

# *Stratiform cloud electrification: comparison of theory with multiple in-cloud measurements*

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1                   **Stratiform cloud electrification: comparison of theory with multiple in-cloud measurements**

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4

5                   **Abstract**

6                   Stratiform clouds constitute ~40% of global cloud cover and play a key role in determining the planetary  
7                   radiation budget. Electrification remains one of the least understood effects on their microphysical  
8                   processes. Droplet charging at the top and bottom edges of stratiform clouds arises from vertical current  
9                   flow through clouds driven by the Global atmospheric Electric Circuit. In-cloud charge data are central in  
10                   assessing the role of charge in droplet growth processes, which influence droplet size distributions and  
11                   associated cloud radiative properties and precipitation. This study presents the first high vertical  
12                   resolution electrical measurements made in multiple layer clouds. Of the 22 clouds sampled, all were  
13                   charged at their edges, demonstrating unequivocally that all stratiform clouds can be expected to contain  
14                   charge at their upper and lower boundaries to varying extent. Cloud base and cloud top are shown to  
15                   charge asymmetrically, with mean cloud top space charge  $+32 \text{ pCm}^{-3}$  and base space charge  $-24 \text{ pCm}^{-3}$ .  
16                   The larger cloud top charges are associated with strong temperature inversions and large vertical  
17                   electrical conductivity gradients at the upper cloud boundary. Greater charging was observed in low  
18                   altitude ( $<2\text{km}$ ) clouds ( $20.2 \text{ pCm}^{-3}$ ), compared to higher altitude ( $>2\text{km}$ ) cloud layers ( $7.0 \text{ pCm}^{-3}$ ),  
19                   consistent with the smaller air conductivity at lower altitudes caused by reduced cosmic ray ionisation.  
20                   Taken together, these measurements show that the greatest cloud droplet charges in extensive stratiform  
21                   clouds occur at cloud tops for low altitude ( $<2\text{km}$ ) clouds, when vertical mixing is suppressed by  
22                   appreciable temperature inversions, confirming theoretical expectations. The influence of cloud  
23                   dynamics on layer cloud edge charging reported here should inform modelling studies of cloud droplet  
24                   charging effects on cloud microphysics.

25

26

27

28        **1. Introduction**

29        Earth's atmosphere is a weak conductor of electricity due to the presence of atmospheric cluster ions,  
30        formed, near the surface, from natural radioactivity in rock and soil, and at higher altitudes by Galactic  
31        Cosmic Rays (GCRs). GCRs are the primary source of ionisation above the Earth's surface, creating a  
32        cascade of charged energetic particles by collision with O<sub>2</sub> and N<sub>2</sub> molecules as they enter the atmosphere  
33        from above. The presence of cluster ions, and the existence of a Global atmospheric Electric Circuit (GEC)  
34        permit a small current density, J<sub>c</sub>, ( $\sim 10^{-12}$  A m<sup>-2</sup>) to flow vertically from the lower ionosphere (approx.  
35        60km altitude) to Earth's surface (e.g. Wilson, 1929; Israel, 1971). This provides a link between the upper  
36        atmosphere and the lower troposphere, in all fair weather regions of the globe. Conventional atmospheric  
37        electricity (e.g. Israel, 1971) distinguishes between disturbed weather (i.e. thunderstorm conditions) and  
38        fair weather regions (where there is no local charge generation), but there also exists a very common, but  
39        seldom mentioned, situation of semi-fair weather conditions – defined here as conditions pertaining to  
40        extensive, non-precipitating layer clouds. These are different from electrified shower clouds which, like  
41        thunderstorms, contribute significantly to the GEC current flow (e.g. Mach et al, 2011; Blakeslee et al,  
42        2014) and are typically associated with precipitation. Layer clouds are prevalent, covering almost 40% of  
43        the planet's surface at any one time (Klein and Hartmann 1993), and play a large role in the terrestrial  
44        radiation balance. When such extensive clouds are present, the fair weather current must pass through  
45        the cloud (experimentally verified by Bennett and Harrison, 2009 and Nicoll and Harrison, 2009; modelled  
46        by Baumgaertner et al, 2014). The consequence of current flow through cloud layers is for charge  
47        accumulation to occur at the top and bottom horizontal cloud edges (Tinsley 2000, Zhou and Tinsley  
48        2007), due to the conductivity transition between the clear air and cloudy regions (see e.g. Harrison et al  
49        2015, figure 1). We define the horizontal cloud edge here as the transition that occurs between cloud  
50        and cloud-free air at the cloud base or top (not the sides of the cloud).

51

52        Charging of cloud droplets has implications for their behaviour and can affect cloud microphysical  
53        processes such as droplet-droplet interactions (e.g. Khain et al, 2004), aerosol-droplet interactions (e.g.

54 Tinsley et al, 2000) and droplet activation (e.g. Harrison and Ambaum, 2008). Harrison et al 2015 discuss  
55 these mechanisms in detail. The magnitude of cloud droplet charge required to affect each of these cloud  
56 microphysical processes varies between a few electronic charges to a few thousand, depending on the  
57 process and the droplet size. The likely large scale effect of charge on the cloud droplet population is a  
58 change in size or number concentration of the droplets, which can lead to a change in the radiative  
59 properties of the cloud, and potentially precipitation changes. This is supported by recent observations  
60 of changes in cloud base properties associated with well-known atmospheric electrical variations  
61 (Harrison and Ambaum, 2013; Harrison et al 2013). As stratus clouds are common globally (being  
62 particularly prevalent over oceanic regions), (Klein and Hartmann, 1993) and the vertical current is always  
63 present in semi-fair weather regions, electrical microphysical effects may provide an underlying source of  
64 variability in cloud properties.

