

# ATMOSPHERIC ELECTRICITY AND CLOUD MICROPHYSICS

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## **Abstract**

The terrestrial atmospheric electrical system covers a range of dimensional scales, from charged molecular clusters to convective cloud systems. Charge-exchange associated with thunderclouds leads to positive charge in the upper conductive regions of the atmosphere and a net negative charge on the planetary surface. In non-thunderstorm regions, a vertical ionic current flows, replenishing the air with molecular ions otherwise removed by attachment, recombination or nucleation processes. Ions may have indirect effects on non-thunderstorm clouds, and therefore conceivably on climate, via cloud microphysical processes. Cloud Condensation Nuclei (CCN) and Ice Nuclei (IN) are necessary for the formation of water clouds and freezing of ice clouds respectively. In both cases, ionisation may be important: it is now known that ultrafine aerosol can be formed from ionisation, probably providing an additional source of CCN. It is also known that electrified aerosol, perhaps active as IN, can be collected by droplets more effectively than neutral particles.

## **1. INTRODUCTION**

Electrical processes in atmospheric air arise from the combined effect of natural ionisation and the natural electric fields generated indirectly by charge separation in thunderclouds. In non-thunderstorm regions, which probably constitute the majority of the global cloud area, the electrical processes will not generate the large breakdown electric fields associated with lightning, but microscopic aerosol particles acquire charges by diffusion of the molecular cluster ions formed from ionisation.

In this overview, the effect of small charges on aerosol particles and droplets are considered. Since the charge arises from radiolysis of air by cosmic rays and natural radioactivity, the discussion here is structured in terms of the processes associated with charge generation and removal, including tutorial material on microphysical cloud processes. It has been observed (Marsh and Svensmark, 2000) that there is a correlation between low cloud properties and the neutrons produced by cosmic rays.

## **2. THUNDERSTORMS AND GLOBAL ELECTRIFICATION**

The atmospheric electrical *system* originally discussed by Wilson (1929) can be simplified into an electric circuit in which thunderstorms separate charge in convective regions. The charge separation leads to a potential difference between conductive regions of the upper atmosphere and the surface, which causes an ionic leakage current to flow vertically (figure 1). Currents of order 2000A flow in the circuit, with an upper atmosphere potential of ~300kV. The conduction current density in undisturbed regions is ~2pA.m<sup>-2</sup>.

The charge-exchange processes within thunderclouds are complicated, and probably result from the interaction between rising ice crystals rising and riming soft hail (graupel). Typical microphysical collisions exchange charge with typical magnitudes of tens of femtoCoulombs, but the precise magnitude and polarity is greatly influenced by the liquid water content and temperature (MacGorman and Rust, 1998).

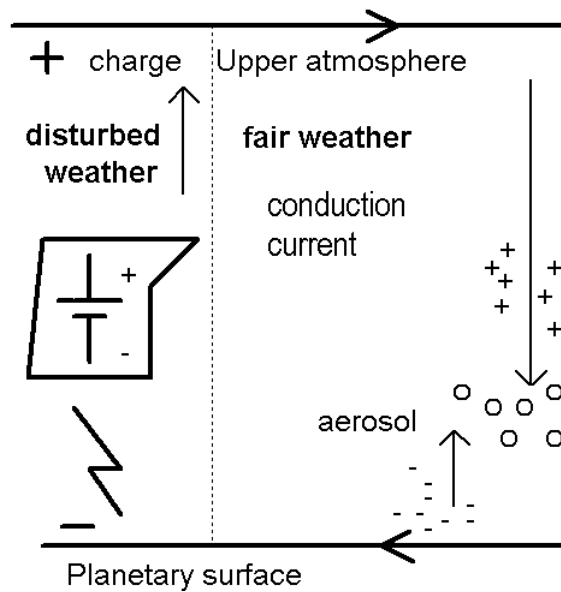


Figure 1. The global atmospheric electrical circuit (from Harrison, 1997).

### 3. ATMOSPHERIC PROPERTIES AND CLOUD MICROPHYSICS

#### 3.1 Atmospheric properties

##### 3.1.1 Bulk properties

The troposphere (lower atmosphere) shows variations in temperature and water content, and partitioning of the water concentration between liquid, solid or vapour forms is critical to the formation and distribution of clouds. Figure 2 shows a vertical sounding of temperature and humidity, which illustrates the atmospheric structure. The presence of low cloud (which was observed from the surface) is evident from the sharp increase in relative humidity, marked as A. B shows a slight temperature inversion associated with the top of the planetary boundary layer, and at C the temperature ceases to fall with height, at the tropopause. It is clear that there is considerable variability in the relative humidity during the ascent, and in the region where cloud was identified optically.

