

Profiles of Ion and Aerosol Interactions in Planetary Atmospheres

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Abstract In planetary atmospheres the nature of the aerosols varies, as does the relative importance of different sources of ion production. The nature of the aerosol and ion production is briefly reviewed here for the atmospheres of Venus, Mars, Jupiter and Titan using the concepts established for the terrestrial atmosphere. Interactions between the ions formed and aerosols present cause (1) charge exchange, which can lead to substantial aerosol charge and (2) ion removal. Consequences of (1) are that (a) charged aerosol are more effectively removed by conducting liquid droplets than uncharged aerosol and (b) particle–particle coagulation rates are modified, influencing particle residence times in the relevant atmosphere. Consequences of (2) are that ions are removed in regions with abundant aerosol, which may preclude charge flow in an atmosphere, such as that associated with an atmospheric electrical circuit. In general, charge should be included in microphysical modeling of the properties of planetary aerosols.

Keywords Ion aerosol attachment · Aerosol charging · Atmospheric conductivity · Planetary dust

1 Introduction

Ion–aerosol interactions are very important in understanding the electrical nature of atmosphere. A direct effect of aerosol charging is the removal of small cluster ions, which reduces the atmosphere’s electrical conductivity (Borucki et al. 1982). Electrical conductivity in an atmosphere is a necessary requirement in permitting an atmospheric electrical global circuit (Aplin 2006a; Aplin et al. 2008).

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Molecular cluster ions, consisting of a few tens of molecules, determine electrical conductivity in the terrestrial lower atmosphere, where weather processes are important. Cluster ions have sufficiently large mobilities to be moved appreciably under the influence of an electric field. The vertical ion concentration profile in the lower atmosphere is determined by various ionizing mechanisms for the production of ions and electrons and the loss processes of these species (Bazilevskaya et al. 2008). Cosmic ray induced ionization, in the lower atmospheres of planets and satellites, lead to the formation of primary negative and positive ions, which readily form ion clusters (Harrison and Tammet 2008). Besides cosmic rays, solar UV photons of very low energies are not absorbed in the ionosphere, and reach the lower atmosphere and surface, causing photoemission of electrons when the energies of the incident photons exceed the threshold energy for photoemission of electrons from particles (Grard 1995; Borucki et al. 2006; Michael et al. 2008).

The cluster ion concentration in a planetary atmosphere is very sensitive to the presence of aerosols, tiny particles suspended in the air originating from various sources. In all atmospheres, aerosols reduce the ion concentrations through the transition of the highly mobile ions into large and massive charged aerosols, by ion–aerosol attachment. As well as ion–aerosol attachment, clusters can be lost by ion–ion or ion–electron recombination. Whereas computation of ion–ion recombination coefficient is relatively straightforward, this is not the case for ion–aerosol attachment coefficient (Hoppel and Frick 1986). Various theories exist for calculating the ion–aerosol attachment coefficient, which depend on the comparative size of the aerosols with the cluster ionic mean free path of the atmosphere. The atmosphere of Venus is very dense and the ionic mean free path is smaller than the aerosol size; this permits ion diffusion to the particle surface. On the other hand, for a tenuous atmosphere like that of Mars, with the ionic mean free path larger than the aerosol size, a kinetic theory approach is required.

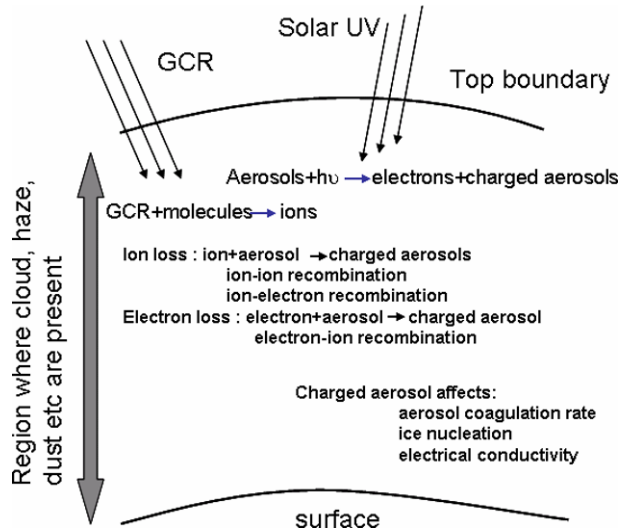
In addition to affecting the conductivity of the atmosphere, aerosol charging reduces aerosol coagulation rate (Harrison and Carslaw 2003), thereby increasing the residence time in the atmospheric column. One consequence is a concentration increase, modifying the optical depth, therefore electrical charging of aerosols is a factor in governing the atmospheric aerosol profiles. Another consequence is an enhancement of aerosol–cloud interactions as the collision efficiency between a particle and a water droplet increases if the particle carries a large charge (Tripathi et al. 2006). In particular, should the drop be supercooled, its freezing probability may be enhanced by the electrically-assisted collection of aerosol facilitating the likelihood of ice nucleation by the contact mode (Tripathi and Harrison 2001, 2002).

In the present review, aerosol charging by ion and electron attachment and the consequences are studied for various atmospheres (see Fig. 1). The review is organized as follows: Ion production in the lower atmospheres of Venus, Mars, Jupiter and Titan is discussed in Sect. 2, Aerosol characteristics (Sect. 3), calculation of ion–aerosol attachment coefficient (Sect. 4), aerosol charging (Sect. 5), and consequences of aerosol charging in atmospheres (Sect. 6). Possible future work is discussed in Sect. 7.

2 Ion Production

Galactic cosmic rays (GCR) have energies spanning a wide range, and contribute to ionization in a planetary atmosphere. GCR have been shown to be the most significant source of ionization (Dubach et al. 1974; Capone et al. 1976, 1977, 1979, 1980), especially in relatively dense regions of a planetary atmosphere with little ionizing solar ultraviolet radiation. GCR originate in interstellar medium, which encounter the solar wind and solar

Fig. 1 Schematic of ion–aerosol processes in a planetary atmosphere. “Top boundary” defines the altitude at which the effect of particles on electrical conductivity ceases; Venus (80 km), Mars (70 km), Jupiter (0.1 mbar), and Titan (400 km)



magnetic field in the heliosphere. In times of higher solar activity the heliosphere deflects GCR from a planet. Therefore, GCR flux is at its minimum during maximum solar activity. The spectrum of the incident cosmic ray flux falls off sufficiently slowly at high energies that a significant portion of the total energy flux is carried by high-energy particles. The largest energy fraction in the cosmic ray flux is typically carried by particles having kinetic energies of at least 1 GeV. Such high energies produce electromagnetic and particle cascades in the atmosphere. A detailed discussion about cosmic rays and their general interaction with planetary atmospheres is presented in Bazilevskaya et al. (2008). Here we consider processes in the planetary atmospheres separately.

