = 1.45 \mu m \) and \( \Delta r_{LIN2} = 0.32 \mu m \) respectively, are enough to completely cover a 10 \mu m ash core. This is given by functions \( f_{LIN1} \) and \( f_{LIN2} \). The effect of aggregate porosity is examined in the experiments \( POR1 \) and \( POR2 \), where we employ the respective functions.

Figure (11) shows the fraction of particles in the larger size classes during the course of the simulation. The
large particles contribute 60% of the ash initially erupted at the vent, aggregation transfers additional particles from the small size classes to the larger ones. Our simulations reveal a high dependency of the fraction of aggregated particles on the collection efficiency. In the experiment POR2, particle growth is nearly suppressed, because the hydrometeor fraction in the aggregates is in general lower than 20%, see section (4.1), whereas collection becomes efficient for hydrometeor fractions larger than 40%, see figure (11). It is also delayed in the experiment POR1, where the fraction of particles in the larger class is less than half of that in the reference experiment. In the experiments LIN1 and LIN2, the fractions of large ash are increased, especially in the experiment LIN2, where particle growth is so efficient that the millimeter sized particles are formed. Aggregates reach the ground about 10 min after the end of the eruption. (This removal of large particles from the atmosphere causes the apparent increase of the fraction of small particles in figure (11)).

Figure 11.: Mass fraction of particles in the larger size classes during the course of the simulation. The vertical line at 30 min indicates the end of the eruption. Shown for the sensitivity studies regarding the sticking coefficient conditions.

The overall aggregation rate is highest at the beginning of the eruption, when all particles occur in the rising core of the eruption column, where the particle density is extremely high (in the order of $10^5$/cm$^3$ and $10^4$/cm$^3$ small and large particles, respectively). When the particles reach the HNB after about 5 min, the aggregation rate decreases, because the particle density decreases by about one order of magnitude. In addition, the collection
efficiency decreases with decreasing temperatures, as discussed below. A small increase is noticeable after the eruption end. This can be attributed to an interim formation of liquid water in the central region of high particle density during plume collapse. After 40 min of simulation, aggregation is not very efficient any more, because of increasing dilution of the plume and decreasing still availability of liquid water. This is due to the low temperatures in most regions of the eruption column, see figure (6). Aggregates, which precipitate to warmer altitudes, meet small relative humidities in the lower troposphere in our scenario. Thus, ice evaporates before it melts to liquid water can occur, and dry particles do not aggregate in this series of sensitivity experiments.

All eruption columns reach the buoyancy height as in the reference experiment, however, the subsequent fate of ash in the atmosphere is quite different. Figure (12) shows the mass fraction of erupted ash above a certain height at 90 min simulation time. The lower edges of the settling ash layers can be found at heights which decrease with increasing collection efficiencies in the experiments $POR_1 \approx POR_2 > REF > LIN_1 > LIN_2$. At the end of the latter simulation, only a layer of less than 10% of the erupted ash remains in the stratosphere, the large aggregates have been entirely deposited to the ground. In all sensitivity experiments, most of the small particles are suspended in the stratosphere. The amount of ash in the stratosphere increases with decreasing aggregation efficiency. In the experiment $POR_1$, ash aggregation was much stronger reduced than in $POR_2$, and consequently the stratospheric cloud in the former contained more than 40% of the erupted ash, whereas the latter contained only about 30%, see figure (12). However, the lower edges of these two experiments occur both at a height of about 5 km at the end of our simulation. Although a larger fraction of particles is contained om aggregates in $POR_2$, and thus removed from the stratosphere, the size of the particles, i.e., their fall velocities, is not much larger than in $POR_1$. 
Figure 12: Mass fraction of erupted ash above a certain height at 90 min simulation time. The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines. Shown for the sensitivity experiments about the collection efficiency.

