

Effect of sea salt on dust settling to the ocean

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ABSTRACT

Dust particles frequently become mixtures of mineral dust and sea salt during their transport in the marine boundary layer, consequently growing in size, which causes changes in their settling velocities. In this study, the effect of sea salt on the gravitational settling of dust particles is investigated. Results show that the adhering of sea salt to dust particles can dramatically increase the gravitational settling of the particles, in particular if the particles become larger than 3–4 μm . Estimates with the observational data from six dust events in southwestern Japan revealed that, due to sea salt adhering, the gravitational settling flux of mineral dust increased approximately 14–17% in well-mixed events and 4–6% in less-mixed events, indicating a potential significant effect of sea salt on dust settling and the importance of considering this effect in the schemata of particle gravitational settling when mapping dust flux to the ocean.

1. Introduction

Input of mineral dust from continents to the ocean via the atmosphere plays a crucial role in sustaining the development of marine ecosystems. It provides nutrients to phytoplankton growth over vast areas of the ocean and makes up the gradual loss of soil-derived elements on islands (Chadwick et al., 1999; Bishop et al., 2002). It is therefore of great importance to correctly map dust deposition in the marine atmosphere to assess the roles of atmospheric dust in the global biogeochemical cycle.

Gravitational settling fluxes of particles in the atmosphere depend on the concentration and settling velocities of the particles. The size and density of a particle are two key parameters in determining its settling velocity. Field measurements have shown that sea salt emitted from the ocean could significantly change dust particles and cause the particles to grow in size (Andreae et al., 1986; Zhang and Iwasaka, 2004, 2006). Clearly, this kind of growth can cause changes in the settling velocity of particles, thus modifying dust deposition and making the process of dust settling to the sea surface fundamentally different from that in the continental atmosphere. It has been reported that the number fraction of mineral dust mixed with sea salt ranged from 80 to 90% in the remote marine atmosphere (Andreae et al., 1986) and from 50 to 90% around Japanese islands (Okada et al., 1990; Niimura et al., 1998; Zhang et al., 2003), suggesting the physical significance of the coagulation between sea salt and dust. However, the effect of sea salt has never been considered in estimates of dust fluxes to the ocean.

In this study, the effect of sea salt on dust settling is investigated by calculations, based on Stokes drag law. To investigate the dependence of settling velocity on particle density, particles at a given size are considered as a mixture of mineral dust and sea salt at different mass ratios. To investigate the dependence of the velocity on particle size, dust particles in different size ranges are considered to grow in size due to sea salt adhering. The change of dust gravitational settling fluxes due to the adhering is estimated with the observational data obtained in southwestern Japan.

2. Methods

The gravitational settling velocity of a particle is calculated by using Stokes drag law, the settling of particles is due to gravity and the drag forces are proportional to the relative velocity between particle and fluid, with the assumption that all particles are in spherical shape. According to Stokes drag law with slip-flow correction, the gravitational settling velocity of a spherical particle smaller than 20 μm in still air, V_s , is expressed as

$$V_s = \frac{D_p^2 (\rho_p - \rho_a) g C_c}{18\mu}, \quad (1)$$

where D_p is the diameter of the particle, ρ_p the density of the particle, ρ_a the density of air, g the gravitational acceleration, μ the dynamic viscosity of air and C_c the Cunningham slip-flow correction and Knudsen–Weber term (Jacobson, 1999).

At standard temperature and pressure, ρ_a is $1.29 \times 10^{-3} \text{ g cm}^{-3}$, which is much smaller than that of mineral dust particles ρ_m (2.65 g cm^{-3} for quartz and approximately 2.6–3.0 g cm^{-3} for clay minerals) and that of sea salt ρ_s (2.17 g cm^{-3} for sodium chloride). As an approximation, V_s

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for a particle composed of sea salt and/or soil-derived minerals can be expressed in a simplified form as

$$V_s = c_0 \left(1 + \frac{2.514\lambda}{D_p} \right) D_p^2 \rho_p, \quad (2)$$

where $c_0 = g/18\mu$, and C_c has been written in terms of D_p and the mean free path of air λ (approximately 8×10^{-8} m; Seinfeld and Pandis, 1997).