65

66 Characterisation of the typical droplet charge in layer clouds is a key factor in understanding cloud edge  
67 electrical effects, and is at present not well known. Previous measurements of cloud droplet charging in  
68 non-thunderstorm cloud have mostly been made from mountaintop observatories in the 1950s (e.g.  
69 Twomey, 1956 Phillips and Kinzer, 1957; Allee and Phillips, 1959), where droplet charges of typically  
70  $\pm 20$  elementary charges ( $e$ ) were found (Phillips and Kinzer, 1958). There remain, however, concerns that  
71 the electrical conditions at mountaintops are not representative of meteorological conditions in clouds in  
72 the free atmosphere. Aircraft measurements of droplet charging were made by Beard et al (2004) within  
73 altostratus, although charge measurements were only reported for the middle of the cloud rather than  
74 on the upper and lower edges, where the charge accumulates. In the UK, balloon borne measurements  
75 by Jones (1957) and Jones et al (1959) demonstrated appreciable gradients in electric field and  
76 conductivity at horizontal cloud edges inside a stratiform cloud layer, whilst half a century later Nicoll and  
77 Harrison (2010) reported high vertical resolution charge measurements from a similar free balloon  
78 platform through a layer of stratocumulus cloud. A region of negative charge (up to  $-35\text{pC m}^{-3}$ ) was found  
79 at cloud base, with a magnitude similar to that predicted by theory. These sparse measurements  
80 confirmed the existence of charging at upper and lower edges of layer clouds, however further

81 quantitative study of the location, polarity and magnitude of charge carried by cloud droplets in layer  
82 clouds is required to inform modelling studies of charge influences on cloud microphysical processes.

83

84 This paper presents a unique new dataset of observations of layer cloud charging at horizontal edges  
85 using specially designed charge and cloud instrumentation flown from free balloon platforms.  
86 Quantitative analysis of the electrical characteristics of a large number of stratiform cloud layers is  
87 reported, providing a new dataset with which to better understand the role of electrification in layer  
88 clouds. In section 2 the theory behind accumulation of charge on cloud edges is discussed, whilst in  
89 section 3 the instrumentation is described. Section 4 presents new measurements of edge charging for  
90 multiple clouds, and section 5 discusses the various factors controlling cloud edge charging in terms of  
91 the measurements obtained. Section 6 discusses estimates of individual cloud droplet charges, whilst  
92 section 7 includes a discussion and section 8 presents the conclusions.

93

## 94 **2. Separation of charge at edges of extensive layer clouds**

95 Charge accumulation is associated with the upper and lower horizontal edges of layer cloud due to the  
96 transition in electrical conductivity,  $\sigma_t$ , between clear air and droplet laden air. Droplet laden air has a  
97 lower conductivity (i.e. higher resistivity) than clear air due to the attachment of cluster ions (typically nm  
98 diameter) to the cloud droplets (typically  $\mu\text{m}$  diameter). The horizontal cloud edge boundary region  
99 therefore has an associated vertical gradient in conductivity, with, in turn, a vertical gradient in the  
100 electric field,  $E$ . From Gauss' law of electrostatics, the change in the vertical component of the electric  
101 field  $E_z$  gives rise to a region of space charge of density,  $\rho$ , (defined as the net difference between positive  
102 and negative charge per unit volume):

$$103 \quad \frac{dE_z}{dz} = -\frac{\rho}{\epsilon_0} \quad 1,$$

104 where  $\epsilon_0$  is the permittivity of free space and  $z$  is positive upwards. Assuming Ohm's law in the vertical  
105 direction, and expressing  $E_z$  in terms of conductivity  $\sigma_t$ , (where the subscript  $t$  denotes the total

106 conductivity, i.e. the contribution from both positive and negative conductivities) and the atmospheric  
107 potential gradient, PG, where  $PG = -E_z$  and

108 
$$J_c = \sigma_t PG \quad 2,$$

109 it follows that equation 1 can be re-written to give  $\rho$  in terms of the fair weather conduction current  
110 density,  $J_c$ , as

111 
$$\rho = -\varepsilon_0 J_c \frac{d}{dz} \left( \frac{1}{\sigma_t} \right) = -\varepsilon_0 J_c \frac{1}{\sigma_t^2} \left( \frac{d\sigma_t}{dz} \right) \quad 3.$$

112 The electrical changes which occur across a layer cloud are illustrated conceptually in Figure 1, for an  
113 assumed conductivity gradient at the upper and lower horizontal cloud boundaries. If the gradient is made  
114 sharper, a larger amount of space charge but over a narrower layer results. As will become apparent,  
115 this example is very much that of an idealised cloud layer.

116

117 The polarity of the charge layers generated by the vertical conductivity change is, accordingly, positive at  
118 the top and negative at the base of the cloud, due to the difference in the direction of the vertical  
119 conductivity gradients between the upper and lower cloud edges and the downward direction of the  
120 current flow. From the instant that cloud droplets start to form, a vertical gradient in conductivity will  
121 become established and charge will accumulate in these regions. The horizontal charge layers created at  
122 upper and lower horizontal cloud boundaries are maintained by the current flow through the cloud, which  
123 ensures a continuous supply of ions into the top and bottom of the cloud.

124

125 It is clear from equation 3 that cloud edge charging depends on the vertical conductivity gradient which  
126 will be determined by local meteorological conditions, as well as the fair weather current density,  $J_c$ . It  
127 follows that modulation of  $J_c$ , either from external sources such as GCR flux changes due to the solar  
128 magnetic field, or internal sources such as a change in thunderstorm generators in the GEC, will also  
129 modulate the space charge (e.g. Nicoll, 2014, Mach et al, 2011). This is discussed in more detail in section  
130 7. A further consideration, as pointed out by Harrison et al (2015) is the variations with height which

131 occur in the cosmic ray ion production rate and therefore in  $\sigma_t$ . The associated expectation is that the  
132 cloud edge charging will vary with the height dependence in  $\sigma_t$ , assuming similar vertical profiles in the  
133 cloud droplet properties at different heights.