2.1 Venus

Cosmic rays incident on the atmosphere interact with the atmospheric gas atoms and molecules. The incident radiation is mainly atomic nuclei, consisting of $\sim 90\%$ protons, $\sim 10\%$ He nuclei, and about 1% heavier nuclei (Upadhyay et al. 1994).

Chen and Nagy (1978) have shown that the ionizing solar ultraviolet radiation does not penetrate much below ~ 120 km, whereas solar flare X-rays are the most important source of ionization in the 60 to 100 km altitude region. Thus, below ~ 60 km or during the Venusian night, galactic cosmic rays are the principal ionizing agent for the atmosphere. The shape of the cosmic ray spectrum is such that a significant fraction of the total energy flux is carried by particles with kinetic energies above 1 GeV. In fact, incident cosmic rays with energies as high as 10 TeV contribute to a significant ionization at penetration depths¹ of ~ 100 g cm⁻². Borucki et al. (1982) used the method developed by O’Brien (1969, 1970, 1971, 1972) to calculate the cosmic ray-induced ionization rates in the Venusian atmosphere. Ionization of the atmosphere by energetic particles produces primary ions CO₂⁺, CO⁺, and O₂⁺ and electrons. Because the collision frequency with neutral species is large, the primary ions and

¹Depth is defined as the integrated mass density of the atmosphere above any given altitude. The total depth of the Venusian atmosphere is approximately 10⁵ g cm⁻².

electrons rapidly form secondary ions and ion clusters. The conductivity of the atmosphere is governed by the mobility of these long-lived secondary ions and ion clusters, rather than by the very mobile, but short-lived, primary ions and electrons. Borucki et al. (1982) estimated that ions such as $\text{H}_3\text{O}^+\cdot\text{SO}_2$ (81 amu), $\text{H}_3\text{O}^+\cdot\text{H}_2\text{O}\cdot\text{CO}_2$ (81 amu), $\text{H}_3\text{O}^+\cdot(\text{H}_2\text{O})_3$ (73 amu), and $\text{H}_3\text{O}^+\cdot(\text{H}_2\text{O})_4$ (91 amu) are the most abundant positive ion clusters. In the atmosphere of Venus, sulfur dioxide and oxygen are the major gaseous species to which free electrons may attach. O_2^- readily transfers its charge to sulfur dioxide and the subsequent reactions of SO_2^- are uncertain. The study of Keesee et al. (1980) suggested that $(\text{SO}_2)_2^-$ would prevail above about 25 km.

2.2 Mars

Whitten et al. (1971) carried out a detailed study of the lower ionosphere of Mars. They considered the ionization by GCR and solar radiation in an ion-neutral model, and calculated the concentration of ions and electrons below 80 km. Molina-Cuberos et al. (2001) improved the study of the lower ionosphere of Mars by developing a more detailed ion-neutral model which includes more neutral compounds and improved reaction rates than previous studies. Molina-Cuberos et al. (2001) calculated the cosmic ray ionization rates of CO_2 , N_2 and Ar. The ionization by cosmic rays in the Martian atmosphere is mainly due to the slowing down of protons and Molina-Cuberos et al. (2001) used measurements of the dissociative and non-dissociative ionization of CO_2 and N_2 by proton impact. The maximum ion concentration occurs at the surface. Hydrated hydronium ions $\text{H}_3\text{O}^+(\text{H}_2\text{O})_n$ (n varies from 1 to 4) are the most abundant positive ions for all altitudes below 70 km. CO_2^+CO_2 becomes important at altitudes more than 65 km. The most abundant negative ion is $\text{CO}_3^-(\text{H}_2\text{O})_2$. Recently, Haider et al. (2008) studied in detail the chemistry of the ion production at high latitudes in the atmosphere of Mars.

2.3 Jupiter

In the Jovian atmosphere (from 10 bar to 1 mbar) muon flux component of galactic cosmic rays are the main ionization source (Whitten et al. 2008). They penetrate to the deep atmosphere at levels where the total number density exceeds 10^{25} m^{-3} and ionize the neutral constituents producing the primary ions H_2^+ , He^+ , CH_4^+ , CH_3^+ , CH_2^+ , and electrons. Fast reactions with the neutrals rapidly convert these into secondary ions and ion clusters. The He^+ ions formed initially are converted into CH_5^+ due to the abundance of H_2 and CH_4 and the inverse temperature dependence of three-body association reaction (Hiraoka and Kebarle 1975). CH_5^+ reacts with C_2H_6 and NH_3 to produce C_2H_7^+ and NH_4^+ , respectively (Capone et al. 1979). The formation of the cluster between NH_4^+ and NH_3 controls the loss process of NH_4^+ in the lower deep troposphere of Jupiter. At equilibrium, the cluster ion $\text{NH}_4^+(\text{NH}_3)_n$ is expected to be dominated by ions with ammonia composition $n = 4$ between 10 bar and 1 bar, but by $n = 2$ ions at 100 mbar due to increasing temperature and decreasing ammonia concentration. $\text{CH}_5^+(\text{CH}_4)_4$ is formed at 0.01 bar, and $\text{CH}_5^+(\text{CH}_4)_2$ is formed at pressures 0.001 bars (Whitten et al. 2008).

2.4 Titan

Since the intensity of the solar UV radiation decreases with the square of the distance from the Sun, GCR-induced ionization assumes greater relative importance in the atmospheres of the outer planets and satellites. GCR-induced electromagnetic and particle cascade penetrate

Table 1 Ion clusters compositions in planetary atmospheres

	Venus	Mars	Titan	Jupiter
Positive ion cluster	$\text{H}_3\text{O}^+\cdot\text{SO}_2$, $\text{H}_3\text{O}^+\cdot\text{H}_2\text{O}\cdot\text{CO}_2$, $\text{H}_3\text{O}^+\cdot(\text{H}_2\text{O})_3$, $\text{H}_3\text{O}^+\cdot(\text{H}_2\text{O})_4$	$\text{H}_3^+\text{O}(\text{H}_2\text{O})_n$	$(\text{H}_2\text{CN}^+(\text{HCN})_3)$	$\text{NH}_4^+(\text{NH}_3)_n$, $\text{CH}_5^+(\text{CH}_4)_n$
Negative ion cluster	$(\text{SO}_2)_2^-$	$\text{CO}_3^-(\text{H}_2\text{O})_2$	–	–

to relatively great depths (i.e. high pressures) in the extended, massive atmosphere of Titan. Ionization also occurs in the atmosphere of Titan through particles precipitating from the magnetosphere of Saturn.

