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Charging Mechanism of Lightning at the Molecular Level

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Abstract

Cloud electrification is one of the oldest unresolved puzzles in the atmospheric sciences. Though many mechanisms for charge separation in clouds have been proposed, a quantitative understanding of their respective contribution in a given meteorological situation is lacking. Here we suggest and analyze a hitherto little discussed process. A qualitative picture at the molecular level of the charge separation mechanism of lightning in a thundercloud is proposed. It is based on two key physical/chemical natural phenomena, namely, internal charge separation of the atmospheric impurities/aerosols inside an atmospheric water cluster/droplet/ice particle and the existence of liquid water layers on rimers (graupels and hailstones) forming a layer of dipoles with H^+ pointing out from the air-water interface. Charge separation is achieved through strong collisions among ice particles and water droplets with the rimers in the turbulence of the thundercloud. This work would have significant contribution to cloud electrification and lightning formation.

Keywords

Cloud Electrification, Charge Separation, Collision, Molecular Level

1. Introduction

The charging mechanism of lightning is an age-old hitherto unresolved problem.

More than a hundred years ago, Wilson [1] measured electric field changes at the ground caused by intra-cloud lightning and found that the momentary electric currents constituting the discharges propagated upwards in nearly all cases. He proposed that the falling rain drops of the thunder cloud were positively charged, leaving behind them a negative charge in the upper part of the cloud. This is the first picture of the vertical charge dipole, which was confirmed by Krebhiel *et al.* [2] and Stolzenburg *et al.* [3] with extensive electric field change measurements and in more complicated charge center distributions, respectively. The generally accepted concept for the development of the thunderstorm charged dipole is the physical separation of oppositely charged particles within the cloud. Many mechanisms have been proposed to account for the observed charges on cloud particles [4]. A century ago drop break-up has been proposed according to the observed electrical effects which are associated with drop break-up near waterfalls: the larger droplets became positively charged while the fragments were negative [5]. Drop break-up may only occur during collisions of two particles, when other, stronger, charging processes may take place leading to an ordered separation of opposite charges in the cloud. Convective mechanism as proposed by Wilson [1] involved the movement of charges carried by the natural convection current in a storm cloud. However, the convective charging hypothesis by itself is unable to produce significant charging [6]. The inductive charging process relies on the pre-existing vertical electric field to induce charges [7]. This process may help account for observations in thunderstorms of regions of cloud particles that have acquired their charges very rapidly in later stages of storm development when substantial electric fields are already present. Observations in thunderstorms have shown that strong electrification follows the development of ice particles [8]. Measurements of the charge transfer during collisions of vapor-grown ice crystals and a riming (graupel) target were reported by many groups, such as, Takahashi [9], Saunders *et al.* [10], Caranti *et al.* [11], Jayaratne [12], Brooks *et al.* [13], Pereyra *et al.* [14], Berdeklis and List [15] and etc. Among them the measurement results are different, which are highly dependent on the experimental environment and conditions [16]. A model proposed by Dash *et al.* [17] described charge and mass transfer in ice-ice collisions in terms of fundamental molecular physics. The principal features that the theory captures are as follows: 1) Particles experiencing more rapid vapor growth charge positively. 2) Positive charging is due to the loss of negative charge. 3) Charging is proportional to growth at low rates but increases more slowly with growth at high rates. 4) Charge transfer is enhanced by surface disorder. 5) Liquid like mass transfer accompanies charging, with the colder particle gaining mass. In their model it is supposed that the OH^- ions are bound to sites by their remaining hydrogen bonds, while positive ions diffuse more readily away from the surface into the ice, thereby creating a charged double layer and a negative surface potential. Summaries of the many propositions of thundercloud charging mechanism are given, for example, in [4] [16] [18]-[23].

A thunderstorm is characterized by vigorous convection where several classes of hydrometeors, such as liquid cloud droplets, drizzle and rain droplets as well as frozen droplets, graupel and hail particles are lifted up to heights between 5 and 15 km where the temperature can be anywhere between −5˚C and −40˚C. In this highly turbulent environment, frequent collisions among the various types of hydrometeors occur; this would give rise to charge separation. The general consequence is that the lightweight ice particles are positively charged and float up to the upper altitude while the much heavier graupel and hail particles are negatively charged and stay at lower altitude. Eventually, the charge separation becomes so strong that a discharge (lightning) occurs between the two charged cloud layers or clouds. The question is how charges are separated into positive and negative groups in the above environment.