134

135 **3. Methodology**

136 **3.1 Cloud and charge instrumentation**

137 To estimate the typical charges on cloud droplets, knowledge of both the in situ cloud properties and  
138 charge profiles are required. A meteorological radiosonde provides a suitable platform with a typical  
139 vertical sampling resolution of  $\sim 5\text{m}$  from its ascent rate of  $\sim 5\text{ms}^{-1}$  and sampling rate of 1Hz. The standard  
140 radiosonde only measures pressure, temperature, relative humidity (PTU) and position, however a  
141 specially developed data acquisition system known as PANDORA (Programmable ANd Digital Operational  
142 Radiosonde Accessory) (Harrison et al, 2012), allows the attachment of additional science sensors, and  
143 transmission of the data synchronously with the PTU data. Adopting this approach, an optical cloud  
144 droplet sensor has been developed to provide information on the cloud droplet profile and determine  
145 the thickness of the cloud to clear air transition (which is generally not well studied, either at horizontal  
146 cloud edges or vertical ones, as the so called “twilight zone” demonstrates (Koren et al, 2007)). The sensor  
147 employs a backscatter method using an ultra-bright Light Emitting Diode (LED) at 590nm as the source,  
148 with a photodiode receiver (Harrison and Nicoll, 2014). The cloud droplet sensor returns a measurement  
149 of visible range,  $X_r$ , which shows considerable change at the cloud edge boundary (which occurs in the  
150 same region as the conductivity changes). The photodiode also provides a direct measurement of solar  
151 radiation, which provides a further independent determination of the cloud edge boundary region in  
152 daylight (Nicoll and Harrison, 2012). In addition, a small charge sensor is flown alongside the optical cloud  
153 droplet sensor to measure the net space charge density within the cloud layer. This employs a spherical  
154 electrode connected to a sensitive electrometer. It primarily responds to induced displacement currents  
155 generated by electric field changes as the sensor moves through the cloud layer (Nicoll, 2013). Details of  
156 the space charge derivation from the measurement of charge sensor current are given in the appendix.

157 The combined package of the PANDORA, optical cloud droplet sensor and charge sensor has a mass of  
158 250g and is attached to the side of a standard Vaisala RS92 radiosonde, all flown under a 200g helium  
159 filled carrier balloon.

160

161 A useful comparison can be made between the space charge profile measured by the charge sensor and  
162 that calculated from theory. This can be achieved from the data measured by the cloud droplet sensor,  
163 which can be used to estimate the vertical in cloud conductivity profile, and equation 3. To estimate the  
164 conductivity profile one must first consider the droplet concentration profile through the cloud layer. The  
165 cloud droplet sensor was laboratory calibrated to provide optical extinction,  $\xi$ , by measuring the  
166 transmission of LED light through a region of droplet laden air simultaneous with the optical backscatter  
167 (Harrison and Nicoll, 2014). The optical extinction is related to the visual range,  $X_r$ , by

168 
$$X_r = \frac{\ln \varepsilon}{\xi} \quad 4,$$

169 where  $\varepsilon$  is normally taken to be 0.05 (HMSO, 1982) (see e.g. Harrison, 2012). Assuming an optical  
170 extinction coefficient for cloud droplets of approximately twice their projected area (e.g. Bohren and  
171 Huffman 1983; Harrison 2012), the total extinction coefficient  $\xi$  is related to the droplet diameter,  $d$ , and  
172 droplet number concentration,  $Z_d$  by

173 
$$Z_d = \frac{2\xi}{\pi d^2} \quad 5.$$

174 Thus, by assuming an average cloud droplet diameter (taken here to be 10 $\mu\text{m}$  – a typical cloud droplet  
175 size for stratocumulus cloud (Miles et al 2000)),  $Z_d$  can be estimated. The cloud droplet number  
176 concentration is a key parameter in calculating the conductivity profile through the cloud layer. Using the  
177 steady state ion-balance equation (see e.g Harrison and Carslaw 2003), which characterises the ion  
178 concentration in the presence of cloud droplets, the total conductivity is

179 
$$\sigma_t = \frac{\mu e}{\alpha} \left( \sqrt{(\beta_a^2 Z_a^2 + 2\beta_a Z_a \beta_d Z_d + \beta_d^2 Z_d^2 + 4\alpha q_i)} - (\beta_a Z_a + \beta_d Z_d) \right) \quad 6,$$

180 where  $\mu$  is the mean ion mobility (taken here as  $1.7 \times 10^{-4} \text{ m}^{-2} \text{ V}^{-1} \text{ s}^{-1}$ ),  $e$ , the elementary charge ( $1.6 \times 10^{-19} \text{ C}$ ),  $\alpha$  the ion-ion recombination coefficient ( $1.6 \times 10^{-12} \text{ m}^{-3} \text{ s}^{-1}$ ),  $Z_a$  the background aerosol number concentration (assumed here to be  $1000 \text{ cm}^{-3}$ ), and  $\beta_a$  and  $\beta_d$  the size-dependent ion-aerosol and ion-droplet attachment coefficients respectively (Gunn, 1954). Equation 6 assumes that the bipolar ion concentrations are equal and represents the cloud droplets by a single (monodisperse) size.  $q_i$  is the ion production rate per unit volume (typically  $2 \text{ ion pairs cm}^{-3} \text{ s}^{-1}$  near the surface) which is described in more detail in the following paragraph. There is likely to be considerable variability in many of the parameters in equation 6 in real cloud environments, particularly in terms of  $Z_a$  and the mean aerosol radius, on which  $\beta_a$  depends, therefore a considerable range of values for  $\sigma_t$  is expected. Application of equations 4 to 6 to the cloud droplet sensor data allows the estimation of  $\sigma_t$  from the optical sensor, which can be applied to equation 3 to derive an estimate of the in-cloud space charge.