Particle removal reduces the density of the remaining ash layer in the stratosphere. Thus the HNB rises during the post-eruptive evolution, as shown in figure (13). This post-eruptive rise of the umbrella cloud becomes increasingly evident with increasing sedimentation of large particles, i.e., separation of the two particle classes. In the same way, sedimentation results in a separation of ash and volcanic gases, thus increasing the injection of the latter into the stratosphere.
Figure 13.: Mass weighted mean height of the eruption clouds of the small ash particles during the course of the simulation for the experiments REF, LIN1, LIN2, POR1, POR2, and EICE.

The effect of the aggregation efficiency of frozen particles was tested in the next sensitivity study called EICE. We apply the aggregation coefficient for ice cloud based on Lin et al. (1983). This formulation is applicable for meteorological clouds, but has not been verified for icy ash particles at great altitudes. In the experiment EICE, we employ the collection efficiency used in Murakami (1990), which neglects the temperature dependence. The collection efficiencies were modified in all processes, in which two frozen classes interact.

\[ E_{sl,\text{hydro},\text{REF}} = e^{0.05 \times T} \]  \hspace{1cm} (7)

\[ E_{sl,\text{hydro},\text{EICE}} = 0.1 \]

The aggregation coefficient of frozen particles in the experiment EICE is larger than that in the reference experiment at temperatures \( T \) below \(-46^\circ C\). The efficiencies of pure ice particles given in equation 7 are modified by the parameterizations representing the availability of hydrometeors at the particles’ surfaces, given in the function \( f_{REF}(x) \) in the equations 6 for the reference experiment. Figure (12) shows that the scavenging of small particles is less efficient in the experiment EICE than in the reference experiment. The contribution of
large particles to the total ash mass decreases from about 92 to 87%, when the temperature dependence of the collection efficiency is neglected. As a consequence of diminished particle growth, sedimentation is reduced and the particle residence time in the atmosphere is increased, see figures (12) and (13).

For the lack of observations, the dependence of the collection efficient $E$ on the availability of hydrometeors at the particle’s surface is represented by coarse estimations in this study. In all likelihood, our these do not exactly reflect the real dependencies. We could, however, show the relevance of having detailed information on the particle morphology, which determines the collection efficiency of the wet and icy aggregation process.

Particle growth has a secondary effect on the stream pattern. Falling aggregates induce an eddy structure in the flow around the eruption column, which in turn influences the residence time of particles in the atmosphere and the amount of ambient water vapor entrained into the plume. Hence, particle aggregation govern the plume dynamics and the injection of volcanic emissions into the stratosphere. Our results await urgently needed experimental verification in the laboratory and possibly in the field.

### 4.4. Electrostatic Forces

Electrostatic forces have been shown to be important for aggregation of especially small dry ash particles (Sorem, 1982; Gilbert and Lane, 1994; Schumacher, 1994; James et al., 2002), see part I of this paper. In ATHAM, however, we do not calculate electric fields. Therefore, it is impossible to explicitly simulate the effects of electrostatic forces on ash aggregation. Instead, we perform a series of experiments, in which we allow for the aggregation of dry ash particles in the small categories in order to get an appraisal of the potential importance of electrostatic aggregation. We modify the collection efficiencies for small particles in the equations for autoconversion, given in part I of this paper. In the studies called $ELS_{0.50}, ELS_{0.25}, ELS_{0.10}, ELS_{0.05},$ and $ELS_{0.01}$, we set the following collection efficiencies: $f_{ELS_{1}} = 0.50, f_{ELS_{2}} = 0.25, f_{ELS_{3}} = 0.10,$ $f_{ELS_{4}} = 0.05,$ and $f_{ELS_{5}} = 0.01$, respectively. As soon as the particles are coated with hydrometeors, our usual parameterizations for the wet or icy aggregation processes become active.