Scientists have proposed some mechanisms to explain and parameterize charging processes among cloud particles through laboratory experiments and atmospheric observations [9]-[15]. However, most of these studies have focused on macroscopic charging processes; the only fundamental theory at the molecular level proposed is restricted in ice-ice collisions [17]. The detailed microscopic charging processes in the cloud, especially at the molecular level, remain unclear. In this paper, we shall concentrate only on the mechanism of charge separation at the molecular level under the above turbulent condition in the cloud.

2. Overview

Our explanation is based largely on previously published experimental observations by other groups [9]-[15]. Essentially, it is due to impurity charge separation inside a droplet (raindrops for example) and inside an ice crystal (to be called internal charge separation) followed by strong collisions of these particles with much larger graupels and hailstones inside the strong turbulence. The strong collisions would shatter the droplets and ice crystals into positively and negatively charged fragments (to be called external charge separation). The negatively charged fragments would be attracted by the liquid water layer's dipole field on the surface of wet graupels and hailstones and "dissolve" into the water (to be called solvation) while the positively charged fragments would be repelled from the graupel's surface, with some of them eventually ending up in the upper part of the cloud. The consequence is charge separation in the cloud. The following section gives a detailed analysis of the proposed mechanism.

3. Analysis

3.1. Internal Charge Separation

According to atmospheric sciences [see for e.g. in [18] [19] [21] [23] [24] [25], the nucleation and growth of droplets in a cloud is initiated by cloud condensation nuclei (CCN), many of which contain salts with low ionization potentials, such as NaCl and $(NH_4)_2SO_4$ or other similar ions. According to the pioneering experimental results of Gebhardt, Schroeder and Kompa (GSK) [26], once the cluster/droplet is formed, the impurity molecules such as NaCl, $(NH_4)_2SO_4$ etc.

will dissolve in the liquid and charges are separated. The salts dissolve because it is energetically more favorable to separate them into opposing charges by the dipole electric field of the water (polar) molecules forming solvate shells or solvent cages around them. This is solvation. For example, NaCl would become Na^+ and Cl^- each enveloped by a solvent cage. Figure 1(a) shows the solvation of a negative charge e schematically. The solvation of a positive charge is similar except that the water dipoles surrounding the positive charge would reverse directions. The above is only possible for atoms/molecules with low ionization energy (This is true for most, if not all, CCN inside cloud droplets). The solvated positive and negative charges will redistribute themselves inside the liquid reaching a new equilibrium. We call this internal charge separation. These droplets inside the cloud will grow quickly in a humid environment through droplet-droplet collision and coalescence, etc. Since inside each droplet, there is already an internal charge separation of the CCN, the fusion of two droplets would mean that there are more charges distributed inside the larger droplet. These droplets in the cold environment of the thunderstorm would become super-cooled.

Similarly, freezing of water becoming ice in the cloud is usually initiated by ice nuclei (IN) [23] [24], the most common being clay minerals such as illite e.g. Feldspar ($KAlSiO_4$) etc. which contain low ionization atomic and molecular impurities similar to the case of CCN. Inside an ice embryo, the impurities in the IN would be separated into atomic and molecular ions by the dipole field of the water molecules. Through interaction with the dipole fields of the surrounding water molecules, these ions would migrate within the ice structure and be trapped at different sites of the ice body so as to reach an equilibrium state (Figure 1(b)). The ice embryos would grow rapidly in the supercooled/supersaturated environment through collision with supercooled droplets. Small ice would be formed. Within a cloud of supercooled droplets inside a thundercloud, they would grow rapidly through collision until the latent heat of freezing has heated the whole droplet to 0˚C. The resultant larger size ice particles would grow further through