191

## 192 **3.2 Ionisation instrumentation**

193 Equation 6 demonstrates that knowledge of the ion production rate,  $q_i$  is required to derive the  
194 conductivity. Using additional instrumentation to detect the vertical profile of ionisation,  $q_i$  was measured  
195 directly on some of the charged cloud flights. This approach uses two Geiger tubes (LND714) with a  
196 compact high tension voltage supply, and an interval timing technique for improved resolution at low  
197 count rates (described by Harrison et al 2013). The electrical conductivity of air depends directly on the  
198 ion production rate, which varies as a function of altitude and latitude, and plays a role in determining  
199 the cloud edge charge through equation 3. Over land, near the surface, ions are generated from natural  
200 radioactivity such as radon gas ( $4$  to  $8 \text{ ion pairs cm}^{-3} \text{ s}^{-1}$  (Hirsikko et al 2007)), which are typically lofted to  
201 altitudes of  $1$ - $2$  km within the atmospheric boundary layer. Galactic Cosmic Rays (GCRs) provide an  
202 additional source of ionisation of around  $2 \text{ ion pairs cm}^{-3} \text{ s}^{-1}$  at the surface, which increases approximately  
203 exponentially with altitude to become the dominant ion source, reaching a maximum (the Regener-  
204 Pfotzer maximum) at  $\sim 20$  km (e.g. Bazilevskaya et al, 2008). A consequence of the increase in ion  
205 production rate means that the conductivity also increases with height from a few  $\text{fS m}^{-1}$  at the surface to

206 a few  $\text{pS m}^{-1}$  at 20 km. From equation 3, the space charge generated at cloud edges is proportional to  
207  $1/\sigma_t^2$ , therefore, since  $\sigma_t$  varies with height,  $\rho$  is also expected to vary. This prediction is tested in section  
208 5 using the measurements of ionisation rate obtained from the balloon Geiger sensor.

209

210 **4. Charged cloud measurements**

211 **4.1 Typical stratiform cloud**

212 To investigate layer cloud charging, a series of specially instrumented balloon flights were carried out  
213 during 2013-2015, primarily from the Reading University Atmospheric Observatory (RUAO), UK ( $51.44^\circ \text{N}$ ,  
214  $0.95^\circ \text{W}$ ), but also from Hyttiala, Finland ( $61.85^\circ \text{N}$ ,  $24.30^\circ \text{E}$ ), and Halley, Antarctica ( $75.35^\circ \text{S}$ ,  $26.66^\circ \text{W}$ ).  
215 Low altitude non frontal stratus and stratocumulus clouds of large horizontal extent (8/8ths cloud cover  
216 from a single site) were targeted since cloud edge charging is expected from theory to be at its greatest  
217 with sharp cloud boundaries which require little vertical mixing, and at low altitudes (<5km) where  $\sigma_t$  has  
218 its smallest values.

219

220 Figure 2 shows data from an instrumented balloon flight through stratocumulus cloud over RUAO on 18<sup>th</sup>  
221 November 2013, which had properties typical of stratiform cloud at Reading. From the thermodynamic  
222 measurements measured by the radiosonde (Figure 2(a)), the temperature inversion at 1.2km indicates  
223 the cloud top, however, combined with the RH determination, the position of the cloud base is much  
224 more ambiguous. In contrast, both the optical cloud droplet (Figure 2(b)) and charge sensor (Figure 2(d))  
225 show the cloud boundary regions very distinctly, with the location of the lower and upper cloud edges  
226 clearly at 0.85 and 1.2km respectively. Inside the cloud the visual range measured by the optical sensor  
227 decreases to 120m (from >1000m outside the cloud), and the solar radiation increases as the sensor  
228 travels upwards through the cloud layer, consistent with the decrease in optical depth. Figure 2 (d)  
229 demonstrates space charge of up to  $\pm 160 \text{pCm}^{-3}$  at both the upper and lower cloud edges, with very little  
230 charge in the middle of the cloud. The fact that the charge is concentrated in narrow layers at these cloud  
231 edges suggests that the mixing processes removing the charge occur at a lesser rate than the charging

232 rate. In addition, the opposite polarity of charging between the upper and lower cloud boundaries  
233 (positive at cloud top, and negative at cloud base), in this case, agrees well with that predicted by the  
234 theory outlined in section 2.

235

236 The conductivity profile calculated from the estimation of  $Z_d$  and equations 4, 5 and 6 is plotted in Figure  
237 2 (c), where it is seen that the presence of the cloud layer decreases the conductivity substantially (from  
238 approx.  $10 \text{ fSm}^{-1}$  to  $2 \text{ fSm}^{-1}$  from outside the cloud to inside), generating the expected vertical gradient in  
239 conductivity on the horizontal cloud edges. Also shown in Figure 2 (c) is the estimated potential gradient  
240 profile through the cloud layer, which is calculated from the derived  $\sigma_t$  profile and equation 2, assuming  
241 that  $J_c$  is constant through the cloud layer (with  $J_c = 2 \text{ pA m}^{-2}$ ). Note the similarity in the mean structure  
242 between the theoretical profiles in Figure 1 and the measured ones in Figure 2. Finally, the predicted  
243 cloud edge space charge, shown in Figure 2 (d) in grey, can be calculated from equation 3, again with  $J_c =$   
244  $2 \text{ pA m}^{-2}$ , and using  $\sigma_t$  calculated from equation 6. Figure 2 (d) also depicts the space charge measured  
245 directly by the cloud sensor (in black), demonstrating good agreement between the locations of the  
246 predicted and measured charge layer, as well as the polarity. In this cloud layer, both the predicted and  
247 observed charge is located at the cloud edge regions, where there is a vertical gradient in cloud droplet  
248 number and size and therefore a conductivity gradient. It should be noted that exact agreement between  
249 the measured and theoretical profiles is not expected as the theory is based on a very simple (and static)  
250 view of clouds, and does not incorporate any vertical mixing which is known to exist in stratiform clouds  
251 (e.g. Shupe et al, 2008). The magnitude of the calculated charge is somewhat smaller than that measured,  
252 but this varies with the assumptions made in equations 3-6, unlike the location of the charge layers, which  
253 depends solely on the location of the vertical conductivity gradient and therefore the vertical gradient in  
254 cloud droplet concentration. This sensitivity can be illustrated by perturbing the assumed values:  
255 increasing the value of  $J_c$  from 2 to 3  $\text{pAm}^{-2}$  and decreasing  $q_i$  from 2 to 1  $\text{cm}^{-3}\text{s}^{-1}$  increases the maximum  
256 predicted space charge from 37 to 111  $\text{pCm}^{-3}$ .