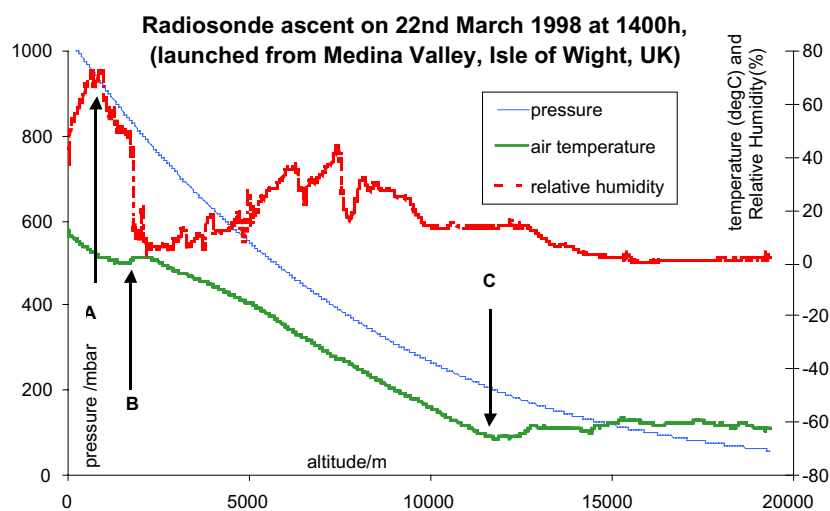


Figure 2. Vertical atmospheric sounding in non-frontal synoptic conditions, showing relative humidity, temperature and pressure variations with height. (A, B and C are discussed in the text.)

### 3.1.2 Microphysical properties

In addition to variability in temperature and humidity, there is a considerable variety in the sizes and abundance of aerosol particles and cloud droplets present in the atmosphere. Figure 3 shows a comparison of the sizes of cloud droplets, raindrops, and a condensation nucleus. The typical molecular cluster comprising an atmospheric small ion will have a diameter less than one nanometre.

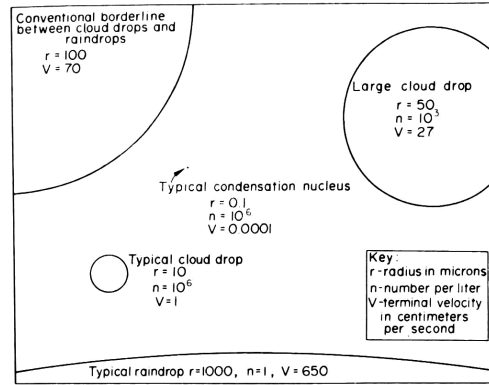


Figure 3. Size spectrum of particles present in a typical cloud. (from Rogers and Yau, 1989).

## 3.2 Cloud Microphysics

The concentration of water vapour in air can be determined by its gaseous partial pressure, and, at any given temperature there is an associated maximum value of partial pressure due to water vapour, the saturation vapour pressure. Air containing sufficient water vapour to generate the saturation vapour pressure is saturated, with a relative humidity of 100%. Slightly greater relative humidities (supersaturations) can occur in localised regions, but they are never greater than a few percent, because of the abundance of aerosol particles on which the water can condense. Many different kinds of aerosol particles are capable of acting as condensation nuclei. Below  $0^\circ\text{C}$  however, liquid water droplets may persist without freezing, although  $0^\circ\text{C}$  is the temperature at which ice melts. Any liquid water droplet with a temperature below  $0^\circ\text{C}$  is supercooled, in a thermodynamically unstable state in which freezing may be readily initiated by heterogeneous or homogeneous nucleation. In heterogeneous nucleation, the supercooled water freezes as a result of the presence of a suitable ice nucleus. Homogeneous nucleation occurs if cooling is continued further, and all supercooled water in atmospheric clouds becomes ice at temperatures colder than  $-40^\circ\text{C}$  by this process.

### 3.2.1 Saturation vapour pressure, temperature and relative humidity

At any given temperature  $T$ , the maximum partial pressure of water vapour, the *saturation vapour pressure*  $e_s(T)$  is given by the Clausius-Clapeyron equation as

$$\frac{1}{e_s} \frac{de_s}{dT} = \frac{\lambda}{R_v T^2} \quad (1)$$

where  $\lambda$  is the latent heat of vaporisation of water, and  $R_v$  is the gas constant for water vapour ( $461.5 \text{ J.kg}^{-1}.\text{K}^{-1}$ ).  $e_s(T)$  can in principle be found by integration from equation (1), but  $\lambda$  is also a function of temperature, which leads to many empirical formulae for  $e_s(T)$ . Common forms include the exponential (Magnus) equation *e.g.*

$$e_s(T) = 6.112 \exp [17.67 T / (243.5 + T)] \quad (2)$$

where  $e_s$  is given in millibars and  $T$  is in Celsius.