The ion clusters formed in the lower atmosphere of Titan were studied by Borucki et al. (1987, 2006) and their masses estimated to be 30 to 102 amu. $\text{HCO}^+\cdot\text{H}_2$ and $\text{CH}_5^+\cdot\text{CH}_4$ are the major positive ions at altitudes below 80 km. C_7H_7^+ and $\text{H}_4\text{C}_7\text{N}^+$ are the most abundant positive ion clusters for altitudes 80 to 260 km, and 260 to 400 km, respectively. Studies showed that there is a lack of electrophilic species in the atmosphere of Titan (Borucki et al. 1987, 2006; Lara et al. 1996; Molina-Cuberos et al. 2000), and there is therefore a large abundance of electrons. During the descent of Huygens probe through the atmosphere of Titan on 14th January 2005, electron conductivity and density profiles were derived using the Huygens atmospheric structure instrument (HASI). This carried two different instruments, a relaxation probe and a mutual impedance probe (Hamelin et al. 2007). The observations suggest that the electron concentration is at least a factor of two lower than that predicted by previous studies (Borucki et al. 1987, 2006; Molina-Cuberos et al. 1999). Though it was generally accepted earlier that electrophilic species are absent in Titan's atmosphere, the recent observations by HASI suggest that they could be present with a maximum concentration of a few ppm. The most abundant ion clusters in the atmospheres of Venus, Mars, Jupiter and Titan are presented in Table 1.

3 Aerosol Characteristics

Aerosols are sub-micron to micron sized solid particles or liquid droplets suspended in air. In the terrestrial atmosphere, some aerosols are generated naturally, originating from volcanoes, dust storms, forest and grassland fires, living vegetation, and sea spray. Human activities, such as fossil fuel combustion and the alteration of natural surface cover, also generate aerosols. In planetary atmospheres, the sources of aerosols are also very variable.

3.1 Venus

The clouds of Venus are generally thought of as a photochemical haze observed in the altitude range of 48–70 km. According to Rossow (1977) there are two different mechanisms for the formation of these clouds. Near the cloud top, photochemistry leads to the formation of cloud particles; near the cloud base, condensation of sulfuric acid vapor on hydrated sulfuric acid particles is responsible for the particle formation.

In situ probe measurements detected three cloud layers (upper, middle, and lower) based on distinctive cloud particle size distributions of ~ 0.4 , ~ 2 and ~ 7 μm (Esposito et al. 1983; Crisp et al. 1991; Carlson et al. 1993; Grinspoon et al. 1993). The cloud particles have a number density ranging from 100 to 1000 cm^{-3} and the particle size distribution is bimodal

(Knollenberg and Hunten 1980; Pollack et al. 1993; Grinspoon et al. 1993; Krasnopolsky 1989). The thick opaque cloud region between 48–56 km was clearly observed on Galileo and ground-based near-infrared images of Venus (Crisp et al. 1989; Bell et al. 1991; Carlson et al. 1993; Grinspoon et al. 1993). Microphysical processes in the cloud layer are described in detail in various papers (e.g., Toon et al. 1982; James et al. 1997; Imamura and Hashimoto 1998, 2001). An upper haze layer is observed in the altitude range of 70–90 km, containing particles with an effective radius of 0.2–0.3 μm , composed of $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ aerosol of 75% sulfuric acid (Kawabata et al. 1980). A transport model between this haze layer and the atmospheric cloud is discussed in Yamamoto and Takahashi (2006).

3.2 Mars

Dust enters the atmosphere of Mars mainly by the thermally driven wind and then by saltation and strong updraft by the dust devils. Dust particle sizes in the Martian atmosphere have been known since the Mariner 9 and Viking missions (Conrath 1975; Pang and Ajello 1977; Toon et al. 1977). Observations of Martian aerosols from Viking orbiter and lander cameras are discussed in Clancy and Lee (1991) and Pollack et al. (1979). Clancy and Lee (1991) suggested that the effective radius of the dust particles, as seen from the Martian surface, is $\sim 2.5 \mu\text{m}$ and the size distribution of dust is constant at sub-solar latitudes. Vertical profiles of the mixing ratio and size of the dust particles in the 15–25 km altitude range were estimated from the solar occultation measurements performed by the Auguste instrument onboard Phobos 2 spacecraft (Korablev et al. 1993; Chassefiere et al. 1992). The effective radius of the particles was found to be $\sim 0.8 \mu\text{m}$ at 25 km and $\sim 1.6 \mu\text{m}$ at 15 km. Their number density is $\sim 0.3 \text{ cm}^{-3}$ in the same altitude range. Chassefiere et al. (1995) extrapolated the solar occultation profiles from the 15–25 km altitude range down to the ground. An effective radius of $\sim 1.25 \mu\text{m}$ was inferred from the analysis of the dust component observed by the ISM infrared spectrometer (Drossart et al. 1991).

Mars aerosol studies with Mars Global Surveyor Thermal Emission Spectrometer during 1999–2001 have been summarized by Clancy et al. (2003). They suggested that the dust particle sizes vary with latitudes. The properties of dust observed by Mars Pathfinder and the Mars Exploration rovers are discussed in Tomasko et al. (1999) and Lemmon et al. (2004). Montmessin et al. (2006) presented the aerosol size distribution from the SPICAM ultraviolet instrument onboard Mars Express. Goetz et al. (2005) studied the dust using Mossbauer spectroscopy and suggest that the dust contains magnetite, olivine and some ferric oxides. The presence of olivine indicates that liquid water did not play a dominant role in the processes that formed the atmospheric dust.

3.3 Jupiter

Haze in the atmosphere of Jupiter extend in altitude as deep as the 10 bar pressure level to as high as the ~ 0.5 mbar (Owen 1969; Smith et al. 1977; West 1979, 1988; Hord et al. 1979; Smith 1980; West et al. 1981, 1992; Tomasko et al. 1986; Banfield et al. 1998; Friedson et al. 2002). The haze chemistry is thought to involve coupling among hydrocarbon photochemistry, ion-neutral chemistry, auroral processes, and particle microphysics. Strobel (1985) suggested that ammonia photolysis in the lower stratosphere leads to the production and condensation of hydrazine. The suggestion of Hord et al. (1979) that the auroral breakdown of methane leads to the formation and growth of carbonaceous particles has been supported by other workers (Kim et al. 1985; Pryor and Hord 1991; Vincent et al. 2000;

Wong et al. 2000, 2003). Detailed discussion of Jovian atmospheric aerosol is provided in West et al. (1986) with the particles' physical properties discussed further in Mishchenko (1990), Moreno (1996), and Lebonnois (2005). A detailed discussion on the cloud and haze particles in the Jovian atmosphere using the observations of Galileo instruments, ground based and Hubble Space Telescope is provided in West et al. (2004). The optical and physical properties of the haze particles, like the composition, size and shape and the formation of the particles are also discussed in West et al. (2004). Atreya and Wong (2005) provided a good picture of the structure of Jovian clouds through combining the Galileo data, remote imaging and thermochemical models. The Galileo probe detected tenuous cloud layers at 0.5, 1.3 and 1.6 bar levels (Ragent et al. 1998) which most likely represent the clouds of NH_3 -ice, NH_4SH -ice, and H_2O -ice, respectively (Atreya et al. 1999).