The density, ρ_p , of a particle composed of soil-derived minerals and sea salt is determined by the mixing degree of the particle, that is, the ratio of the sea salt component to the soil-derived mineral component in the particle. To calculate the density, the ratio, r , is defined as $r = m_s/(m_m + m_s)$, where m_s is the mass of sea salt component, and m_m is the mass of the soil-derived mineral component in the particle. Consequently, $m_m + m_s$ is the mass of the particle, and the density of the particle can be expressed in average as $\rho_p = [r\rho_s + (1-r)\rho_m]$. r is 0.0 for non-mixed particles and approaches 1.0 with the increase of sea salt portion in particles. Thus, the density of the particle can be expressed as $\rho_p = (1 - 0.18r)\rho_m$ with $\rho_m = 2.65 \text{ g cm}^{-3}$ and $\rho_s = 2.17 \text{ g cm}^{-3}$.

To investigate the minute change of V_s of a particle corresponding to its size increase dD_p ($dD_p \ll D_p$), which is supposed to be the result of a surface coating layer by a substance, the increasing rate of V_s corresponding to D_p is derived from eq. (2), with the assumption that the coating layer is homogeneous on the particle surface. The rate is expressed as

$$\frac{dV_s}{dD_p} \approx c_0[(3\rho_e - \rho_p)D_p + 2.514\lambda(3\rho_e - 2\rho_p)] \quad (3)$$

where ρ_e is the density of the coating substance.

Gravitational settling fluxes of mineral dust from the air to the surface are estimated with the data obtained from six dust events in southwestern Japan in 2002. Details of the data were described by Zhang et al. (2006). Here the information for the estimate is briefly introduced. Number concentrations of particles were monitored by two optical particle counters at a site on the southwestern Japan coast with 15 min time intervals from March to May in 2002, a period when Asian dust particles frequently occurred and were widely transported and dispersed over East Asia and the North Pacific. The available ranges of particle number concentrations were 0.1–0.15, 0.15–0.2, 0.2–0.3, 0.3–0.5, 0.5–1.0, 1.0–3.0, 3.0–5.0 and $>5.0 \mu\text{m}$. Because dust particles were rarely detected in the diameter range $>10 \mu\text{m}$ in these events, all particles in the range $>5.0 \mu\text{m}$ are considered smaller than $10 \mu\text{m}$. The average concentrations within 1 h, when particles were collected for individual particle analysis, are applied in the estimate.

The fluxes are calculated with particle concentrations multiplied by their gravitational settling velocity. Surface resistance and turbulence effects are not included because these processes have significant influence only on sub-micrometre or smaller sized particles settling to the surface. Mineral dust particles ob-

served in the marine boundary layer are usually in the range of micrometre, and dust mass is dominated by particles of several micrometres.

Number concentrations of dust particles at different mixing degrees in each event are derived from the particle number concentrations and the results of individual particle analysis reported by Zhang et al. (2006). It is assumed that the number ratios of dust particles with different mixing degrees in each size range are the same as the ratios in total detected dust particles. To obtain the growth of dust particles due to sea salt adhering, a linear relation between the relative growth of dust particles and their mixture degree, $RelativeGrowth = 0.4198MixtureDegree - 0.0432$ with the confidence $R^2 = 0.9321$, is applied; where the $RelativeGrowth$ of a particle is defined as the ratio of its growth due to sea salt to its diameter and the $MixtureDegree$ of a particle is defined as the ratio of the relative mass of sodium and chlorine to that of sodium, chlorine, aluminium, silicon and iron in the particle. This relation was derived from the data published in a previous paper (Zhang and Iwasaka, 2006).

3. Results and discussion

3.1. Influence of sea salt on the gravitational settling velocity

Figure 1 shows the dependence of the gravitational settling velocities of dust particles on their mixture with sea salt. At a given size, V_s is linearly proportional to particle density, and for a mixed particle, it is between the velocities of non-mixed particles and sea salt particles of the same size (Fig. 1a). In the diameter range $<1.0 \mu\text{m}$, the velocities of mixed and non-mixed particles are not very different if they are the same size. In the diameter range $>1.0 \mu\text{m}$, the velocities are somewhat different for particles with different mass ratios of sea salt to soil-derived mineral part, and the larger the particles are, the larger are the differences.