The relative humidity is the actual vapour pressure expressed as a fraction of  $e_s$ , at the same temperature. Supersaturation is expressed either as a percentage relative humidity greater than 100%, or as a saturation ratio  $S$ . (101% RH = 1% supersaturation = saturation ratio  $S = 1.01$ ).

### 3.2.2 Activation of condensation nuclei

In the troposphere, supersaturations are never greater than a few percent, and are typically rather less. Consequently direct condensation onto ions, which permits visualisation of particle tracks in a Cloud Chamber ( $S \sim 4$ ), cannot occur in the lower atmosphere. Condensation on aerosol particles, which are larger, does occur, however, and the minimum size of particle necessary depends on the degree of supersaturation. All aerosol particles are therefore potentially able to act as condensation nuclei (CN), if the supersaturation is sufficiently large, but it is the subset of particles able to cause condensation at atmospheric supersaturations which is of interest in cloud physics. These condensation nuclei are known as Cloud Condensation Nuclei (CCN).

The vapour pressure over the curved water surface of a particle of radius  $r$ ,  $e_s(r)$  is greater than  $e_s$  over a plane surface at the same temperature. If condensation occurs on a particle, its growth rate is proportional to the difference in between the bulk vapour pressure  $e$  and  $e_s(r)$ . For  $e - e_s(r) > 0$  the cloud droplet grows. This situation is rather more complicated in a mixed-phase cloud due to the differences in vapour pressure over ice and supercooled water. Ice particles grow at the expense of supercooled water in a mixed-phase system.

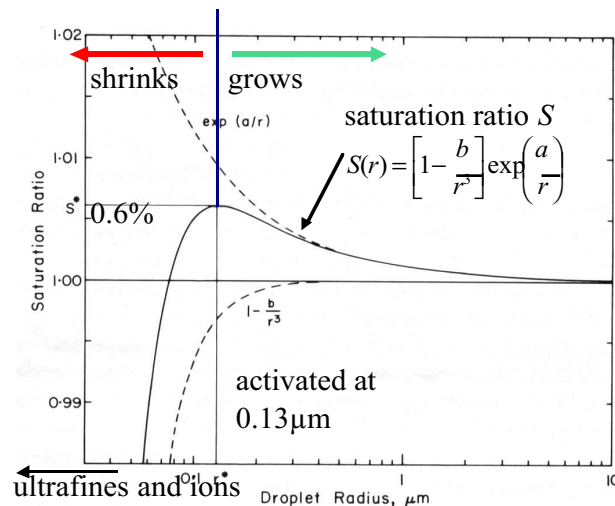


Figure 4. Activation of particles at typical atmospheric saturation ratios. The maximum in the saturation ratio curve  $S(r)$  defines the minimum radius of particle required to act as a nucleus on which a cloud droplet to grow. For a supersaturation of 0.6%, a  $0.13\mu\text{m}$  radius particle is required. A droplet smaller than this will evaporate. The function  $S(r)$  principally depends on a “curvature” term  $a$ , and a “solution” (dissolved salt) term  $b$ . (after Rogers and Yau, 1989).

### 3.2.3 Supercooling and ice nucleation

Supercooled droplets are common under atmospheric conditions, and result from water droplets cooling in the absence of suitable ice nuclei (IN) to permit heterogeneous ice nucleation. At temperatures cooler than  $-40^\circ\text{C}$  all supercooled droplets begin to freeze by homogeneous nucleation.

Only very few atmospheric aerosol particles can act as IN, typically less than 1% although the exact fraction increases as the droplets become colder. The ability of a particle to act as an ice nucleus depends on a variety of physical properties including its shape, solubility, crystal structure and its history in cloud processing. At warmer temperatures ( $-6$  to  $-10^\circ\text{C}$ ) ice multiplication occurs

by mechanical production of ice splinters on freezing, generating additional ice fragments which are also able to act as IN. Figure 5 shows the dramatic temperature change occurring when a supercooled water droplet freezes, releasing latent heat.