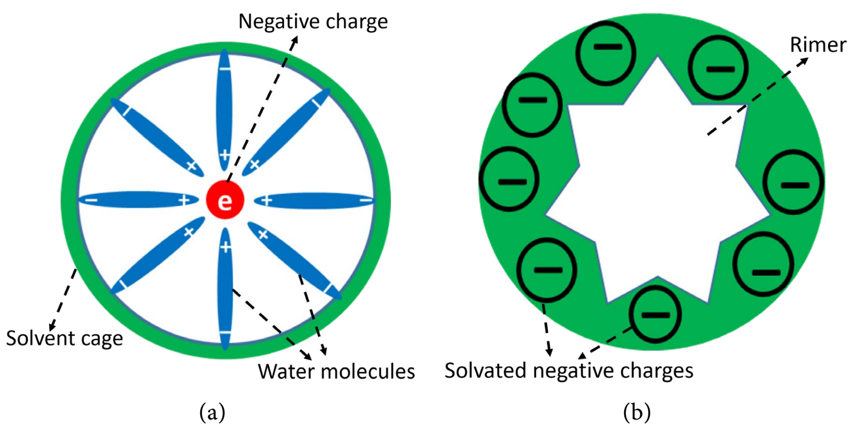

Figure 1. (a) A primitive picture of a solvated negative charge, *e*, in a solvent cage. The cigar-shaped dipoles are water molecules. Hydrogen bonds are not indicated. After solvation, the ensemble becomes a negatively charged "ball". (b) Solvated negative charges (circles) in a surface layer of water on a rimer represented by a white star (Not to scale).

riming resulting in graupels and hailstones. We shall call them collectively as rimers in this paper. Because most of the content in an ice crystal and in a rimer come from the freezing of supercooled droplets containing CCN, the impurities inside an ice crystal and a rimer are mostly CCN.

3.2. External Charge Separation

This process is essentially the result of collisions among the various particles inside the thundercloud. According to the experimental results of GSK [26], when an impurity doped water cluster/droplet collides with any solid surface at a sufficiently high kinetic energy, it will be shattered into many fragments some of which are negatively charged and some positively while others are neutral. The so-called sufficiently high kinetic energy of the particle before collision is to overcome the electrostatic binding energy between the oppositely charged parts inside the cluster/droplet before fragmentation (see also in [27] for similar results). In a thundercloud, the collision would be among the droplets/raindrops/ice crystals and the much larger size solid graupels and hailstones. We should emphasize that inside a thundercloud, there is a turbulence with strong wind blowing around. The wind speed is very high, up to the order of a few tens of meters per second in the extreme. This high wind speed would be the basis of strong collisions among the particles. The following gives a physical/chemical picture of the mechanism of such charge separation due to strong collisions. We shall analyze the collisions among the majority particles inside a thunderstorm, namely, supercooled water droplets, raindrops, ice particles and rimers (graupels and hails).

The collisions among supercooled water droplets would result in either coalescence or the formation of two or more prodigy drops. The collisions among supercooled water droplets and rimers would serve as a wetting agent; *i.e.* the surface of a dry solid rimer would be kept wet with a layer of liquid water soon after the collision with super-cooled droplet. This is because during the freezing of a supercooled water droplet on the surface of a rimer, latent heat is released thus warming up a thin surface layer which becomes liquid for a short period of time. This layer of liquid water would soon freeze up as soon as it cools below 0˚C. Because of the existence of a large number of supercooled droplets and the high wind speed in the turbulence, such collisions would be very frequent so that the surfaces of a large number rimers would be kept wet almost constantly. We shall call them wet rimers (graupels and hails).

Collisions among ice crystals and wet rimers would be the major process giving rise to charge separation. While the fragmentation following ice-ice collisions has not been studied extensively [24], we propose the following scenario. When an ice crystal collides with a wet rimer at a high speed, it will be shattered into positively and negatively charged as well as neutral ice fragments. Meanwhile, on the surface of the wet rimer, there is a layer of positively charged hydrogen atoms sticking out of the air-water interface [28], resulting in a layer of dipoles pointing outward from the liquid surface (**Figure 2**). The electric field

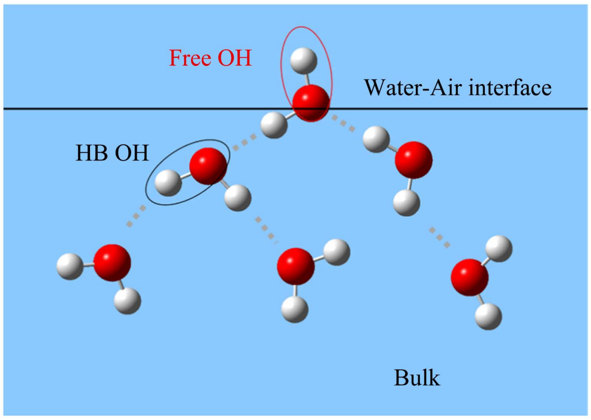

Figure 2. Water molecules at the liquid water-air interface. Red balls, oxygen atoms; grey balls, hydrogen atoms. HB OH: hydrogen bonded OH; dotted lines: hydrogen bonds (idea adopted from [28]).