From the point of view of particle growth, the adhering of sea salt to a dust particle could change the settling velocity of the particle. It can be confirmed from eq. (3) with $\rho_e = \rho_s$ that, in the case where sea salt adheres to a particle, the settling velocity of the particle will be increased due to the adhering if the particle is larger than $0.5 \mu\text{m}$ and its density is smaller than 5.0 g cm^{-3} . It is then concluded that the adhering of sea salt to a dust particle will result in the increase of the gravitational settling velocity of the particle because the density of soil-derived mineral dust particles is usually smaller than 3.0 g cm^{-3} . Details of the increases corresponding to the size growth are shown in Fig. 1b. In the range $<3 \mu\text{m}$, the velocity increases gradually as the particle grows. However, the velocity increases rapidly after it becomes larger than 3 or $4 \mu\text{m}$. This indicates that the removal efficiency of a dust particle by gravitational settling will increase remarkably when it becomes larger than 3 or $4 \mu\text{m}$ compared

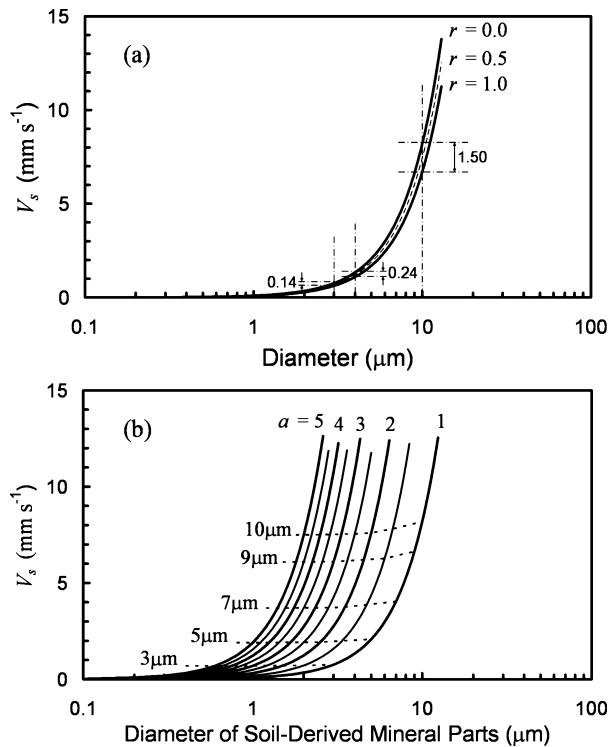


Fig. 1. Dependence of the settling velocity V_s of a particle on the mixing ratios of sea salt and soil-derived mineral dust in the particle. (a) V_s for particles at 0.0, 0.5 and 1.0 mixing ratios [defined as (mass of sea salt)/(mass of the particle); r is 0.0 for mineral dust particles, 0.5 for mixed particles composed of mineral dust and sea salt at the same weight portions and 1.0 for sea salt particles]. The differences between non-mixed particles and sea salt particles at 3, 4 and 10 μm are also shown. (b) V_s for mineral dust particles after they are assumed to be enlarged by sea salt to a folds larger than the size of soil-derived mineral parts in the particles. Results are shown for $a = 1$ to 5 with the increment of 0.5. Note that the line of $a = 1$ is the same as that $r = 0.0$ in (a). Dashed curves mark the referential sizes after the enlargement.

with the stage when it is in the smaller size range if the particle continues to grow in size due to the adhering of sea salt. This result is consistent with the speculation of Zhang et al. (2005), which was based on the observational facts of particle growth and supposed to interpret the consistent mode size ranges of trans-Pacific transported Asian dust particles at different surface sites from Asia to North America.

The size distribution of sea salt particles is usually characterized by a mode diameter at 2–4 μm (O'Dowd et al., 1997), suggesting mixing with sea salt could make dust particles to become larger than 2 μm. The sequence of this sea salt action has been confirmed, and in some cases, sea salt even combined multiple dust particles to produce giant aggregates (Zhang et al., 2005). After travelling from China to Japan, dust particles, in which the mass of adhering sea salt was considerably more than that of soil-derived mineral parts, grew approximately by 1 μm from 2.4 to

3.4 μm on average, and those, in which the mass of adhering sea salt was comparable to that of mineral parts, by 0.4 μm from 2.6 to 3.0 μm (Zhang and Iwasaka, 2004). Therefore, due to sea salt adhering, the gravitational settling of dust particles can be significantly accelerated. This means sea salt from the ocean assists atmospheric dust particles in settling to the ocean. In contrast, there is no similar assistance for dust particle settling in the continental atmosphere.