257

258 **4.2 Horizontal distribution of charge within cloud layer**

259 As the vertical current flows throughout all fair weather regions of the atmosphere, cloud edge charging is  
260 expected across the entire horizontal extent of an extensive layer cloud. Preliminary experimental  
261 evidence for this was given in Rycroft et al (2012) and is further supported here. Figure 3 shows data  
262 from an ascent and descent of an instrumented balloon through the same stratiform cloud layer,  
263 measured 105km apart. Figure 3(a) shows the GPS derived location of the balloon during its ascent from  
264 Reading (black) and descent (grey), and (b) an infrared satellite image showing large scale low cloud  
265 coverage of the southern UK at the time of the balloon launch. Figures 3 (c) to (f) show both ascent (black)  
266 and descent (grey) data from the instrumented balloon flight, whereby a cloud layer exists between 0.7  
267 and 1 km (the lower part of the descent data is missing due to loss of the radiosonde signal close to the  
268 ground). This comparison demonstrates that all four profiles of temperature, RH, visual range and charge  
269 are similar between ascent and decent, with a slight lowering of the cloud top on the descent compared  
270 with the ascent. The charge sensor's response became saturated at its maximum value on both the ascent  
271 and descent stages of the flight at the cloud top (red points), suggesting a very large concentration of  
272 positive charge near the upper cloud edge. For this particular flight, saturation of the charge sensor would  
273 have occurred at approximately  $200\text{pCm}^{-3}$  therefore it is likely that the charge at cloud top exceeded this  
274 value. In contrast there is little evidence of a charge layer at the cloud base. The asymmetry in charge  
275 between cloud base and top is likely to be related to the large temperature inversion (of  $10^{\circ}\text{C}$ ), which  
276 leads to a particularly strong gradient in the visual range at cloud top (Figure 3(e)). From equation 3, this  
277 sharp thermodynamic transition from cloudy to clear air will result in a strong gradient in conductivity at  
278 the cloud top, with the expectation of substantial positive space charge accumulations: this is supported  
279 by the charge data shown in Figure 3(f)). The substantial variability in the charge within the main body of  
280 the cloud (up to  $+\text{-} 100\text{ pCm}^{-3}$  which is larger than on most of the flights observed), with a gradual  
281 transition from positive to negative charge as cloud base is approached, may provide an explanation for  
282 the lack of a well defined charge layer at cloud base, despite a clear gradient in visual range at cloud base.  
283 It is possible that downward mixing of the large amount of positive charge at cloud top acts to neutralise  
284 some of the negative charge at the cloud base, thus resulting in the observed slightly negative values

285 towards the base of the cloud layer. Regardless of the explanation for the unusual charging behaviour,  
286 the existence of substantial charge at a similar height in the cloud but widely horizontally separated  
287 therefore confirms that cloud edge charging is a horizontally widespread phenomena.

288

289 **4.3. Multiple cloud layers**

290 An interesting case of cloud edge charging occurs when multiple, vertically-displaced but overlapping  
291 cloud layers exist. Figure 4 shows vertical profiles from an instrumented balloon flight through two  
292 distinct cloud layers between 0.7-1.0km and 1.4-1.6km (visual range was not available on this flight).  
293 Figure 4 (c) demonstrates the presence of bipolar space charge in both layers, each with positive charge  
294 at cloud top and negative charge at cloud base, in agreement with the theory set out in section 2. The  
295 magnitude of the space charge in the upper cloud layer is much smaller than that in the lower cloud layer,  
296 which is likely related to a shallower conductivity gradient between clear and cloudy air in the upper cloud  
297 layer. This is likely to result from 1) differences in the cloud droplet profiles between the two clouds, with  
298 lower liquid water content expected in the upper cloud layer and 2) an increase in the ion production rate  
299 in the upper cloud layer which acts to reduce the conductivity gradient (as modelled by Zhou and Tinsley  
300 2007). The effect of variations in ionisation rate on cloud edge charging is discussed more fully in  
301 section 5. Nevertheless, the presence of space charge in both cloud layers confirms the continuity of  
302 current density through the upper cloud layer to the layer below, allowing the distinct cloud layers directly  
303 above each other to both become charged at their horizontal boundaries.

304

305 **4.4 Summary of all stratiform cloud flights**

306 Section 4.1 demonstrates a typical example of a stratiform layer cloud and the parameters measured by  
307 the specially-instrumented radiosondes. The current section summarises the 22 different stratiform cloud  
308 layers sampled to date in order to quantify the variability between different cloud layers. The properties  
309 of the 22 stratiform cloud layers analysed are summarised in Table 1.