Figure (14) shows the fraction of particles in the larger size classes for the experiments $ELS$. The difference of the fractions of large particles is almost entirely determined by the aggregation rate in the central rising zone, as pointed out in the description of figure (11). The amount of aggregates increases with increasing collection efficiency from experiment $ELS_{5}$ to $ELS_{1}$; in the experiment $ELS_{1}$ almost all small particles are scavenged by larger aggregates. The delay of particle aggregation onset at the beginning of the eruption is the shorter than in the reference experiment, because it does not depend on the onset of hydrometeor condensation.

At times later than 40 min of simulation, particles do not aggregate to a large degree in this series of experiments, similar to the finding as in the previous studies (see section 4.3), in which aggregation depended
entirely on the amount of hydrometeor at the particles’ surfaces. However, the amount of dry particles in the small particle classes is rather limited after the eruption’s end. Thus, the modified processes of ‘electrostatic autoconversion’ are not important any more at this state of the simulation.

**Figure 14.** Mass fraction of particles in the larger size classes during the course of the simulation. The vertical line at 30 min indicates the end of the eruption. Shown for the sensitivity studies ELS.

In figure (15) the mass fraction of erupted ash above a certain height is plotted at 90 min simulation time. Although aggregation efficiencies are quite different for the single experiments, and higher than in REF, the difference in the lower edges of the plume heights is not. The sizes of the aggregates, and thus their fall velocities are only slightly larger than in the reference experiment, and they increase only slightly from experiment ELS5 to ELS1. This can be explained by the modified aggregation mechanism in these experiments. The modified ‘electrostatic autoconversion’ intensifies the transfers of small particles to aggregates, but all particles have the same growth probability. Non-linear particle aggregation, which would promote the growth of single, larger particles at the cost of others, is not affected by electrostatic aggregation in our study. Thus, the size and the fall speed of the aggregates does not change significantly. This finding is similar to the well known ‘second indirect effect’ in climate research (Albrecht, 1989). High aerosol concentrations in polluted air lead to high number concentrations of cloud drops. This causes a delay of the development of large, falling drops. Thus, precipitation development is delayed.
The amount of particles that are suspended in the stratospheric cloud decreases with increasing collection efficiency, because the amount of particles in the small classes decreases. In the experiment $ELS1$ only about 4% of the particles remain in the stratosphere, see figure 15, in contrast to about 13% in the reference experiment.

![Figure 15: Mass fraction of erupted ash above a certain height at 90 min simulation time. The ground level at 900 m and the tropopause at 17 km are indicated by horizontal lines. Shown for the sensitivity experiments $ELS$.](image)

The consideration of electrostatic forces for the aggregation of dry, small particles leads to a stronger increase of the fraction aggregated particles than in the previous experiments, where we increased the collection efficiency of aggregation in the presence of hydrometeors in the experiments $LIN1$ and $LIN2$. However, sedimentation was not much enhanced in the studies $ELS$, in contrast to the previous study. This result of enhanced aggregation efficiency without enhanced particle growth is true for invariant electric fields and constant electric properties of the aggregating particles. We believe, however, that in reality none of these will be constant, and thus, preferential particle growth might occur.

### 4.5. Size Distribution of Erupted Particles

In this section, we examine the importance of the initial size distribution of erupted particles on the aggregation efficiency. We perform a sensitivity study called $ASK$ under the same conditions as in the reference
experiment except for the initial size distribution, which reflects that of Askja D, as estimated by Woods and Bursik (1991) (WB) as an example for a plinian eruption.

Figure (16) depicts the particle size distribution for the Askja D ash, and the gamma functions which we fitted to this distribution. The initial mean volume radius of ash in the small class is $r_{ac} = 10 \mu m$, and $r_{ar} = 100 \mu m$ in the large class. The smaller class contributes a mass fraction of 20%.

**Figure 16.** Particle size distribution of the Askja D ash. The log-normal distribution given by WB (thin solid line) is shown. The two gamma distributions (dashed and dotted lines) represent the particle size distributions of the two ash classes in ATHAM, which have been fitted to the WB data.

Highly efficient particle growth leads to the formation of millimeter-sized aggregates, as shown in figure (17).
Figure 17: Radii of particles in the large mode of frozen particles. Shown at 30 min for the experiment ASK.