3.4 Titan

Spacecraft observations show aerosols are abundant in the Titan atmosphere. Titan's haze particles consist of complex organic molecules, derived from methane and nitrogen photochemistry (Khare and Sagan 1973; McKay et al. 2001; Khare et al. 2002; Atreya 2007). Size information is derived from the Voyager photopolarimeter observations (West et al. 1983), Voyager high-phase-angle images (Rages et al. 1983) and photometry and polarimetry measurements from Pioneer 11 (Tomasko and Smith 1982). Analysis of Voyager 2 images by Rages and Pollack (1983) indicate that between 220 km and 350 km the aerosol particles have radii near 0.3 μm and have number densities that range from about 0.2 particles/cc at 350 km to 2 particles/cc near 220 km. Benzene was detected by the Infrared Space Observatory (Coustenis et al. 2003). Benzene, in a chain of reactions, leads to formation of polyaromatic hydrocarbons (PAH) by continued removal of hydrogen atom and acetylene addition. Cassini instruments detected large positively charged ions (100–350 amu) and negatively charged ions (20–8000 amu) and Waite et al. (2007) confirmed the large negatively charged particles as tholins and predicted the size as ~ 100 –260 nm. Observations of haze particles by Huygens probe Descent Imager / Spectral Radiometer (DISR) were reported by Tomasko et al. (2005). The DISR instrument measured the linear polarization of scattered sunlight and the comparison of polarization with model computations for different sized fractal aggregate particles indicate that the radii of the monomers comprising the aggregate particles is near 0.05 μm , almost independent of the number of monomers in the particle. Contrary to the expectation that the Titan haze clears below an altitude of about 50 km, the DISR downward-looking spectrometer observed significant haze opacity at all altitudes throughout the descent, extending all the way down to the surface (Tomasko et al. 2005).

4 Ion–Aerosol Interactions

The problem of droplet charging by ionic diffusion was originally addressed by Millikan (1911). During the experiment, Millikan (1911) observed that negatively charged droplets were able to catch more negative ions. This was interpreted as evidence that the ions must have sufficient kinetic energy to maintain the surface of the drop against the electrostatic repulsion generated by the charge on the drop. It was concluded that the only way a droplet could lose or gain charge was to capture an ion of appropriate sign. The diffusion of ions to droplets is a real and continuous process. Once captured, ions can never escape because of the work they must do to overcome the local electric image forces. The image force increases faster than the inverse square law and is appreciable for distances considerably less than one mean free path away from the droplet.

Later Arndt and Kallmann (1925) and Frenkel (1946) studied quantitatively the charging of droplets, but the assumptions they adopted were not reasonable. Gunn (1954) estimated the role of ionic diffusion in the electrical charging of drops and noticed that not more than a single ion may be carried by a droplet of radius less than 0.01 μm , unless energetic chemical forces modify. On the other hand, if drops are appreciably larger than the ionic mean free path, relatively many ions may be transferred to the droplet. Gunn (1954) suggested that ions of both signs diffuse into the droplet surface and the ion attachment coefficient can be expressed using the following equations.

$$\beta_+ = \frac{4\pi Q\mu_+}{\exp(Qe/akT) - 1}, \quad (1)$$

$$\beta_- = \frac{4\pi Q\mu_-}{1 - \exp(-Qe/akT)}. \quad (2)$$

Here, β_+ and β_- are the positive and negative ion attachment coefficients, respectively, Q is the free electrical charge resident on the droplet, μ_+ and μ_- are the mobilities of positive and negative ions, respectively, e is the elementary charge, a is the droplet radius, k is the Boltzmann constant and T is the temperature.

The different theories for ion–aerosol attachment are summarized by Fjeld and McFarland (1986). These can be divided into three different types. Diffusion theory applies to particles having radii larger than the ionic mean free path; it assumes ion diffusion to the particles' surface. Here ions are trapped by image force (Keefe et al. 1968; Hoppel 1977). At the other extreme (small particle size) are the free molecular or effusive theories, which use a kinetic approach. Here the attachment occurs via 3-body trapping (similar to the ion–ion recombination). In the intermediate region (size of the particles and the ionic mean free path become comparable), transition regime theory is used. Image capture has a maximal effect for uncharged particles, and its importance relative to the Coulomb force (for charged particles) increases with aerosol particle radius. For aerosols much smaller than the ionic mean free path, attachment is also dominated by image capture, but for discharging events a three-body collision process as in ionic recombination is dominant. In the transition regime, a combination of diffusion and effusion mechanisms is used. The mean free path is smaller than the radius of aerosols in the atmosphere of Venus, whereas the opposite is true in Mars. In Titan's atmosphere the mean free path is smaller for altitudes less than 50 km; above this height the mean free path is larger than the aerosol size.

As discussed in Gunn (1954) the theory of the diffusion of ions to aerosols with radii larger than the ionic mean free path is well established. The study of the attachment of ion to aerosols of radii comparable to or smaller than the ionic mean free path has been more difficult (Hoppel and Frick 1986). The concept of a limiting sphere—concentric with the aerosol but with radius the order of a mean free path larger than the aerosol was defined by Fuchs (1964). The diffusion—mobility treatment holds outside this sphere, with kinetic theory applied for the region inside.

If an aerosol particle carries a single charge then the ion–aerosol recombination coefficient reduces to the ion–ion recombination coefficient and can be explained using 3-body trapping theory, which requires the trapping distance. The ion–aerosol trapping distance is obtained from the ion–ion trapping distance which itself can be calculated from the ion–ion recombination coefficient (Natanson 1960). In addition to the 3-body trapping there can be trapping of the ion by image force. The image force can lead to orbits which spiral into the aerosol, which means any ions which approach the aerosol closer than the image capture distance will spiral into the aerosol under the influence of the image force. The image capture sphere and the resulting attachment coefficient were calculated by Keefe et al. (1968).