(in [23]: p. 167) from the layer of dipoles would attract the negatively charged ice fragments into the water layer (See more detailed discussion in Section 3.3). The negatively charged fragments would "dissolve" in the liquid layer which would then freeze up quickly. Thus, the negatively charged impurities in the fragment would become trapped inside the frozen layer. Such trapping and freezing would prevent any charge transfer to other particles during subsequent collisions. The positively charged ice fragments, being very light, would "fly" away into the higher part of the cloud after many collisions with other particles. The neutral fragments would either stick onto the surface of the wet rimer or "fly" away. Consequently, the rimer would become more and more negatively charged through more and more collisions with the ice crystals and would grow in size. Being heavy, the rimers would stay at the lower part of the cloud.

Collisions among supercooled raindrops and wet rimers would mostly involve large rimers (hails). Since both species are not as abundant as those smaller particles in a thundercloud, it would be a minor process as compared to the collision between ice crystals and wet rimers. The collision would result in the "splashing" of the liquid droplet at the surface of the large wet rimer. Much of the central part of the splashing would stick on the rimer's surface. At the periphery, liquid fragments would be "splashing out" radially. The relative kinetic energy between the super-cooled raindrops and the rimer inside the strong turbulence would be sufficiently high such that after the collision, the fragments would become positively and negatively charged as well as being neutral. As in the case of ice crystals' collisions, the negatively charged fragments would be attracted by the electric field from the positively charged water surface of the wet rimer. They would merge into the liquid layer which would freeze up quickly on the main body of the solid rimer. This would prevent any charge transfer to other particles during subsequent collisions. The positively charged super-cooled fragments would "fly" away, quickly freeze up, collide with other particles and eventually float up into the upper part of the cloud. The neutral fragments would probably stick onto the surface of the rimer (riming) after the collision or undergo more collisions with other particles. Consequently, the rimer would become more and more negatively charged through more and more collisions with the raindrops and would grow in size.

The collision among rimers would result in either breakup or scattering but probably no charge transfer.

When more and more charges are separated into the positively charged ice particles in the upper altitude and the negatively charged rimers in the lower altitude, sooner or later, the electric field between the two oppositely charged clouds is sufficiently strong so that corona discharge followed later by lightning flash would occur.

3.3. Positively Charged Hydrogen Atom Sticking out of a Water Surface

It is well-known [28] that at the water-air interface, a positively charged hydrogen atom which is linked to the parent molecule sticks out into the air (**Figure 2**). The other part of the parent molecule is linked to the surrounding hydrogen bonds in the following way.

Below the water-air interface (**Figure 2**), there are two hydrogen bonds, one linking the O atom, one linking the second H atom of the parent molecule. Consequently, more electronic charge cloud will be sucked away from the O atom at the surface. This O atom would thus attract more electronic charge from the H atom sticking out of the interface. Consequently, this H atom would be charged more positively than the H atom of a free water molecule. The positively charged hydrogen atoms above the interface would form a positively charged layer. Since the bulk liquid as a whole is neutral electrically, the consequence is that a layer of dipoles is formed with the positive charges pointing upward at the liquid–air interface. This layer of dipoles would have a surface dipole electric field which will attract negative charges in its vicinity.

For a bath of water at room temperature, there will be thermal jittering of the surface molecules such that the effective surface dipole field would be negligible. However, at a temperature at or below the freezing point, the liquid water surface would become stable presumably at the molecular level; *i.e.* local jittering of the molecules at the surface could be negligible. A stable surface layer of positive charges, hence, a stable layer of dipoles would be formed. This surface layer of dipoles would have a rather strong electric field above the surface. (See next paragraph.) This field would be able to attract the negatively charged fragments of water droplets/ice particles in its vicinity. In the case of a rimer whose surface is wet with a layer of liquid water at 0˚C, negatively charged fragments of droplets/ice particles would be attracted toward (and hence into) the surface's liquid layer.