3.2. Enhancement of gravitational settling flux

Particles in the size range 1.0–10 μm were considered in the estimate of the fluxes. This is because the typical size range of Asian dust particles in the downstream marine areas is 1.0–10 μm. Some dust particles appeared in the range <1.0 μm, but their contribution to the flux in terms of mass was very small compared with particles in the range 1.0–10 μm. With the data of six dust events reported by Zhang et al. (2006), the gravitational settling fluxes of particles in the range 0.1–1.0 μm were estimated as they were dust particles and the fluxes were compared with that of particles in the range 1.0–10 μm. The results showed that the former one was less than 1% of the latter one and could be disregarded.

Figure 2 shows the enhancements of the gravitational settling fluxes due to sea salt adhering in the six events and their dependence to the degree of dust particles mixed by sea salt. The dust events were categorized as well-mixed events, in which dust particles were frequent mixtures of mineral dust and sea salt, and less-mixed events, in which dust particles were considerably less mixed with sea salt. For well-mixed events, the fluxes increased approximately 14–17%; for less-mixed events, 4–6%, indicating a crucial enhancement in some cases (Fig. 2a). Increase of the mixing degree will result in the increase of dust gravitational settling. Comparisons of the enhancement of settling fluxes in dust events with different mixing degrees show a clear proportional relation between the enhancement and the mixing degree (Fig. 2b).

The number ratios of dust particles in which sea salt dominates the mixture were frequently more than 10%, and even more than 30% in well-mixed dust events in southwestern Japan, although at this area, Asian dust plumes had not left the continent far away and were still in the early stages of their long journey over the North Pacific. As more and more dust particles are removed from dust plumes by the settling, and consequently, the ratios of sea salt to soil-derived minerals in dust plumes increase gradually as the plumes are aging in the marine atmosphere, the influence of sea salt on the settling of the remaining dust particles in the plumes can be expected to become more and more crucial to the fate of the particles. The settling of the remaining dust particles from the air to the ocean in the remote areas is thus more closely dependent on adhered sea salt rather than the soil-derived parts in the particles.

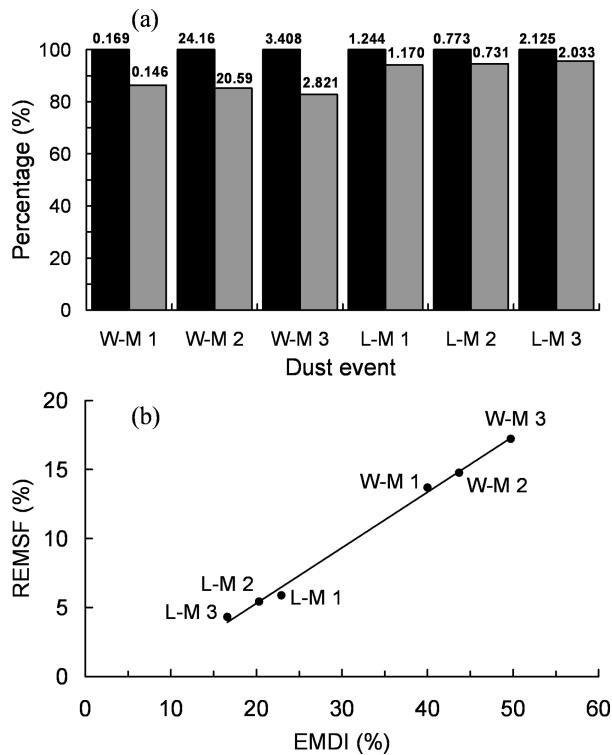


Fig. 2. Enhancement of gravitational dust settling by sea salt adhering. (a) Gravitational settling fluxes of mineral dust (black bars as 100%) and the fluxes assuming no sea salt effect (shaded bars, as ratios vis-à-vis the black bars) in three well-mixed (W-M) dust events and three less-mixed (L-M) dust events. The fluxes in $\times 10^{-7} \text{ g m}^{-2} \text{ s}^{-1}$ are shown above each bar. (b) Relative enhancement of mineral settling flux [REMSF; from (a)] via the equivalent mixture degree index (EMDI). The line shows the linear regression. The EMDI for a dust event was defined as the integral of the number ratios of the relevant mixed dust particles multiplied by the central mixing degree in each mixing degree ranges given by Zhang et al. (2006). It is an indicator for the degree of dust particles mixed with sea salt in a dust event, and the larger it is, the more severe the dust particles were mixed in the event.