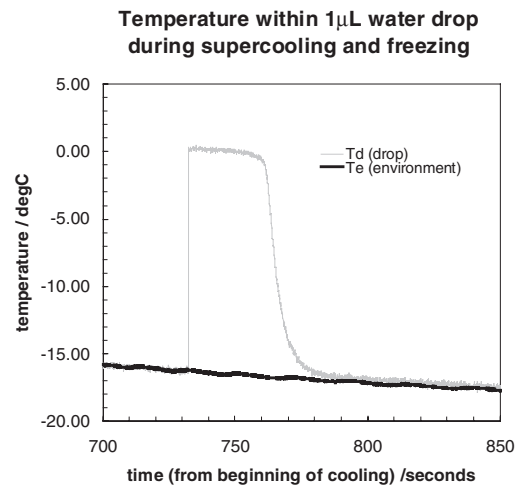


Figure 5. Time series of temperature  $T_d$  within a supercooled water droplet, in an environment at temperature  $T_e$ , as it freezes and releases latent heat (from Harrison and Lodge, 1998).

## 4. ATMOSPHERIC IONISATION AND ELECTRIFICATION

### 4.1 Steady-state ion concentrations

Ion-pairs are continually produced in the atmosphere by radiolysis of air molecules, figure 6. The ions produced are rarely single species but clusters of water molecules around a central ion. Typical atmospheric ion concentrations in unpolluted air and fine weather are about  $500 \text{ ions.cm}^{-3}$  (Chalmers, 1967).

There are three principal sources of high-energy particles which cause radiolysis: Radon isotopes, cosmic rays and terrestrial gamma radiation. The partitioning between the sources varies vertically. Near the surface, ionisation from turbulent transport of radon and other radioactive isotopes is important, together with gamma radiation from isotopes below the surface. Ionisation from cosmic rays is always present, comprising about 20% of the ionisation at the surface. The cosmic fraction increases with increasing height in the atmosphere and dominates above the planetary boundary layer.

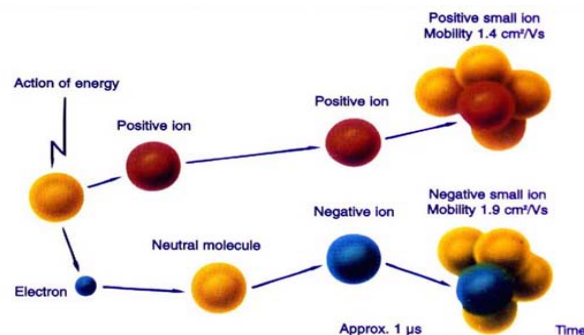


Figure 6. Formation of small ions by radiolysis of air molecules.

Small ions consist of clusters of water molecules collected around a singly charged ion. They have a lifetime of the order of a hundred seconds. Clusters such as  $\text{H}_3\text{O}^+(\text{H}_2\text{O})_n$ ,  $\text{H}^+(\text{H}_2\text{O})_n$ ,  $\text{NO}^+(\text{H}_2\text{O})_n$  and  $\text{NO}_2^+(\text{H}_2\text{O})_n$  are common for the positive ions and  $\text{O}_2^-(\text{H}_2\text{O})_n$ ,

$\text{CO}_4^-(\text{H}_2\text{O})_n$ ,  $\text{NO}^-(\text{H}_2\text{O})_n$  or  $\text{NO}_2^-(\text{H}_2\text{O})_n$  for the negative ions (Volland, 1984). The chemical difference between the species in the positive and negative ions leads to some physical asymmetries in the ion properties, with the negative ions more mobile. The ratio of mobilities  $\mu_-/\mu_+ \sim 1.2$ .

## 4.2 Ion balance equation

Atmospheric small ions of both signs with number concentrations  $n_+$  and  $n_-$  are governed by

$$\frac{dn_{\pm}}{dt} = q - \alpha n_{\pm} n_{\mp} - n_{\pm} \int_{a=0}^{\infty} \sum_{j=-\infty}^{\infty} \beta_{\pm 1,j}(a) N_j(a) da \quad (3)$$

where the ions are produced at a rate  $q$  per unit volume. Ions (which are assumed to carry unit charges) are removed by ion-ion recombination (with recombination coefficient  $\alpha$ ), and by attachment to aerosol particles, which causes charge transfer to the aerosol. The aerosol attachment rate  $\beta_{\pm 1,j}(a)$  depends on aerosol particle radius  $a$  and the number of elementary charges  $j$  present on the aerosol particle of radius  $a$  (Gunn, 1954). In equation (3), the size and charge distributions of atmospheric aerosol particles are accounted for by the integral of number concentration  $N(r)$  over all particle radii, and by a sum across all possible particle charges at each radius. Recombination is the principal loss mechanism of ions in clean, aerosol free air. If aerosol is present, then ions are also lost by aerosol attachment.