The following gives a crude estimation of the electric field from a layer of water dipoles sticking out of the air-water interface. Such a dipole layer would in turn induce other dipoles below it and the latter in turn induces other dipoles below, and so on throughout the interior of the liquid structure. A model is thus proposed. It consists of a uniform layer of positive charges on the surface and a similar layer of negative charges with the same charge density at the bottom of the water bath. This is equivalent to having a polarization P distributed uniformly across the layer. This layer is assumed to be pancake in shape [29] [30] with a thickness T and radius R (**Figure 3**).

Assume that all the water molecules (dipoles) at the surface contribute to P. The dipole moment of a free water molecule is p = 1.84 Debye. In the current situation, sticking out of the interface is the dipole OH. Its dipole moment would be smaller than that of the parent molecule H_2O. We thus postulate that the dipole moment at the interface is reduced to 30% of the dipole moment of a free molecule. The density of water $D \cong 3 \times 10^{28}$ molecules/m^3; 1 Debye = 3.34×10^{-30} Coulomb*m. Assume that the thickness $T = 1$ μm, the radius $R = 1$ mm so that its volume is $V = \pi R^2 T$, $P = D_p \cong 0.054$ Coulomb/m^2. The electric field ([29] [30]) is

$$E = \frac{1}{4\pi\epsilon_0}\frac{2PV}{r^3} \cong \frac{3}{r^3} \text{ V/m} \tag{1}$$

where ϵ_0 is the vacuum permittivity. Thus, the field at the distance $r = 1$ cm is $E = 3 \times 10^6$ V/m = 3×10^4 V/cm. This is a respectable field for a layer of water 1 μm thick and 2 mm in diameter.

Supercooled water droplets, in principle, would also have positively charged hydrogen atoms sticking out of their surfaces. However, a droplet in the strong turbulence keeps changing in shape and size; *i.e.* the surface of the droplet keeps changing such that the net electric field from the surface dipoles would become negligible. We shall thus assume that the electric fields from the surfaces of super-cooled water droplets inside the turbulence are negligible. The surface of a rimer is much larger in size as compared to a droplet and is irregular. Thus, the field from the surface layer of water on a rimer would be less than the estimated value. But it would still be significant from the following point of view. The lifetime of the thin liquid water layer is relatively short before it freezes. The influence of the turbulence on the stability of the thin liquid layer would be negligible; *i.e.* the liquid layer on the rimer could be considered as a quasi-stationary layer during the collision with a droplet, raindrop or ice crystal. Hence, the electric

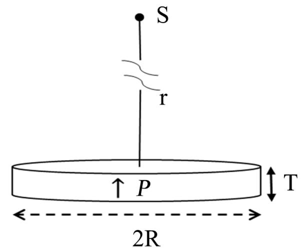

Figure 3. A model for the dipole distribution of water molecules in a slab of liquid water with charges Q and $-Q$ distributed uniformly across the surfaces. It is equivalent to a slab of dipole whose total dipole moment per unit volume is P. Assume that the slab is pancake in shape with thickness T and radius R. We shall estimate the electric field at the position S at a distance r above the surface.

field from the liquid surface of the rimer would be significant. It would attract the negatively charged fragments in the vicinity.

It should be noted that, physically speaking, the dipole field is valid at a distance large compared to the size of the water dipole at the surface while right on the surface, it is zero [29] [30]. During the attraction of a negative charge by the dipole field above the surface, the dipole field would "suddenly" decrease towards zero as the distance decreases. This would mean that the relative acceleration would slow down between the two. Nevertheless, they would still approach each other strongly and finally, the negative charge would "slam" into the liquid surface. Freezing would follow. Once frozen, they would stay within the body of the rimer. The positively charged fragments would be repelled away from the rimer. The neutral fragments would probably stick on the surface of the rimer or undergo more collisions inside the turbulence.