3.3. Uncertainties in the estimate

The particle sizes applied here were obtained from the electron micrographs of the particles on the collection grids. The particles were assumed to have a spherical shape in the calculation. Mixed particles should have had a liquid surface coating layer in the air before they were captured onto the collection grids. This is because sea salt particles are usually in aqueous phase or as wet aerosols in the marine boundary layer, due to their high hygroscopicity. Therefore, the sea salt parts in mixed particles could keep the particles, at least as wet particles or droplets in the marine boundary layer. Evidence is available from published photographs of mixed particles collected in the marine boundary layers, which usually showed the morphologies with sea salt crystals in part (e.g. Zhang et al., 2003). Because of the liquid surface layer, mixed particles should have shown a more

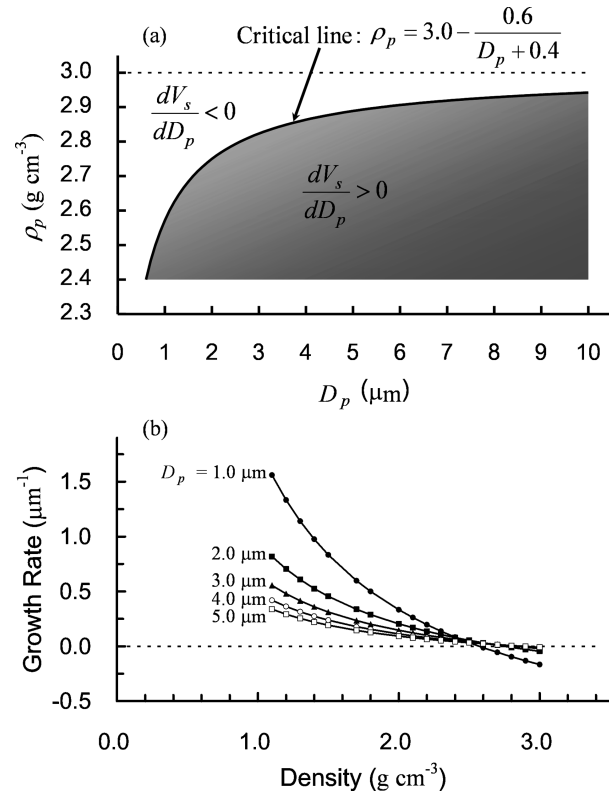


Fig. 3. V_s changes due to minimal surface uptake of liquid water. (a) $\frac{dV_s}{dD_p}$ corresponding to particle diameter. The critical line is derived from $\frac{dV_s}{dD_p} = 0$. (b) Growth rate of V_s relative to diameter ($\frac{1}{V_s} \frac{dV_s}{dD_p}$) at different particle densities. These results are estimated from eqs. (2) and (3) with the density of liquid water 1.0 g cm^{-3} .

spherical shape and exhibited a slightly larger size than those in electron micrographs. The sizes applied in the estimate were somewhat smaller than their size in the air because of the loss of liquid water on the particles and the evaporation of possible volatile species in particle conservation and in electron microscopes. Note that soil-derived parts in the particles had the same shapes on the collection grids as in the air.

It is learnt from eq. (3) that the uptake of liquid water by a particle does not always result in an increase of its gravitational settling velocity. An increase or decrease of the velocity depends on the size and density of the particle (Fig. 3a). However, it can be confirmed that in the density range (ρ_p) $2.6\text{--}3.0 \text{ g cm}^{-3}$ and diameter $D_p > 1 \mu\text{m}$, the growth rate of the gravitational settling velocity of a dust particle ($\frac{1}{V_s} \frac{dV_s}{dD_p}$), in the case of liquid water coating, is very small; in particular when the particle is larger than $2 \mu\text{m}$ (Fig. 3b). A little change of the surface layer due to the loss or uptake of liquid water could not result in a significant change of the settling velocity. On the contrary, if the particles absorb considerable liquid water, the size of the particles will increase considerably and their density will decrease, the result of which will be an increase of their gravitational settling velocity

(Fig. 3b). Most of the dust particles as used in the estimate were larger than $2.0\ \mu\text{m}$. If their major compositions were clay or quartz, their density should be about $2.6\text{--}3.0\ \text{g cm}^{-3}$. For such particles, their settling should have been somewhat underestimated because of the ignorance of the possible liquid surface layer coated on the particles. For this reason, the enhancements shown in Fig. 2 are more likely the lower bounds. Including the effect of mixed particles as wet aerosols will show more significant enhancement.