Fig. 2 Schematic of image force, and three body trapping distance. Δ is the image capture distance, δ is the three-body trapping distance, a is the radius of the particle and b_{Δ} is the impact parameter

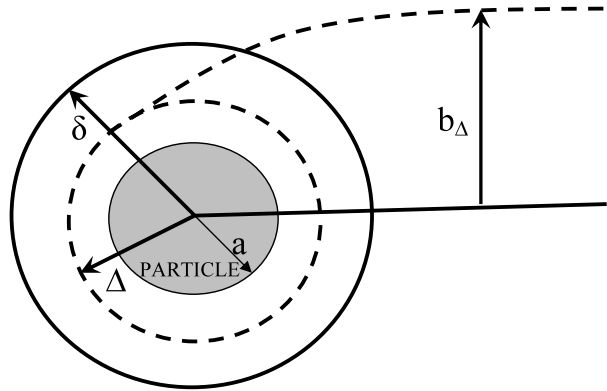
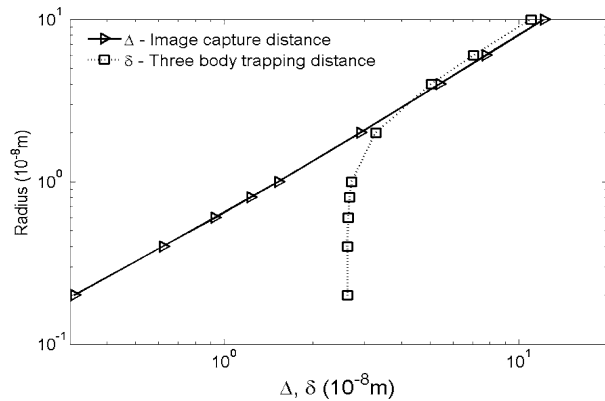


Fig. 3 Importance of image force distance and three body trapping distance with respect to the size of the aerosol. (Data from Hoppel and Frick 1986)



Jensen and Thomas (1991) used the expression developed by Natanson (1960) to calculate the ion capture rates by small ice and meteoric particles in the terrestrial mesosphere.

Hoppel and Frick (1986) estimated the relative importance of image capture and three-body trapping and included both effects in a single theory to calculate the attachment coefficients. Figure 2 demonstrates the two trapping spheres. The image capture sphere extends to the distance (Δ) corresponding to the minimum apsidal distance (i.e. the distance of closest approach where radial velocity vanishes). If the impact parameter is greater than b_{Δ} , the ion escapes, and if the impact parameter is less than b_{Δ} , the ion spirals into the aerosol. When an ion collides with an aerosol particle, part of its kinetic energy is lost to the aerosol. If this energy is sufficiently high the ion will be trapped in the coulomb field of the aerosol. The three-body trapping distance δ is defined as the average separation distance where the removed energy is enough to ensure trapping. If the image capture distance Δ is greater than δ for uncharged aerosol, but if the ion and the aerosol have opposite polarity, there is a critical radius of the aerosol below which the three-body trapping becomes important. Hoppel and Frick (1986) calculated Δ and δ as a function of the radius of the aerosol and the same is presented in Fig. 3. The relative importance of Δ and δ at Venus, Mars and Titan are shown in Fig. 4.

Hoppel and Frick (1986) calculated the attachment coefficients for ionic mass 150 amu and found, for radii less than about 2×10^{-8} m, no aerosol is-doubly charged and, for

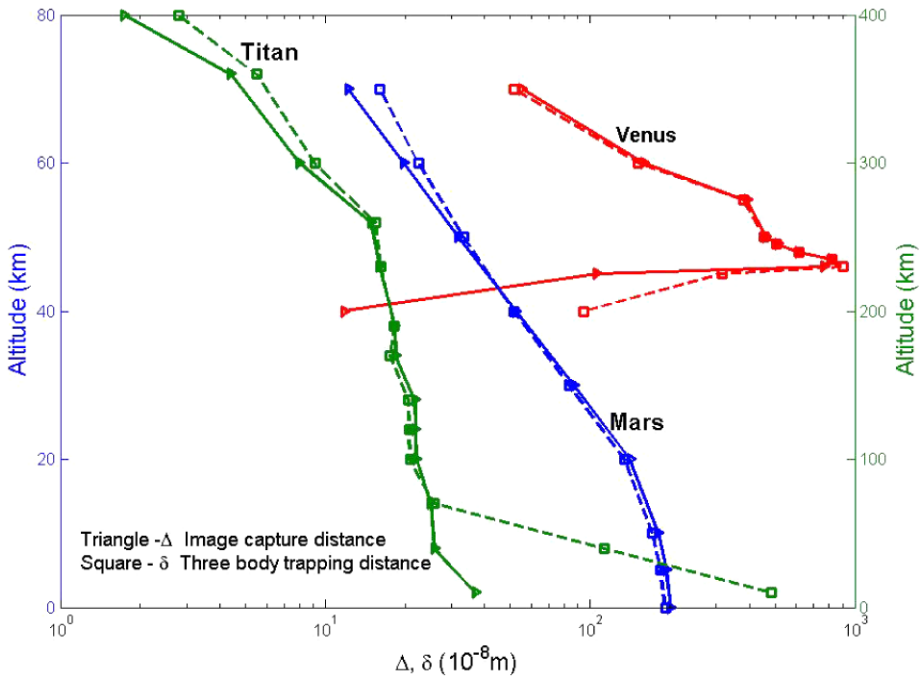
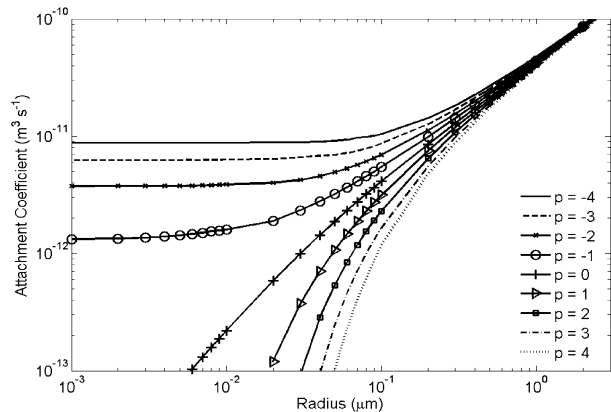


Fig. 4 Relative importance of Δ and δ for various planets. The *green lines* (Titan) correspond to the right y-axis and the *blue* (Mars) and *red* (Venus) lines correspond to the left y-axis

Fig. 5 Ion–aerosol attachment coefficients for ions of 150 amu (Hoppel and Frick 1986)



aerosols less than about 3×10^{-8} m, triply charged aerosols can be neglected. The attachment coefficients as a function of aerosol radius was estimated by Hoppel and Frick (1986), as presented in Fig. 5 and found that the attachment coefficient increases as the radius increases. Tripathi and Michael (2008) describes the calculation of ion–aerosol attachment coefficients using the theory of Hoppel and Frick (1986). Figure 6 presents the attachment coefficients estimated for Venus, Mars and Titan.