3.4. The Number of Charges Separated from the CCNs in a Thunderstorm

In the proposed charging mechanism, the majority, if not all, of the charges originate from the impurities inside the CCN and IN. They initiate the formation of water droplets, rimers and ice particles. In particular, the impurities inside an ice crystal is mostly CCN. This is because the formation of ice crystals is largely due to the coalescence between ice and supercooled water droplets which contain CCN. One might question if the number of charges from the impurities is sufficient to account for the large number of charges responsible for a lightning discharge. We shall make an estimation and compare it with the measured charges created in a lightning discharge.

We assume that the thunderstorm takes place over land. Before the arrival of the thunderstorm, the typical CCN concentration in air is between 10^2 to 10^3 cm^{-3} based on many measurements (in [23]: p. 168). In the current case, the density of CCN in static air is assumed to be the mean value; *i.e.* 5×10^2 cm^{-3}. (For simplicity, IN is not included or could be neglected as compared to CCN.)

Meanwhile, a strong updraft blows the existing CCN continuously upward into the turbulence zone of the thunderstorm. For simplicity, we idealize that the turbulence zone is a cylinder as shown in **Figure 4**. The size of the cylinder is similar to that of a typical thundercloud; *i.e.* h ~ 15 km, diameter of the cross section A is ~20 km. The updraft blows the CCN uniformly into the cylinder of turbulence. The turbulence zone is assumed to be a sink for the CCN being carried up by the updraft (**Figure 4**). Inside the turbulence zone, cloud formation and charge separation, etc. take place. Since the maximum speed of an updraft can be up to the order of a few tens of m/sec, we conservatively assume that the mean speed v of the updraft is a few m/sec; say, $v = 5$ m/sec. This updraft carries a fixed density D of CCN's into the turbulence zone. We further assume that this density is uniformly distributed in the updraft across the area of the cylinder (thundercloud), its value being the static value of 5×10^2 cm^{-3}; *i.e.* $D = 5 \times 10^2$ cm^{-3}.

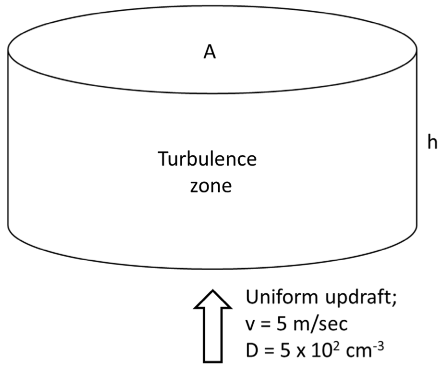

Figure 4. A schematic diagram of a hypothetical thunderstorm geometry.

The flux of CCN moving up with the updraft is thus

$$F = D_v \left[\mathrm{cm}^{-2} \cdot \mathrm{sec}^{-1}\right] \tag{2}$$

This flux is assumed uniform across the cross section A of the cylinder. The total number of CCN carried into the turbulence zone per second is

$$n_{\mathrm{CCN}} = D_v A \left[\mathrm{sec}^{-1}\right] \tag{3}$$

The volume of the turbulence zone is Ah. Thus, the number of CCN flowing into the turbulence zone per unit volume per unit time is

$$N_{\mathrm{CCN}} = n_{\mathrm{CCN}} / Ah = D_v / h \left[\mathrm{cm}^{-3} \cdot \mathrm{sec}^{-1}\right] \tag{4}$$

This number is re-distributed inside the turbulence zone. For simplicity, we assume that the impurity responsible for charge separation inside each CCN is NaCl without loss of generality. Assume that each CCN contains N_{NaCl} molecules. The total number of NaCl molecules per unit volume per unit time is thus given by

$$N_{\mathrm{NaCl}} = N_{\mathrm{CCN}} N_{\mathrm{NaCl}} = \left(D_v / h\right) N_{\mathrm{NaCl}} \left[\mathrm{cm}^{-3} \cdot \mathrm{sec}^{-1}\right] \tag{5}$$

Assume that each NaCl contribute to one positive and one negative charge e, the absolute value of the electronic charge. Hence, the total number of negative/positive charges N_e in one CCN is

$$N_e = N_{\mathrm{NaCl}} \tag{6}$$

And the total number of negative/positive charges per cm^3 per sec is

$$N_e = N_{\mathrm{NaCl}} = \left(D_v / h\right) N_e \left[\mathrm{cm}^{-3} \cdot \mathrm{sec}^{-1}\right] \tag{7}$$

Let Q = quantity of charges/cm^3/sec = $N_e e$ [cm^{-3}·sec^{-1}]; $e = 1.6 \times 10^{-19}$ Coul.

$$Q = \left(D_v / h\right) N_e e \left[\mathrm{Coul} \cdot \mathrm{cm}^{-3} \cdot \mathrm{sec}^{-1}\right] \tag{8}$$

$D = 5 \times 10^2$ cm^{-3}; $v = 5$ m·sec^{-1}; $h = 15$ km; N_e = not yet defined.