The increased particle size in the larger class, and the greater size difference between the two classes (i.e., an increased differential fall velocity) in this experiment causes significantly enhanced particle growth, because the mechanism of gravitational capture becomes more efficient. Consequently, sedimentation is much more pronounced than in the reference experiment. At the end of the simulation, about 75% of the erupted mass is deposited to the ground. The formation of two ash layers is less obvious, and the stratospheric mass amounts to about 5%. In addition, the enhanced sedimentation reduces the horizontal expansion of the umbrella cloud by 20 km compared to the reference experiment, after 30 min of eruption.

We conclude that the knowledge on the initial particle size distribution is needed in addition to the information on the mass eruption rate in order to assess the ash dispersal during explosive volcanic eruptions.

4.6. Ambient Conditions

The impact of the environmental conditions on the plume’s shape and height has been investigated by Herzog (1998); Graf et al. (1999). Their experiments revealed, that the plume height is not only determined by the composition and temperature of the erupting material and its vent exit velocity. But it is also dependent on the actual meteorological conditions in the environment in terms of atmospheric stability and relative humidity. The latent heat release due to phase changes of water contributes a considerable amount to the plume’s total thermal energy and therefore influences its vertical evolution. The subject of this section, however, is the effect of the ambient conditions on ash aggregation efficiency.

We use various vertical profiles for temperature and humidity in the background atmosphere, but leave the volcanic forcing the same as in the reference experiment as shown in figure (18).

The experiments called SAS, SAW, MLS and MLW reflect subarctic summer, subarctic winter,
Figure 18.: Atmospheric profiles for temperature and relative, and specific humidity (McClatchey et al. (1972)) in the sensitivity experiments regarding the ambient conditions. The legend is explained in the text.

midlatitude summer and midlatitude winter conditions, respectively (McClatchey et al., 1972). The subtropical ambient conditions of the reference study, REF (see 1), are modified in the experiments STH and STW, where we increase the temperature and the humidity, respectively, to simulate daily variations. In the experiment TRO we increase both the temperature and the humidity as in the previous two experiments, to resemble tropical conditions.

Figure (19) displays the mass of condensed hydrometeors (liquid water + ice) during the course of the simulation. It is in general accordance with the specific humidity in the atmosphere, although this is not exactly true for the arctic and midlatitudinal conditions. The amount of ice in the model is about two orders of magnitude higher than that of liquid water in all sensitivity studies. Aggregation of water-coated particles is insignificant. Thus, we do not find an impact of the amount of liquid water on the aggregation efficiency.
Figure 19: Mass of total liquid water and ice in the model during the course of the simulation. The vertical line at 30 min indicates the end of the eruption. Shown for the sensitivity studies regarding the ambient conditions.

Figure (20) shows the fraction of particles in the larger size classes during the course of the simulation.
Figure 20.: Mass fraction of particles in the larger size classes during the course of the simulation. The vertical line at 30 min indicates the end of the eruption. Shown for the sensitivity studies regarding the ambient conditions.

The aggregation efficiency increases in the same sequence as the amount of condensed hydrometeors in the eruption column shown in Figure (??). It is strongest in the experiment TRO and least effective in the experiment SAS. After the collapse of the eruption column at about 40 min simulation time, aggregation persists to some degree in the experiments SAW, MLW, MLS, and especially in SAS. This could also be caused by the warmer stratospheres in these experiments when compared to tropical conditions, and by the higher relative humidities between 4 and 8 km, see figure 18, which leads to aggregation of the settling particles in the lower troposphere.
The HNB of a volcanic eruption column is dependent on the volcanic forcing, the release of latent heat from the condensation of both volcanic and ambient water vapor, and on the stability of the atmosphere. In this series of experiments, however, it is mainly determined by the strong volcanic forcing, and is situated between 20 and 23 km (in SAS it is somewhat lower). The altitude of the NBH increases slightly with increasing specific humidity in the ambient atmosphere. Figure 21 shows the mass fraction of erupted ash above a certain height at 90 min simulation time. All curves show a layer of small particles in the stratosphere at HNB, and a layer of larger, aggregated ash in the troposphere, as described for the reference experiment in figure 3. The altitude of the lower edge of the settling ash layer decreases in general with increasing aggregation efficiency. Sedimentation is most pronounced in the experiment TRO. The studies STH and STW are similar to the reference experiment. Sedimentation is at least effective in SAS, where the tropospheric ash layer is about 5 km higher than in the reference experiment, and a higher fraction of particles remains at the HNB.