310

311 Table 1 demonstrates that the range of cloud base heights sampled was from from 475 to 4838m, with a  
312 median height of 1367m , with cloud depths (i.e. cloud top height minus cloud base height) ranging from  
313 the shallowest cloud layer of 86m to the deepest of 1700m (median cloud depth 285m). The mean space  
314 charge density in each cloud layer varied between 2 to 43 pC m<sup>-3</sup>, with a median of 16 pC m<sup>-3</sup>, with the  
315 maximum space charge density in each cloud layer ranging from 10 to 253 pC m<sup>-3</sup> (median = 109 pC m<sup>-3</sup>).  
316 These values are calculated from the magnitude of the charge only, as taking the polarity into account  
317 results in values very close to zero due to the presence of opposite polarity charge at cloud base and cloud  
318 top. Figure 5 shows the variability in the space charge inside each of the 22 cloud layers in more detail,  
319 with each cloud layer plotted as an individual boxplot. The magnitude of space charge encountered  
320 during these flights is in agreement with previously reported values of stratiform cloud charge, which is  
321 typically found to be up to 1000 pC m<sup>-3</sup> (e.g. Ilyanitov and Chubarina, 1967; Nicoll and Harrison, 2009;  
322 Nicoll and Harrison, 2010; Nicoll 2013)

323

324 Figure 2 presents measurements from an individual cloud/charge sensor flight through a stratiform cloud,  
325 which demonstrates agreement with theoretical expectations in terms of the location and polarity of the  
326 cloud edge charge layers, however the boxplots in Figure 5 show that considerable variability in charge  
327 can exist between different cloud layers. Figure 6 further demonstrates the variability between different  
328 cloud layers by showing the individual vertical profiles from 16 of the 22 stratiform clouds sampled,  
329 normalised by the measured cloud depth. Only clouds with mean altitude <3km were selected as these  
330 lie within the typical altitude for stratocumulus clouds, and below this altitude the ion production rate  
331 varies less with height than at higher altitudes (see section 5.2). Figure 6(a) shows the in-cloud visibility  
332 as calculated from measurements from the cloud droplet sensor using the method explained in section  
333 3.1, and (b) the space charge density measured by the charge sensor. There is obvious variability in both  
334 the shape of the vertical profiles and magnitude of the space charge between individual cloud layers. It  
335 is therefore useful to consider the average profile through a stratiform cloud layer (Figure 6 (c) and (d)).  
336 Figure 6(d) demonstrates that, as in the case of the individual cloud layer shown in Figure 2, on average,  
337 positive charge exists at the cloud top region, and negative charge in the cloud base. At the cloud top, the

338 maximum mean charge is  $+32 \text{ pC m}^{-3}$ , and at the base  $-24 \text{ pC m}^{-3}$ , suggesting an asymmetry between cloud  
339 top and cloud base. The theoretical expectation is that this results from the vertical conductivity gradient,  
340 which, in general is larger at cloud top than in the base (see Harrison et al 2015), and discussed in more  
341 detail here in section 5.1. It is also interesting to note that, on average, the negatively charged region is  
342 relatively shallow and rapidly reaches a maximum value (at  $\sim 20\%$  cloud depth fraction), whilst  
343 the positively charged region is more vertically extensive (from 40-90% of the cloud depth fraction).  
344 The fact that charge is observed away from the upper and lower cloud boundaries is likely to be  
345 due to mixing of charge through vertical motion from updrafts and downdrafts within the cloud  
346 volume. Although the stratiform nature of the clouds will minimise vertical motions, local  
347 variability and updrafts of up to  $1 \text{ ms}^{-1}$  nevertheless occur (e.g. Peng et al, 2005). An asymmetry  
348 in the vertical extent of updrafts compared with downdrafts will result in different mixing profiles  
349 between cloud top and cloud base, which would influence the vertical extents of average positive  
350 and negative charge regions. Although the polarity of the average vertical space charge profile agrees  
351 with theoretical predictions, on an individual cloud to cloud basis, considerable variability exists. This was  
352 originally documented by Imyanitov and Chubarina (1957) in a series of aircraft flights measuring electric  
353 field,  $E_z$ , from which the space charge profiles could be derived using equation 1. They found that only  
354 41 % of 70 flights contained positive charge in the upper regions and negative charge in the base, with a  
355 mean space charge density of  $7.3 \text{ pC m}^{-3}$  in the upper part, and  $-5.3 \text{ pC m}^{-3}$  in the lower part. Similarly to  
356 Imyanitov and Chubarina (1957), the instrumented balloon flights from Reading, Hyttiala and Halley find  
357 that 57% of the 22 sampled stratiform clouds contain positive charge in their upper regions, and negative  
358 charge in the lower regions, whilst only 5% (1 flight) have an inverted charge polarity structure (i.e.  
359 negative charge at top and positive in base). 33% of cloud layers contained positive charge at both cloud  
360 top and base, and 5% (i.e. 1 flight) negative charge at cloud top and base. Thus, on average the charge  
361 structure within low level stratiform clouds agrees with that predicted by theory, but individual clouds  
362 may not. This is likely to be attributable to the oversimplification of cloud edge charging theory, which  
363 does not take into account the dynamical motions within a cloud. For example, the existence of entirely  
364 positively charged clouds may be due vertical mixing processes which act to transport positive charge

365 downwards into the base of the cloud. The observed asymmetry between cloud top and cloud base  
366 charge could hypothetically cause positive charge to dominate over the negative, thus leading to a net  
367 positive charge throughout the cloud. The extent to which the theory holds for the set of measured layer  
368 clouds will be examined in the next section.

369 -

370 **5. Factors controlling cloud edge charging**

371 Equation 3 describes the cloud edge charge in solely steady state electrostatic terms, specifically the  
372 variation with height of the air conductivity and the local vertical conductivity gradient. This section will  
373 now deal with each of these factors in turn.