We now need to give an estimation of the quantity N_e which is equal to the number of NaCl molecules N_{NaCl} inside a typical CCN. We note that particles larger than the Aitken particles (particles with radius less than 0.1 μm) are usually better CCN (in [23]: p. 171) if their chemical compositions are the same.

We thus assume that most of the CCN are particles larger than the Aitken particles and that the volume of NaCl inside each CCN particle is equivalent to a cube of salt whose volume is (0.01 μm)^3. This is more than two orders of magnitude smaller in volume than the overall size of the CCN, probably a pessimistic estimation.

$$N_e = N_{\mathrm{NaCl}} = d\left(0.01\,\mu\mathrm{m}\right)^3 \Big/ \left[\mathrm{NaCl}\right]_{\mathrm{mass}} \tag{9}$$

where d = 2.17 g·cm^{-3} is the density of salt, $[\mathrm{NaCl}]_{\mathrm{mass}}$ = atomic mass of NaCl = 58.44 amu; 1 amu = 1.66 × 10^{-24} g.

Thus,

$$N_e = 2\times10^4\ \mathrm{cm}^{-3} \tag{10}$$

Using Equation (8), we obtain

$$Q = 5.3\times10^{-16}\ \mathrm{Coul}\cdot\mathrm{cm}^{-3}\cdot\mathrm{sec}^{-1} = 31.8\ \mathrm{Coul}\cdot\mathrm{km}^{-3}\cdot\mathrm{min}^{-1} \tag{11}$$

In a typical thunderstorm, the rate of charge generation is about 1 Coul. km^{-3}·min^{-1} [25]. Q in Equation (11) is thus more than one order of magnitude larger than the rate of natural charge generation. This is reasonable because during the fragmentation of ice particles and raindrops, not all the internal charges are separated into fragments. In other words, even with a probably pessimistic estimation, there are enough impurities to charge up the thundercloud. This would justify the proposal that most of the charges in a thundercloud come from the impurities inside the CCN (and to a minor extent, IN).

There are occasional cases in which the bottom part of the thundercloud is charged positively. This phenomenon might be due to the downdraft from the upper part of the thundercloud around the cloud's periphery into the base of the thundercloud. Positively charged ice crystals at the upper part of the thundercloud would be carried down resulting in a positively charged bottom region. One might ask if it is possible for positively charged fragments to descend directly along the central column of the cloud and form the positively charged zone at the bottom of the cloud. The answer is negative. That is because positively charged ice crystals are light-weight particles. In the turbulence with high buoyancy, they would "float" upward along the central column of the updraft. Since the downdraft would blow mostly around the periphery of the central turbulent zone, some positively charged ice crystals would descend around the periphery.