The interpretation of the results of this series of sensitivity study is quite different, because of the complex mutual dependencies of macroscale physical and microphysical quantities. The stability of the atmosphere...
influences the dynamics and the temperature distribution of the eruption column. The ambient humidity affects the amount of latent heat release and, thus, the plume dynamics. At the same time, the aggregation efficiency is changed, which in turn determines the sedimentation of ash and the structure of the flow in the vicinity of the eruption column. In addition, not only the specific humidity influences the aggregation efficiency but also the relative humidity. It is not possible to assign individual microphysical effects to a single ambient parameter.

Our results show the influence of the ambient conditions on ash microphysics in the eruption column. During explosive volcanic eruptions with high columns the aggregation of frozen particles is highly dominant, and the amount of ice determines the amount of aggregation. The process seems to be enhanced at warmer stratospheric temperatures, but the temperature dependence is not well established, see section (4.3), and awaits detailed investigation. The amount of hydrometeors is strongly enhanced in phreatomagmatic eruptions, and we would assume a much more efficient particle aggregation.

5. Discussion and Conclusion

The microphysics of hydrometeors and ash particles in an explosive volcanic plume has been studied by numerical experiments with the plume model ATHAM. An extended version of the microphysics has been applied: based on a two-moment scheme it describes the interactions of hydrometeors and volcanic ash in the plume. The experiments have been performed in a two dimensional model version in cylindrical coordinates without cross wind effects. It corresponds, therefore, to the special case of an eruption without background wind in the ambient atmosphere. We did not try to simulate a specific volcano, but chose conditions typical for highly explosive eruptions.

Our results show that by far the highest portion of condensed water freezes to ice in the eruption column. The fast plume rise to regions, which are too cold for even supercooled liquid water to exist causes most particles to occur as ice-ash aggregates. The total mass of tephra in the model domain is about 100 times that of condensed hydrometeors during the eruption. Hence, the hydrometeor-ash aggregates are rather dry, with an ash fraction in the aggregates usually higher than 80 wt.%. The small relative humidity in the lower troposphere in our scenario leads to total evaporation of condensed water or ice and results in the deposition of completely dry aggregates. No traces of ice or water would be observed in the fallout of the simulated eruption, although it has played a significant role during the transport in the plume. Similar little evidence could be found for the history of ice by remote sensing, because it needs an extremely high ice content to distinguish its signals from that of silicate particles in the plume (Rose, 1999).

The interaction of hydrometeors and volcanic ash during transport in the eruption column can lead to very efficient particle growth. The aggregation efficiency depends on a number of parameters that are discussed below.
Aggregation is governed by the availability of hydrometeors at the particles’ surface, which determines the collection efficiency. Our results show that the particle morphology is a key parameter, because it determines the amount of hydrometeors acting as adhesive for aggregate formation. Particle microphysics has also been found to be sensitive to the temperature dependence of the collection efficiency of frozen particles. These results indicate an urgent need of research about the porosity and the surface properties of volcanic particles, especially at low temperatures, and their aggregation properties within eruption columns. The parameters applied for the collection efficiency should be improved by observational data, which are not yet available.