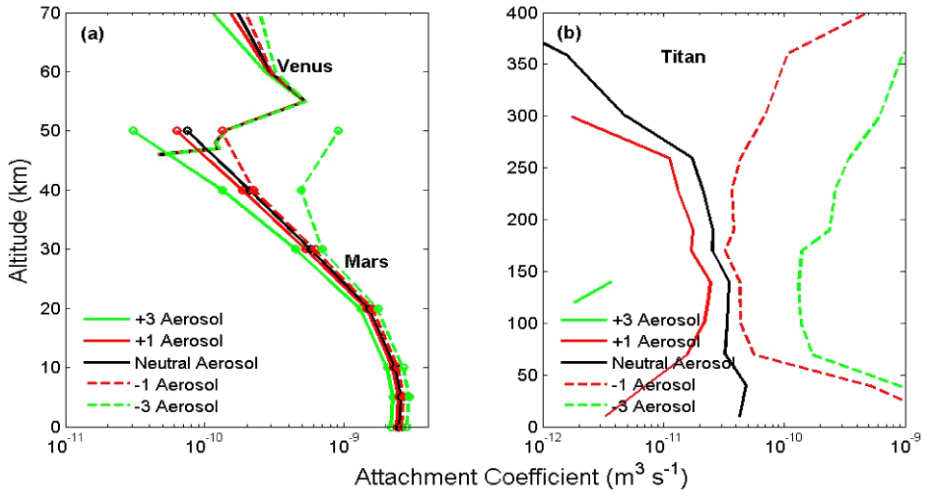


Fig. 6 Attachment coefficients estimated for various planets. **(a)** Venus and Mars, **(b)** Titan. Positive aerosols mean they are of opposite polarity (i.e. “+3 aerosol” means the attachment coefficient of a positive ion to a particle of charge -3 or the attachment coefficient of a negative ion to a particle of charge $+3$). In Venus, the effect of ion–aerosol attachment in the atmospheric conductivity occurs only for altitudes 45–70 km

5 Aerosol Charging

Aerosol charging depends on ion–aerosol attachment coefficients and the ion–ion and ion–electron recombination coefficients. As the attachment coefficients and the recombination coefficients vary with temperature and pressure, and therefore with altitude in atmosphere, the aerosol charging rate also varies with altitude. The concentrations of ions, electrons and aerosols can be found from the three-level probabilistic master equations. These constitute a set of $2s + 4$ simultaneous differential equations, where s is the maximum number of elementary charges allowed on a particle (Yair and Levin 1989). The ion and electron charge balance equations can thus be written as

$$\frac{dn^+}{dt} = q_{\text{ion}} - \alpha n^+ n^- - \alpha_e n^+ n^e - n^+ \sum_i (\beta_1^{(i)} N^i), \quad (3)$$

$$\frac{dn^-}{dt} = q_{\text{ion}} - \alpha n^+ n^- - n^- \sum_i (\beta_2^{(i)} N^i) - F n^-, \quad (4)$$

$$\frac{dn^e}{dt} = q_e - \alpha_e n^+ n^e - n^e \sum_i (\beta_e^{(i)} N^i) + F n^- - n^e \sum_j \beta_j n^j. \quad (5)$$

Here, α is the ion–ion recombination coefficient, α_e is the electron–ion recombination coefficient, q_{ion} is the ion production rate, q_e is the electron production rate, β_j^i is the attachment coefficient for ions of polarity j (1 for positive and 2 for negative) to particles with charge i , β_e^i is the electron attachment coefficient to particles with charge i , β_j is the electron attachment coefficient to species j , and N^i is the density of particles of charge i . F represents the detachment of electrons from negative ions and is evaluated using the equation from

Borucki et al. (1982).

$$F \approx p\pi^{1/2} \frac{L_m}{L_I} \frac{P}{KT} \sigma^2 \left(\frac{8KT}{\pi m} \right)^{1/2} \left(\frac{E_a}{KT} + 1 \right) \exp\left(-\frac{E_a}{KT} \right). \quad (6)$$

Here p is the probability of an energetic collision removing an electron, L_M/L_I is the ratio of the mean free path of neutral molecules to that of ions, P is the pressure, σ is the collision cross section, E_a is the electron affinity of the negative ions, m is the mass of the negative ions, k is the Boltzmann constant, and T is the temperature (Borucki et al. 1982).

Parthasarathy (1976) derived a steady-state recurrence relation to compute the build-up of electric charge on aerosol particles due to collision with positive and negative ions and electrons. Whitten et al. (2007) modified the recurrence expression given by Parthasarathy (1976) to estimate the charge distribution on aerosols and used the method of Jensen and Thomas (1991) to reflect the time dependence of the charge accumulation by aerosols. The time dependent charge balance equations for the aerosols are

$$\frac{dN^i}{dt} = \beta_1^{(i-1)} n^+ N^{(i-1)} + \beta_2^{(i+1)} n^- N^{(i+1)} + \beta_e n^e N^{(i+1)} - \beta_1^{(i)} n^+ N^i - \beta_2^{(i)} n^- N^i - \beta_e n^e N^i \quad (7)$$

where β_e is the electron-aerosol attachment coefficient.

5.1 Venus

Aerosol charging in the Venusian atmosphere is appreciable at altitudes below 80 km. For altitudes above 60 km, additional electrons produced by photodetachment substantially increase the electron concentration. Below this altitude, electrons rapidly attach themselves to neutral molecules, which become more abundant as the atmosphere becomes denser. There is a reduction of 10–20% in the ion and electron densities above 70 km (Borucki et al. 1982).

5.2 Mars

In the Martian atmosphere the charging of aerosols is appreciable below 70 km. Michael et al. (2007) showed that the concentration of charged aerosols decreases as the altitude increases. The aerosols are charged up to 36 elementary charges (i.e. $-36e$ and $+36e$) at 0 km in the atmosphere, but at 70 km the aerosols are charged only up to $\pm 2e$. The concentration of charged aerosols decreases as the charge on the aerosol increases, i.e. the neutral aerosols have a maximum concentration whereas the aerosols with maximum charges have their minimum concentration. It was also noted by Michael et al. (2007) that the steady-state concentration of the aerosols with positive charges is somewhat higher than the corresponding negatively charged aerosols. This is because the positive ions have higher mobility and are attached to the aerosols faster. Michael et al. (2008) studied the day-time charging of aerosols when electrons are also present in the atmosphere along with ions. It was found that more negatively charged aerosols are produced when electrons have a much higher mobility than the ions. More than 80% of the neutral aerosols get charged close to the surface and almost none become charged at the upper boundary (70 km). This is due to the characteristics of the ion–aerosol attachment coefficients and the number density of the aerosols.