4. Discussion

Our analysis shows that a few major conditions should be satisfied during charging in a thundercloud. First of all, both the CCN and IN contain atomic and molecular impurities with low ionization potentials. Internal charge separation of the impurities inside the supercooled water droplets and raindrops as well as in small ice particles would naturally take place. The second condition is the existence of supercooled water droplets. It was discovered that supercooled water droplets play a very important role by analyzing and modelling thunderstorms over various cities in China even though the mechanism was not yet clear [31] [32] [33]. These droplets, on collision with small ice particles will readily freeze forming ice crystals. Further collisions between the ice crystals and supercooled water droplets would increase the size of the ice crystals. The latter eventually become rimers. Supercooled water droplets would also contribute to wetting the surfaces of the rimers by the release of latent heat during the freezing of the super-cooled liquid water droplets on the rimers. Positively charged hydrogen of the water molecule would stick out of the wet air-liquid interface (hence, dipoles) and would attract the negatively charged fragments into the liquid layer on the surface of the rimer. This would trap the charges inside the water environment and they would readily freeze up. The third is related to the findings of Takahashi who pointed out in his experiment [34] that simultaneous occurrence of supercooled water droplets and ice crystals is a necessary condition for charge separation. The supercooled water droplets would serve as a wetting agent on the surface of a rimer (as explained in the second condition) while raindrops (including large size supercooled droplets) would be shattered into positively and negatively charged fragment on collision. The ice crystals would become rimers and contribute to charge separation via collisions with wet rimers. The fourth condition is strong updraft and turbulence. The strong wind in the turbulence would provide a high kinetic energy onto the various particles undergoing collisions. This would allow positively and negatively charged fragments to be separated during the collision of the raindrops (including large size supercooled droplets) and ice crystals with the wet rimers. The strong wind in the turbulence would also increase the frequency of collisions between rimers and super-cooled droplets so that the surface of the rimers would almost always be kept wet. The fifth condition is the abundant supply of water molecules. This is satisfied by the abundant moisture in the updraft. The strong moisture (high cloud water content) is to provide enough water molecules to feed the growth of droplets, rimers and ice particles.

In a laboratory experiment, when the voltage difference between two electrodes in air is increased from zero up to a certain high voltage, corona discharge between the two electrodes will occur [29] resulting in a faint bluish glow between the electrodes. This will give rise to ionic winds blowing outward from the electrodes [35]. Note that the wind speed during a corona discharge was measured in the laboratory to be up to a few m/sec [35], while the wind speed in an updraft of a thunderstorm is also a few m/sec according to [25]. When the voltage is further increased, the ionic wind becomes stronger until a threshold electric field is reached when breakdown "suddenly" occurs between the two electrodes giving rise to a strong flash of light together with a strong snapping sound. The ionic wind will "abruptly" stop. The charging of a thundercloud is similar; it would start from zero. As charging increased, the voltage difference (or electric field) between two cloud bodies would become larger and larger. The high voltage difference would sooner or later induce a corona discharge. This would give rise to an ionic wind between/inside the clouds enhancing the wind speed inside the turbulence. This would in turn increase the number and speed of collisions among the particles inside the cloud [35]. In particular, as is popularly believed, the collision between ice particles and the much heavier rimers (graupels and hailstones) would result in charge separation in the cloud. Thus, the higher the speed of collision, the more charge separation there would be and the corona discharge would become stronger, resulting in a stronger ionic wind. The latter would in turn enhance the wind speed inside the turbulence and so on. These cyclic processes of enhancing the wind speed [35] and the charging of the cloud would eventually reach a threshold electric field between the two oppositely charged cloud bodies. This would result in a breakdown between the two charged clouds; *i.e.* lightning together with a "thunderous sound". A rain gush would follow [35]. On the other hand, there could be a situation in which corona discharge could not grow further because of various reasons such as insufficiency of water content in the cloud, or the strength of the turbulence was not sufficient to further increase collisions and charge separation, etc. In this case, the corona discharge produces only faint glows in the cloud without breakdown (no thunderous sound).

5. Conclusion

We offer a qualitative picture at the molecular level of the charge separation mechanism of lightning in a thundercloud. It involves two key physical/chemical "natural" phenomena, namely, internal charge separation of the atmospheric impurities/aerosols inside an atmospheric water cluster/droplet/ice particle and the existence of a liquid water layer on a rimer forming a layer of dipoles with H+ pointing out from the air-water interface. Strong collision (provided by the strong turbulence) is necessary between the super-cooled water droplets and raindrops/ice particles and the rimers. This would separate some of the positively and negatively charged fragments from inside the raindrops and ice particles. The dipole field from the liquid layer of water on the surface of the rimer would attract the negatively charged fragments into the water layer. The negatively charged fragments would then "merge" into the liquid layer followed by "immediate" freezing and would not be "extracted" out during subsequent collisions. The consequence is that the much heavier rimers would be charged negatively and stay at the lower part of the thundercloud. The positively charged fragments would undergo more collisions in the turbulence and would eventually fly (float) up into the upper part of the thundercloud. Finally, after sufficient charge separation, electrical discharge would occur between the upper and lower part of the thundercloud; *i.e.* lightning.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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