We find an influence of the atmospheric background conditions on ash microphysics in the eruption column. The amount of condensed ice, which is in turn dependent on the specific humidity in the ambient atmosphere, influences the degree of aggregation. Aggregation seems to be enhanced at warmer stratospheric temperatures, but the temperature dependence of the aggregation of icy particles is not well established, and awaits detailed investigation. We did not find a strong sensitivity to the amount of liquid water present in the eruption column, although the collection efficiency for water-coated is much higher than for icy particles. This is caused by the overwhelming dominance of frozen hydrometeors in all experiments.

The parameterization of the interaction between ash and hydrometeors applied in this study is based on the assumption that volcanic particles are always active as condensation nuclei for liquid water and ice. This assumption is justified, since ash particles are relatively large, and they are often coated with – sometimes hygroscopic – salts, but it has not yet been proven by observations. We tested our parameterization in a sensitivity study where we reduced the threshold temperature for ice nucleation from $T < 0^\circ\text{C}$ to $T < -15^\circ\text{C}$, and found that the results are not modified significantly. This can be explained by the fairly low temperatures well below $-15^\circ\text{C}$ in most regions of the plume in our simulations.

In our parameterization, particle aggregation is caused by the gravitational capture mechanism, which is believed to be more important in volcanic eruption columns than Brownian and turbulent aggregation (Textor and Ernst, 2003). We investigated, however, the impact of electrostatic forces on ash aggregation, which is most important for small dry particles. Since the ATHAM model does not deliver any information on the electric field during the eruption, we set constant collection efficiencies for small, dry ash in the autoconversion processes, and performed sensitivity experiments where we varied these constants. The simulations show that electrostatic forces strongly increase the fraction of small ash particles scavenged by larger ones, and that this process is highly dependent on the choice of the electrostatic collection efficiency. However, in spite of the enhanced particle aggregation, the sedimentation pattern remains quite unaffected, and the size and fall speed of the aggregates does not change significantly. The modified ’electrostatic autoconversion’ intensifies the transfers of small particles to aggregates, but all particles have the same growth probability. Non-linear particle
aggregation, which would promote the growth of single, larger particles at the cost of others, is not affected by electrostatic aggregation in our study. This result is true for invariable electric fields and constant electric properties of the aggregating particles. We believe, however, that in reality none of these will be constant, and thus, preferential particle growth might occur. Our study provides a first assessment of the microphysical effects of the electrostatic aggregation mechanism. Research on electric properties within volcanic eruption columns are required to improve our knowledge in this field.

The information about particle size distribution upon eruption is rather limited at the moment. Our experiments indicated a major influence on the mechanism of gravitational capture, which becomes is enhanced through higher fall velocity differences. Thus, particle growth and sedimentation is strongly affected by the erupted particle sizes. Volcanoes with identical the mass eruption rate but different particle size distribution at the vent can show completely different plume dispersals in the atmosphere.

Many uncertainties exist about the formation and stability of ash aggregates. The parameterization in this study does not include breakup processes. Large aggregates may be destroyed due to hydrodynamic instabilities during precipitation or due to collision with other particles, similar to the breakup of large rain drops. Water evaporation in the relatively dry lower troposphere can lead to drying of settling particles, this way possibly disconnecting some aggregates.

The sedimentation of aggregated ash strongly influences the shape and height of the eruption column. In addition, the removal of small ash particles by aggregation from the HNB decreases the density of this layer, and the remaining volcanic emissions in the stratosphere are lifted upwards after the end of the eruption. This leads to an increased separation of volcanic ash and gases. Thus, particle aggregation determines the residence time volcanic emissions in the atmosphere, their injection into the stratosphere, and their deposition behavior.

In this study we have used simple parameterizations for ash aggregation in order to identify the important processes controlling particle scavenging efficiency and the parameters, which determine theses processes. Our assumptions will need to be rigorously tested against laboratory data, which we plan to collect. This survey should be understood as a qualitative process study, it does not aim to give quantitative results. However, it provides new information about the evolution of microphysical processes within the eruption column. The results from this study can now be utilized to design urgently needed detailed investigations by laboratory and field experiments.

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