5.3 Jupiter

The cloud charging by ion and electron attachment in the atmosphere of Jupiter (for pressures between 5.5 and 0.1 mbar) has been studied by Whitten et al. (2008). For pressures below 4 mbar the electrons are scavenged by cloud particles and thereby increasing the positive ion abundance as the electron–ion recombination decreases. At most atmospheric pressures greater than 2.2 bars, the charge on the low cloud particles is negative and at atmospheric pressures of 2.2 bars and less, the mean charge on the lower cloud particles becomes positive. The charge on the middle and upper cloud particles is negative (Whitten et al. 2008).

5.4 Titan

Aerosol charging during the nocturnal atmosphere for the altitude region 0–400 km is studied by Whitten et al. (2007). During the night, no negative ions are present (Borucki et al. 1987) and for altitudes between 170 and 350 km electrons get attached to the polycyclic aromatic hydrocarbons (PAH, which are similar in mass and mobility with respect to positive ions), and become negatively charged. The densities of positive ions and electrons are similar at altitudes greater than 350 km as the abundance of PAH is very low. At low altitudes the positive ion and electron densities decrease rapidly because of ion–electron recombination. The mean charges attained by the aerosol after the ion-attachment are -6 , -6 and -3 at altitudes of 50, 150 and 250 km, respectively (Whitten et al. 2007). The charging of aerosols was modeled by Borucki et al. (2006) for the day-time atmosphere of Titan. During the daytime, solar UV radiation knocks out electrons from aerosols. It was inferred by Borucki et al. (2006) that the charge distribution of aerosols moves to larger positive values as the altitude increases. About 350 positive charges were acquired by aerosols at 350 km altitude. This is a result of the increased solar UV flux at higher altitudes.

6 Atmospheric Consequences of Aerosol Charging

6.1 Electrical Conductivity

The atmospheric conductivity depends on the existence of positive and negative ions, and the resulting conductivity can be found from the number densities and mobilities of the individual charged species. In the lower atmosphere the conductivity is maintained by the small ions. The attachment of ions to aerosols reduces the conductivity as they become almost immobile by attachment to aerosol particles. The conductivity of the atmosphere is calculated as

$$\sigma = e(n_+K_+ + n_-K_- + n_eK_e) \quad (8)$$

where e is the electronic charge, n and n_e are the number densities of ions and electrons, respectively and K and K_e are the corresponding mobilities. The atmospheric electrification at various bodies in the solar system is reviewed in Aplin (2006a).

Venus: Borucki et al. (1982) modeled the ion–particle interaction and the atmospheric electricity and Aplin (2006b) debated on the existence of the global electric circuit on Venus. With certain assumptions, Borucki et al. (1982) suggested that the effect of particles in the atmosphere for altitudes less than 47 km, on the conductivity is negligible. Between 47 and

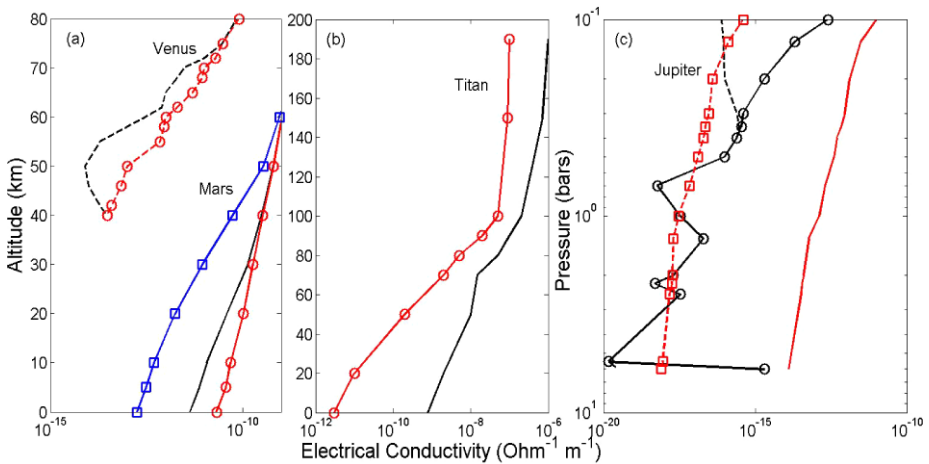


Fig. 7 Effect of aerosols in the atmospheric conductivity in (a) Venus (Borucki et al. 1982), Mars (Michael et al. 2007), (b) Titan (Borucki et al. 2006), and (c) Jupiter (Whitten et al. 2008). Red lines are conductivities of the clear atmospheres and black lines are conductivities in the presence of aerosols. Blue line in (a) shows the effect of dust storm (dust opacity ~ 5) in the atmosphere of Mars (Michael and Tripathi 2008). Solid lines are electron conductivities and dashed lines are ion conductivities in (c)

70 km, the large abundance of particles causes a reduction in conductivity by a factor of about 2–3. The ubiquity of the cloud cover on Venus makes this reduction a global feature. Discovery of frequent lightning discharges (Russell et al. 2007 and references therein) in the atmosphere show the active electrical processes in Venus. The presence of lightning implies that the charge separation mechanisms operate at a rate sufficient to overcome the dissipation of the separated charge by atmospheric conduction. Atmospheric conductivity helps control the local electric field and the current flowing in a global circuit. Figure 7a presents the atmospheric conductivity with and without particles in the atmosphere of Venus.

Mars: Michael et al. (2007) studied the electrical conductivity in the night-time atmosphere of Mars. The maximum variation in conductivity was observed close to the surface (dropped by a factor of five) of the planet and effect of aerosols becomes negligible for altitudes greater than 50 km. The atmospheric conductivity estimated is presented in Fig. 7a. Michael and Tripathi (2008) studied the variation of atmospheric conductivity during the dust storm in 2001. The dust opacity reached a maximum of ~ 5 during the storm, and the conductivity decreased by about 2 orders of magnitude for the maximum dust opacity observed (blue line in Fig. 7a). The atmospheric conductivity during the day-time has been studied by Michael et al. (2008). In addition to ions, electrons are also present in the day-time atmosphere due to the ionization of aerosols by the solar UV radiation. Though the concentration of electrons is about 2 orders of magnitude less than the ions, the high mobility of electrons increases the atmospheric conductivity by a factor of 2.5. But the existing aerosols decrease the conductivity by about a factor of 2 during the day-time.

Jupiter: The consequences for atmospheric conductivity of particle charging by ion and electron attachment in the atmosphere of Jupiter for pressures between 5.5 and 0.1 mbar has been studied by Whitten et al. (2008). Where cloud particles are present, the reduction of the electron conductivity ranges from a factor of 30 at 0.1 bar to 10^4 at 4 bars. At pressures near 1 bar and 4 bars, the positive ion conductivity increases by a factor of 10 over that expected

for the clear atmosphere. Whitten et al. (2008) suggested that negative ions' contributions are likely to be insignificant. The effect of cloud particles in the electron and ion conductivity in the atmosphere of Jupiter from Whitten et al. (2008) is presented in Fig. 7c.