It is instructive to simplify the ion balance equation by neglecting the ion sign (*i.e.*  $n_+ \approx n_- = n$ ) and replacing the aerosol particle size distribution by an equivalent monodisperse particle number concentration  $Z$ . The ion-aerosol equation can then be written as

$$\frac{dn}{dt} = q - \alpha n^2 - n \beta Z \quad (4)$$

### 4.2.1 Time dependent solution

Integrating this equation gives the ion concentration  $n$  as a function of time  $t$ , for a zero initial ion concentration at time zero, as

$$n(t) = \frac{\left[ -\sqrt{(\beta^2 Z^2 + 4\alpha q)} - \beta Z \right]}{2\alpha} \left[ \frac{\left( 1 - e^{-\sqrt{(\beta^2 Z^2 + 4\alpha q)} t} \right)}{\left( 1 + e^{-\sqrt{(\beta^2 Z^2 + 4\alpha q)} t} \right)} \right] \quad (5)$$

which highlights two interesting points. Firstly, if the ion-pair production rate  $q$  is uniform and the removal rates are also steady, the ion concentration tends to a steady value for large values of  $t$ . Secondly the equation can be simplified according to the situations in which attachment or recombination dominates as the removal mechanisms, according to whether  $\alpha n^2$  or  $n\beta Z$  is the bigger term. In the atmosphere in polluted air, these terms are roughly comparable, and therefore all the terms in equation (5) have to be evaluated.

### 4.2.2 Recombination Limit

In the case of ion loss solely by recombination, such as in relatively aerosol-free regions of the atmosphere, equation (5) reduces to

$$n(t) = \sqrt{\left[ \frac{q}{\alpha} \right]} \left[ \frac{\left( 1 - e^{-2\sqrt{\alpha q} t} \right)}{\left( 1 + e^{-2\sqrt{\alpha q} t} \right)} \right] \quad (6)$$

and the steady-state concentration after a long time has elapsed is given by  $n_{\infty} = (q/\alpha)^{1/2}$ .

Inserting typical atmospheric values of  $q \approx 10$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$  and  $\alpha = 1.6 \times 10^{-6} \text{cm}^3 \text{s}^{-1}$  gives  $n_{\infty} = 2500$  ion-pairs  $\text{cm}^{-3}$ . Typical values of small ion concentrations observed in mountain air are about 500 ions  $\text{cm}^{-3}$  of each sign, suggesting that attachment processes are almost always significant in modulating the ion concentrations in the lower troposphere.

### 4.3 Aerosol electrification

Collisions between the ions and atmospheric aerosol lead to charge-exchange and electrification of the aerosol, and the ion asymmetry ensures that the collisions do not lead to an average charge of zero. Local electric fields can cause further asymmetries, by depletion of one sign of ion concentration, and consequently substantial aerosol electrification can occur in such regions.

The number concentration  $N_j$  of monodisperse aerosol particles carrying  $j$  elementary charges is given by the Modified Boltzmann Distribution (Clement and Harrison, 1992), as

$$\frac{N_j}{N_0} = \left[ \frac{n_+ \mu_+}{n_- \mu_-} \right]^j \frac{8\pi\epsilon_0 akT}{je^2} \sinh \left[ \frac{je^2}{8\pi\epsilon_0 akT} \right] \exp \left[ \frac{-j^2 e^2}{8\pi\epsilon_0 akT} \right] \quad (7)$$

where  $\mu_{\pm}$  are the positive and negative small ion mobilities,  $n_{\pm}$  their number concentrations,  $T$  the temperature,  $e$  the modulus of the electronic charge,  $k$  Boltzmann's constant and  $\epsilon_0$  the permittivity of free space. The mean charge  $J$  (Gunn, 1955) is given by

$$J = \frac{4\pi\epsilon_0 akT}{e^2} \ln \left[ \frac{n_+ \mu_+}{n_- \mu_-} \right] \quad (8)$$