374

375 **5.1 Conductivity gradient**

376 One property which shows great variability between clouds is the thickness of the transition zone  
377 between clear air and the horizontal cloud edge, which is directly related to the vertical gradient in  
378 conductivity, and also the generation of space charge in accordance with equation 3. A direct  
379 measurement of the cloud edge transition zone thickness can be obtained from the optical measurements  
380 made by the cloud droplet sensor. An example of a particularly narrow (or “sharp”) cloud edge transition  
381 zone is shown in the cloud layer in figure 3, where the optical transition from cloud to clear air at the  
382 cloud top takes place over a depth of 30m. In this cloud layer the space charge within the cloud top is co-  
383 located exactly where the optical cloud changes occur, with the magnitude of the space charge increasing  
384 as the visibility decreases. The charge at cloud top in this particular cloud layer is so large that it saturated  
385 the charge sensor (giving a lower limit of charge as  $>200\text{pCm}^{-3}$ ). The same cannot be said for the cloud  
386 base region, however, which has a much less distinct change from clear to cloudy air, and hence no  
387 appreciable amount of space charge is located in the cloud base region. For all of the 22 stratiform  
388 clouds studied here the nature of the cloud top and cloud base transitions, with the cloud top transition  
389 extending vertically over a narrower region, is consistent between clouds. This is expected from the often  
390 sharp temperature inversion in the cloud top region, which acts as a lid to further upward motion, which

391 is not present in the cloud base region. The magnitude of the charge which accumulates at horizontal  
392 cloud edges is therefore constrained by thermodynamical processes.

393

394 Figure 7 shows boxplots of the cloud edge transition zone thickness, visibility gradient and space charge  
395 measured at both horizontal cloud edges of cloud top and cloud base for 16 of the cloud layers with  
396 altitudes <3km. The cloud base was identified on the basis of determining where the optical cloud droplet  
397 sensor voltage began increasing, and cloud top from where it was decreasing. The cloud edge transition  
398 zone thickness was also selected on this basis. It is seen that in general, the cloud edge transition zone is  
399 narrower at cloud top (median depth 72m) than cloud base (median depth 128m). Similarly the gradient  
400 in visibility (which is related to the cloud droplet concentration and conductivity by equations 5, 6 and 7),  
401 is also larger at cloud top (42) than cloud base (21). In agreement with this, the magnitude of the space  
402 charge in cloud top regions is also greater at cloud top (median 45 pCm<sup>-3</sup>) than at cloud base (median 29  
403 pCm<sup>-3</sup>), demonstrating an asymmetry in charge between cloud top and cloud base. It follows that the  
404 largest magnitude of cloud edge charging will occur in stable stratified layers with sharp edges and little  
405 vertical mixing. These properties are typical of marine stratocumulus, which are characterised by strong  
406 temperature inversions at cloud top.

407

## 408 **5.2 Conductivity and altitude variation**

409 Conductivity varies substantially with height because of the variation in cosmic ray ion production, and  
410 contribution from radon near the surface, therefore to investigate the height dependence of cloud edge  
411 charge it is first necessary to determine the vertical profile of conductivity. Conductivity profiles can be  
412 measured directly e.g. by a Gerdien type sensor (e.g. Nicoll and Harrison, 2008) or indirectly by measuring  
413 the ion production rate. Measurements showing the ion production rate variation with height have been  
414 presented by Harrison et al, 2014 using the balloon-borne Geiger sensor described in section 3.1. In this  
415 study measurements of vertical profiles of ionisation rate have been obtained at all three cloud  
416 measurement sites of Reading, Hyttiala and Halley which allow the average vertical ion production rate

417 profile from the surface up to 5km for all of the sites together to be found, as well as the clear air  
418 conductivity profile (shown in Figure 8 (a)).

419

420 To determine the effect of cloud height on the generated space charge an estimate of the in-cloud  
421 conductivity profile has been derived from the cloud droplet profile of the cloud shown in Figure 2, typical  
422 of the average cloud profile, and equation 6. The predicted magnitude of the mean space charge in the  
423 cloud is then calculated from equation 3, using the calculated conductivity values from Figure 8 (a). This  
424 is then compared with the measured mean value (magnitude) of space charge measured from all 22 cloud  
425 layers at the 3 measurement sites (where the predicted and measured values of charge are shown as the  
426 grey line and black points in Figure 8 (b) respectively). It is seen that the predicted space charge follows  
427 closely the inverse variation in ionisation rate with height, with a reduction in ionisation at approximately  
428 1km where the radon contribution falls off. Although the same variation is not so evident in the measured  
429 space charge values, the black line depicts a lowess fit to the data (locally weighted scatterplot regression)  
430 which does demonstrate elevated space charge values in the lowest 1.5km, which falls off rapidly with  
431 increasing height. This is supported by the boxplot in Figure 8 (c) which divides the measured mean space  
432 charge values according to the height of cloud in which they were measured. It is clear that there is a  
433 statistically significant difference (at the 95% confidence level) in the mean space charge measured in  
434 clouds below 2km and above 2km, with lower altitude clouds being more highly charged. The median of  
435 the mean space charge distribution for the low altitude (<2km) clouds is  $20.2 \text{ pCm}^{-3}$ , and  $7.0 \text{ pCm}^{-3}$  for the  
436 higher altitude clouds (>2km). The ratio of these two space charge values is 2.9 which is very similar to  
437 the ratio of mean conductivity (=2.8) below and above 2km from Figure 8(a). There is also a noticeable  
438 difference in the range of the space charge values between the two altitude ranges, with the inter-quartile  
439 range (IQR) =  $11.5 \text{ pC m}^{-3}$  for low altitude clouds compared to IQR =  $5.4 \text{ pC m}^{-3}$  for the higher altitude  
440 clouds, potentially related to the smaller magnitude of space charge in the higher altitude clouds. The  
441 data shown in Figure 8 thus supports the hypothesis that the droplet charging effect is greatest for low

442 altitude stratiform cloud, which would be particularly so over the oceans, where any radon contribution  
443 to the total ionisation rate is negligible.