Titan: The atmospheric conductivity of Titan for the night-time was studied by Whitten et al. (2007) and for the day-time was studied by Borucki et al. (2006). For the night-time atmosphere, the conductivity decreases in the diffusion range, where the effective radius of the particles are larger than the ionic mean free path, (below 50 km), while the presence of aerosols does not make a considerable variation in the atmospheric conductivity for altitudes greater than 50 km (effusion range, where the particles are much smaller than the ionic mean free path). During the day-time, a large amount of electrons are produced from aerosols due to the interaction with the solar UV radiation (Borucki et al. 2006) thereby increasing the conductivity by a factor of 2 to more than 2 orders of magnitude. The effect of particles on the electron conductivity during the day-time atmosphere of Titan from Borucki et al. (2006) is presented in Fig. 7b. The direct (HASI) measurements of electrical conductivity in the lower atmosphere of Titan reveal that all models differ from the measurements by a factor about two to three, but the general shapes of modeled and measured profiles are similar (Hamelin et al. 2007). The higher conductivity estimated by Borucki et al. (2006) could be due to the fact that the photoemission threshold used by Borucki et al. (2006) is smaller than the original photoemission cut-off, due to methane absorption.

6.2 Effect of Aerosol Charging on Physical Processes

Scavenging of aerosols by cloud droplets can be enhanced by electrification of the aerosols. Collision efficiency of a particle with a water droplet increases by a factor of 30 for aerosols carrying a large charge (~ 50). For an aerosol population containing a constant fraction of contact nuclei, the droplet's freezing probability can be enhanced by the electrical collection. This is particularly so in the case of small (< 20 microns) super-cooled droplets, or for the few aerosol particles which statistically may transiently carry large charges. A mean charge of $10e$ charges per particle is sufficient to increase the collision efficiency threefold over that for neutral particles (Tripathi et al. 2006). Modest charging of submicron radii aerosols can lead to more collisions with supercooled droplets than neutral aerosol. The change in collision rate occurs from the electrical image force and is, therefore, independent of the charge carried by the water drop (Tripathi and Harrison 2001, 2002). Figure 8 presents the freezing probability with particle charge.

Coagulation of particles in the atmosphere leads to a shift in the aerosol size distribution to larger particles and reduces the build up of extremely high concentrations of ultra-fine aerosols produced by gas-to-particle conversion (Kanawade and Tripathi 2006). The electrical charging of aerosols reduces the aerosol coagulation rate. The lower coagulation rate increases the residence time of the aerosols and thereby increases the aerosol concentration, which affect the optical depth of the atmosphere and hence the atmospheric structure. For ultrafine particles there is a dramatic increase in the coagulation rate if the particles are charged (Harrison and Carslaw 2003). Figure 9 presents the coagulation rate with particle charge. For larger particles the rate of coagulation between particles with like charges is lower than that for neutral particles, while the rate is enhanced for particles with unlike charges (Harrison and Carslaw 2003).

Fig. 8 Freezing probability with particle charge (Tripathi and Harrison 2002)

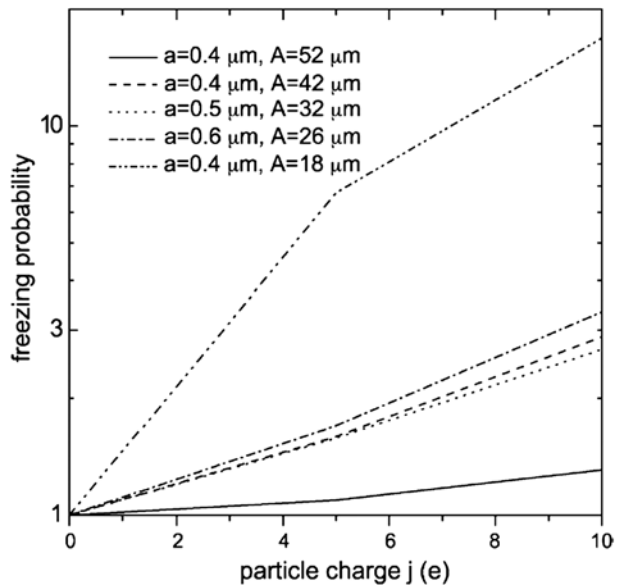
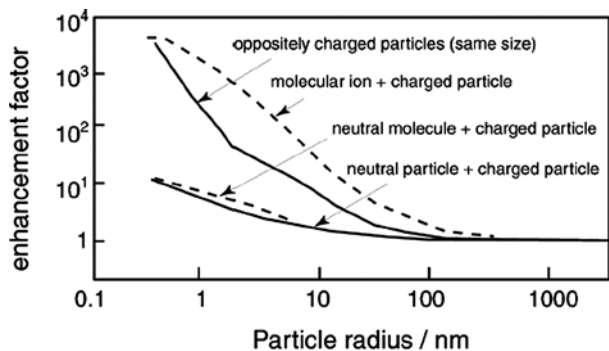


Fig. 9 The enhancement of coagulation rate with particle charge (Harrison and Carslaw 2003)



7 Future Work

Virtually the work on ionic modeling, in regions of interest in the atmosphere of Jupiter from aerosol charging point of view, has not progressed since the seminal work of Capone et al. (1979). Recently the rate constants for ion and ion-neutral reactions occurring in Jupiter's atmosphere have been updated, based on laboratory data. However, these rate constants have not been incorporated into the detailed ion-neutral model of Jupiter's atmosphere. Furthermore, recent detection of electrophilic species leads to the formation negative ion clusters, as shown using simple modeling calculations by Whitten et al. (2008). This needs to be confirmed.

Whereas some experiments have been performed in the laboratory to validate the numerical results pertaining to the aerosol charging in Titan, no such experiments have been performed relevant to Mars. The laboratory simulations for Martian electrification have only been confined to the triboelectric (contact) mechanism (Krauss et al. 2003), however, as shown recently by Michael et al. (2007, 2008) and Michael and Tripathi (2008) that away from the surface dust diffusion charging is the dominant mechanism for calm weather con-

ditions in Mars, which need to be tested, in laboratory on a priority basis. In the terrestrial atmosphere it has been shown that collision rates between electrically charged aerosols and supercooled droplet are greatly enhanced. This should be accounted for while studying cloud formation in the atmosphere of Mars.

Because the smallest particles formed in Titan's ionosphere are electrically charged (Waite et al. 2007), and because aerosol charging can greatly modify the condensation and coagulation processes (Yu and Turco 2001; Toon et al. 1992), it is important to simulate the effect of aerosol charging in an explicit manner in the microphysical models. Planetary aerosol microphysical models currently lack this refinement.

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