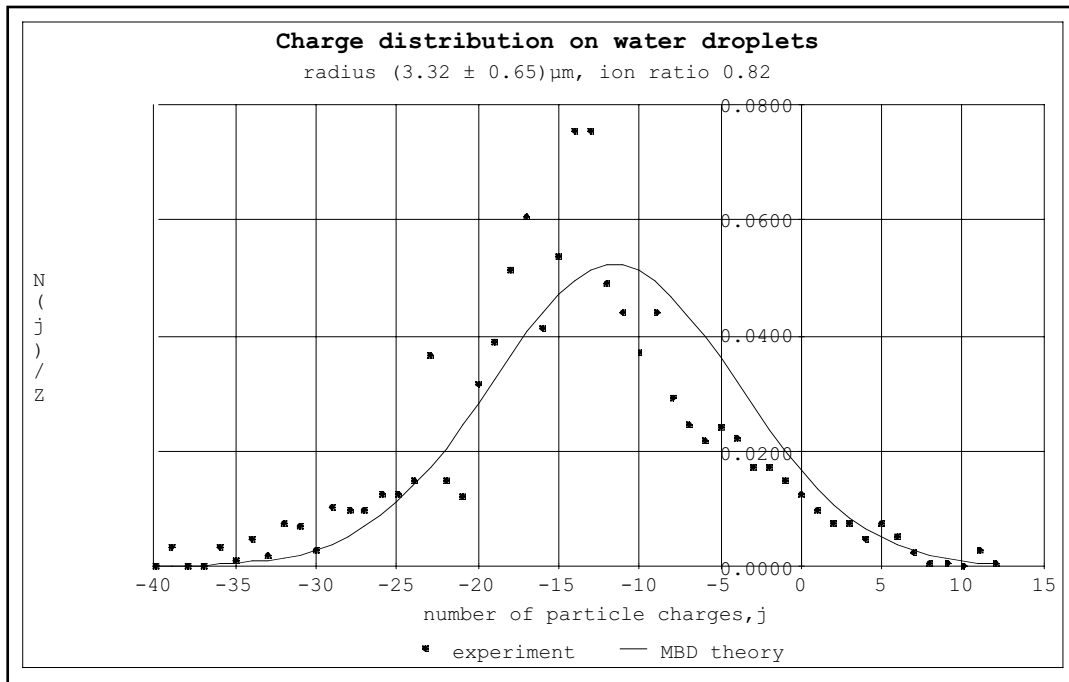


Figure 7. Charge distribution on water droplets in the presence of ion asymmetry using the Modified Boltzmann Distribution (MBD). (Experimental data from Gunn and Woessner, 1956)

## 5. OBSERVED TROPOSPHERIC ELECTRICAL PROPERTIES UNDER NON-THUNDERSTORM CONDITIONS

Many electrical soundings of the atmosphere have been made during disturbed (thunderstorm) conditions, but few such measurements have been made under more quiescent atmospheric conditions. There are difficulties with in-cloud measurements, as a balloon or aircraft platform will be required: this may itself introduce difficulties with sampling, particularly under small electric fields.

Regions of aerosol particles, some of which may acquire appreciable charges, are the principal perturbation to atmospheric electric fields under fair weather conditions. In general the upper and lower surfaces of a horizontal region of particles or droplets will charge from ions flowing vertically as a result of the fair weather conduction current, and the region within the layer will be of low conductivity compared with that of the surrounding air. Figure 8 shows electric field profiles observed in non-thunderstorm clouds, which are typically two orders of magnitude smaller than equivalent profiles determined in thunderstorms.

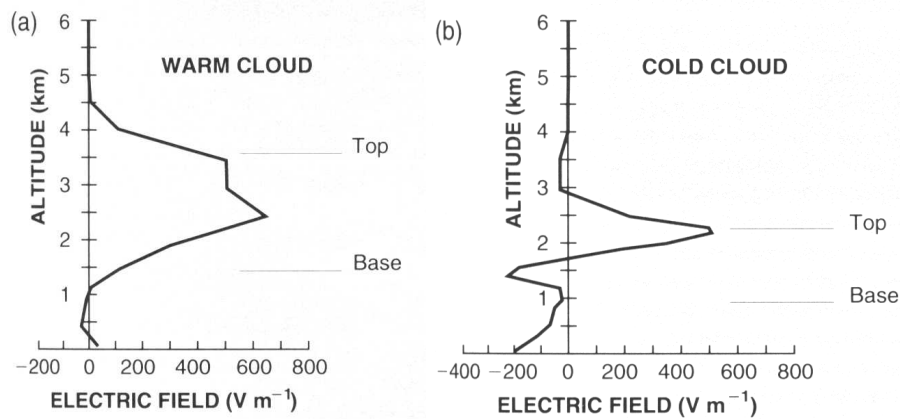


Figure 8. Typical electric field profiles found in non-thunderstorm clouds under (a) liquid water and (b) supercooled conditions (from MacGorman and Rust, 1998).