Another concern on the estimate of the enhancement is the uncertainty due to the assumption of spherical shape of the particles. Recent studies on dust particles settling to surface by Li and Osada (2007a) suggested that the settling velocity of spherical particles is smaller than that of ellipsoids with the same surface area, and the preferential setting of elongated particles lead to the increased proportion of dust particles with large circularity observed in Japan (Li and Osada, 2007b). This implies that dust particles with large circularities are favoured to be transported far away from their source areas, and dust particles settling to remote marine areas exhibit a more round shape rather than a very elongated shape. Ginoux (2003) investigated the settling of dust particles as in spherical shape and as in non-spherical shape and reported that changing the particle shape from spherical to non-spherical made little difference to the results in cases where the non-spherical shape is not too elongated. Shapes of the dust particles used for the above flux estimate were investigated with their median aspect ratios (b/w , where b is the longest dimension of a particle and w is its orthogonal width), circularity factors ($4\pi S/l$, where S is the measured surface area and l is the periphery length of the particle) and ellipse deviations [$(4\pi bw - S)/S$, showing the deviation of the two-dimensional shape of the particle from an ellipse]. These shape indexes were suggested and described by Okada et al. (2001). Results show that rare particles have remarkable elongated shapes. More than 90% of the particles have their aspect ratios smaller than 2.0 with about one third of the particles in the range from 1.2 to 1.4, more than 95% have their circularity factors larger than 0.5 and approximately 90% have their ellipse deviation smaller than 0.2. The investigation of Ginoux (2003) shows that non-spherical shape of particles has considerable influence on the settling only if the aspect ratios of the particles are as large as 5. Therefore, the assumption of spherical shape for the particles in the estimate of dust to sea surface is not expected to cause large deviations in the absolute values, and its influence on the relative values of the estimate shown in Fig. 2 should be small.

4. Summary

By using Stokes drag law, the influence of sea salt on the gravitational settling velocities of dust particles was investigated. In the diameter range $< 1.0\ \mu\text{m}$, the velocities of mixed and non-mixed dust particles are not very different if they are the same size. In the diameter range $> 1.0\ \mu\text{m}$, the velocities are somewhat differ-

ent for particles with different mass ratios of sea salt to mineral dust, and the larger the particles are, the larger are the differences. In the point of view of particle growth, the adhering of sea salt to a dust particle could dramatically enhance the settling velocity of the particle, in particular, if the adhering makes the particle becoming larger than 3 or $4\ \mu\text{m}$.

With data of dust events observed in southwestern Japan, the enhancement of gravitational settling fluxes of mineral dust due to the mixing of sea salt was estimated. Results showed that the fluxes increased by approximately 14–17% in well-mixed events and 4–6% in less-mixed events. There is a clear proportional relation between the mixing degree and the enhancement of the settling fluxes. These results indicate a substantial enhancement of dust settling flux due to sea salt and the importance of including this effect in the schemata of dust particle gravitational settling in the marine atmosphere. Dust settling schemata applicable to calculating dust deposition to the land surface are not applicable to the deposition to the sea surface, and ignoring the effect of sea salt can cause large inaccuracies in the estimate of dust fluxes to the ocean. Simulations of the flux with numerical models were not qualitative so far because of the ignorance of the effect (e.g. Ginoux et al., 2001). To correctly map the input of mineral dust from the air to the ocean, the effect of sea salt on dust settling needs to be considered, particularly in the remote marine atmosphere, where sea salt may play more important roles in dust settling compared with the estimate of this study. Unfortunately, the mechanisms responsible for the production of mixed particles in the marine atmosphere have not been elucidated. Cloud or fog processes have been suggested to be the major routes for dust particles to combine with sea salt to produce mixed particles (e.g. Niimura et al., 1998), although no direct evidences are yet available for this hypothesis.

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