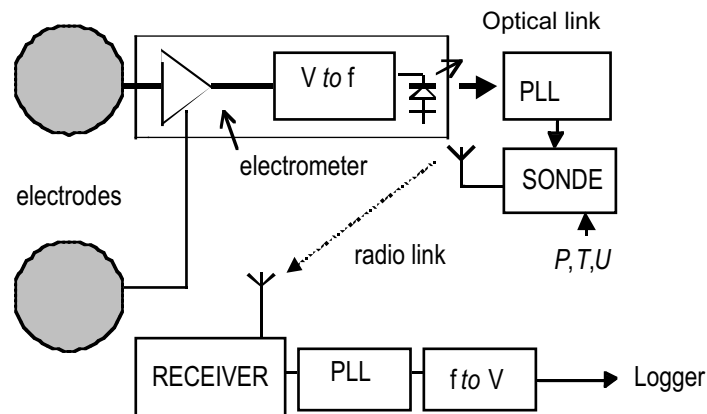


Figure 9. Summary schematic of an atmospheric electric field sensor using vertically-spaced spherical electrodes, flown under a conventional radiosonde balloon. The electrometer circuit and batteries are mounted within the lower electrode, with the data conveyed via a voltage-to-frequency converter and an optical link operating at 100kHz. A phase-locked loop (PLL), recovers the data from an optical receiver, and the signal is injected into a standard meteorological RS80 radiosonde measuring pressure  $P$ , temperature  $T$  and relative humidity  $U$ . The uhf receiver recovers the 100kHz signal, which a further PLL converts back to a voltage. The voltage is logged at 20Hz by a computer and analogue to digital converter (from Harrison, 2001).



A modern sensor suitable for use in low fields using a standard meteorological radiosonde has recently been described (Harrison, 2001), using the displacement current to detect charged aerosol particles from the changes caused in electric field. Figure 9 shows a schematic of the system, which is disposable. Regions of space charge of  $\sim 10\text{nC.m}^{-3}$  were reported in shallow layers, with electrical structures suggesting electrification by the conduction current.

## 6. DIRECT INFLUENCES OF ELECTRIFICATION AND RADIOACTIVITY ON CLOUDS

In considering how natural atmospheric ionisation might influence cloud physics or cloud formation, Harrison (2000) identified two possible routes:

- (1) *direct* processes, such as production of new aerosol (*e.g.* sulphate) by gas-to-particle conversion (GPC) or homogeneous nucleation
- (2) *indirect* processes, such as the modification of existing heterogeneous nucleation processes by affecting the condensation nuclei (CN) or ice nuclei (IN).

### 6.1 Direct processes

#### 6.1.1 CN production

In the presence of high levels of radioactivity, the radiolytic formation of particles has been shown to occur. Bricard *et al* (1968) found the CN concentration could be made to cycle by the regular addition of Thoron (a short-lived source of  $\alpha$ -particles), figure 10. Vohra *et al* (1984) observed particle formation in artificial air in the presence of trace concentrations of sulphur dioxide, ozone and ethene, with naturally-occurring radon concentration levels, suggesting that ultrafine aerosol production could occur in atmospheric air under natural conditions. Recent theoretical work by Yu and Turco (2001), further strengthens the expectation that radiolytic particle production will be found in the atmosphere, and that the conventional ion-aerosol balance equations are incomplete.

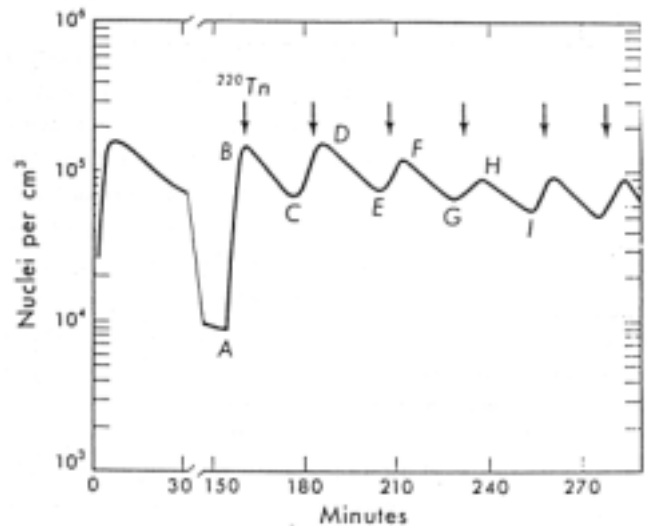


Figure 10. Particles formed in filtered Parisian air, with the addition of regular cycles of Thoron (from Bricard *et al*, 1968).

### 6.2 Indirect processes

#### 6.2.1 Scavenging of charged particles

The removal of aerosol particles by cloud droplets, scavenging, is known to be influenced by many factors, including electrical forces (Pruppacher and Klett, 1997). Figure 11 shows the partitioning of the water drop charge  $Q_d$  in response to the image charge induced by the charged

aerosol particle (carrying a charge  $Q_a$ ) brought close to the drop. Charge conservation requires that  $Q_d = I + D$ . In magnitude,  $I = -(A/s)Q_a$ , where  $s$  is the separation distance between the aerosol and drop centres. The image charge is located at a distance  $c$  from the centre of the drop (Jackson, 1975). Summing the Coulomb and image forces, the net electrical force acting between the particles' centres is

$$F_e = \frac{1}{4\pi\epsilon_0} \left[ \frac{Q_a I}{(b-c)^2} + \frac{Q_a D}{b^2} \right] \quad (9)$$

where a positive  $F_e$  is repulsive.

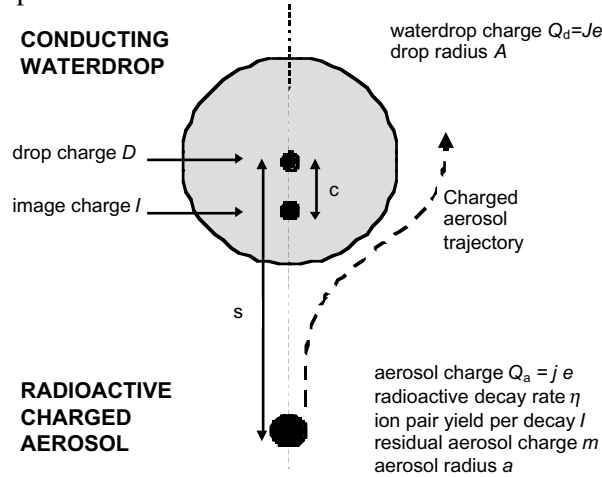


Figure 11. Schematic of the (radioactively) charged aerosol and the image charge  $I$  construction within a water drop of radius  $A$ . The aerosol and falling drop carry charges  $Q_a$  and  $Q_d$  respectively, with  $Q_d = I + D$ , where  $D$  is the non-image charge considered at the centre of the drop (from Tripathi and Harrison, 2001).

### 6.2.2 Electrofreezing

Tinsley *et al* (2000) have shown that the electrical image force is very significant in aerosol-droplet collisions as, unlike the Coulomb force, it is always attractive between the charged aerosol and water droplet at small separations. This process, electroscavenging, is a subset of many processes described more generally as electrofreezing, in which electrical fields or charges influence the freezing of supercooled droplets.

Tinsley and Dean (1991) argued that modification of the electrical properties of aerosols might change their efficacy of aerosol as contact ice nuclei, ultimately leading to storm intensification by triggering latent heat release. Direct ionisation has, however, recently been shown *not* to lead to freezing of supercooled water (Seeley *et al*, 2001). There is currently no definitive evidence that charging influences ice nuclei efficiency or that contact nucleation of ice is the dominant freezing mechanism.

## 7. DISCUSSION

Ionisation in the atmosphere is ubiquitous and part of the atmospheric electrical system, which transports ions in low electrical field regions of the atmosphere, such as clear air and non-electrified clouds. There appear at least two ionisation-related processes of relevance to cloud formation:

- (1) aerosol electrification
- (2) ultrafine aerosol production

## 7.1 Aerosol electrification

Stratified regions of aerosol will charge in the atmosphere under quiescent conditions as a result of ion transport by the conduction current. Although the charges carried are unlikely to be sufficiently large to initiate bulk discharge processes such as lightning, the distribution of charges expected on aerosols under natural ion asymmetry may yield a small fraction of particles with significant charge levels. Such highly-charged aerosol would normally be rapidly neutralised by atmospheric ionisation, but in ion-depleted regions arising from large aerosol concentrations, moderately-high charge levels might persist.

Scavenging processes are influenced by aerosol charge; the water drop charge has negligible effect by comparison. Since heterogeneous ice nucleation requires the collection of suitable aerosol able to operate as ice nuclei, it is therefore conceivable that aerosol charging could influence ice formation. If charge were itself shown to be an enhancing effect for ice nuclei, then the synergy between the effects of increased collection and efficient nuclei could be potent.

## 7.2 Ultrafine particle production

Radiolytic particle production has been observed in laboratory air at atmospheric levels of ionisation, but the particles formed are small and not, at their formation, able to act as cloud condensation nuclei. The recent theoretical work of Yu and Turco (2001) is, however, compelling, in that it shows that in aerosol-deficient regions, such as marine stratus cloud, cosmic ray ionisation could provide an appreciable source of particles. Further microphysical modelling is required to show that sufficient ultrafine particles can survive to become cloud condensation nuclei before the effect on suitable clouds can be assessed.

## 8. CONCLUSIONS

The physical processes, if any, leading to the cosmic ray-low cloud correlation observed by Marsh and Svensmark (2000) remain to be established in the atmosphere. As discussed above, there are atmospheric electrical mechanisms relating ionisation to cloud which remain relatively unexplored in atmospheric physics, and in suitable cloud, could conceivably offer physical explanations for the observed correlation. However without numerical and theoretical estimates of their significance, it is currently impossible to regard ionisation effects as irrelevant to cloud processes.

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