444

445 Substantial scatter in the data points is expected as a number of factors influence the space charge  
446 generation in clouds in addition to the conductivity profile. No allowance has been made here for  
447 differences in cloud edge transition zone thickness between low altitude and high altitude clouds, which  
448 may result from dynamical changes between the different cloud altitudes. Further, it should be noted  
449 that the assumption of using the average cloud droplet profile for all altitudes of cloud between 1 and  
450 5km is another potential source of variability, which also does not take account of dynamical changes in  
451 clouds that typically occur with altitude changes. Although it would be preferable to repeat this analysis  
452 to take these points into consideration, as well plot the data from each of the three measurement sites  
453 individually, the small number of data points limits the statistical analysis possible.

454

## 455 **6. Cloud droplet charging**

456 The results shown in section 4 combined with theory provide strong evidence that widespread charging  
457 occurs within the edge regions of stratiform cloud layers. Simultaneous charge and cloud droplet  
458 measurements also indicate that the charge is likely to reside primarily on the cloud droplets, since charge  
459 is typically observed to be present mainly within the region where the visual range indicates that cloud is  
460 present. As mentioned in section 1, there are several physical consequences associated with the charging  
461 of cloud droplets, including modification of droplet-particle interactions and droplet activation, however  
462 the extent to which the droplet behaviour is influenced depends on the magnitude of the droplet charge.  
463 Since most of the space charge within a cloud will reside on cloud droplets it is possible to represent the  
464 measured space charge density values discussed in section 4 in terms of elementary charges per droplet,  
465 rather than charge per unit volume. If the charge density  $\rho$  is assumed to be partitioned equally between  
466 cloud droplets with concentration,  $Z_d$ , the mean number of elementary charges per droplet  $j$  is given by

467

$$j = \frac{\rho}{Z_d e} \quad 7,$$

468 Summary histograms of individual cloud droplet charges are shown in Figure 9 for (a) cloud base and (b)  
469 cloud top regions, for all 16 cloud layers with altitude <3km (with cloud base and top selected on the basis  
470 of changes in the optical cloud droplet sensor response, as for Figure 7). For each cloud layer, equation  
471 7 was applied to the measured space charge values, together with the calculated equivalent cloud droplet  
472 number concentration profile (calculated assuming cloud droplets are monodisperse with 10 $\mu\text{m}$   
473 diameter). At cloud base, the range of cloud droplet charges from Figure 9 (a) is -270 to 57e, with median  
474 -0.4e, whilst at cloud top droplet charges range from -26 to 31e, with median 1.4e. Since the droplet  
475 charge is calculated directly from the space charge measurements, overall the droplet charges are largely  
476 negative at cloud base and positive at cloud top, with a slight asymmetry between the two in that cloud  
477 droplet charges at the cloud top are slightly larger than in the base.

478

479 **7. Discussion**

480 This work presents the first quantitative comparison of multiple observations of stratiform cloud edge  
481 charging from multiple sites. Although general agreement between observations and theory in the  
482 location and polarity of the charge layers is found on average across all the soundings, substantial local  
483 differences are apparent in individual clouds. The results discussed here therefore demonstrate that cloud  
484 edge charging is dependent on both the dynamical properties of the cloud as well as the background  
485 electrical environment. Turbulence and updrafts and downdrafts inside the cloud layer will act to mix the  
486 charge generated at the cloud edges, and the presence of ice as well as varying aerosol concentrations  
487 (which are not considered here) are also likely to play a role in the resultant charge profile.

488 The confirmation of the asymmetry in charging between cloud base and top is an important factor which  
489 has hitherto not been included in modelling studies of layer cloud electrification, and has potentially  
490 important consequences for larger charging of droplets at cloud top than base. In terms of the  
491 implications for cloud microphysical processes, the magnitude of the estimated droplet charges (from a

492 few  $e$  per droplet to  $270e$ ) are large enough to affect droplet-droplet collision processes, but likely not  
493 droplet activation, which is thought to require charges of  $\sim 1000e$ . Harrison et al (2015) describes the  
494 increased collision rate between small and large droplets when both droplets are equally charged through  
495 an increase in the collision efficiency (i.e. the likelihood of droplets colliding with each other), which  
496 results from the mutual attraction of the droplets from the electrical image force. This occurs even when  
497 the droplets have the same polarity of charge (as would typically be the case in an unmixed cloud layer),  
498 and will be most dominant for droplets ( $< 2\mu\text{m}$ ), likely resulting in a depletion in the number concentration  
499 of small droplets.

500 Over-range of the charge sensor occurred on one flight (shown in Figure3), demonstrating that space  
501 charge regions  $> 200\text{pCm}^{-3}$  can exist within stratiform clouds. Measurements from the optical cloud  
502 droplet sensor presented here provide a more realistic value of cloud edge depth of  $\sim 100\text{m}$  than that  
503 previously used in modelling studies (e.g. 11m in Zhou and Tinsley, 2007), and the large variability  
504 observed in the charge profiles on an individual cloud to cloud basis suggest that the simple modelling  
505 approach adopted up to now needs to be extended for future studies. These therefore present a unique  
506 dataset of in-cloud stratiform charge measurements which can help to inform future modelling studies of  
507 cloud droplet charging effects on cloud microphysics (e.g. Harrison et al 2015; Zhou and Tinsley 2012;  
508 Tinsley and Leddon 2013).

509

510 Although not considered in detail here, variations in the air Earth conduction current may also contribute  
511 to some of the variability in cloud edge charging since it is the flow of this vertical current through cloud  
512 layers which leads to the accumulation of space charge at cloud edges.  $J_c$  is controlled by a number of  
513 factors internal to Earth's climate system, which are mainly associated with the Global atmospheric  
514 Electric Circuit (GEC). This is driven by global thunderstorm activity, therefore changes in thunderstorm  
515 output current and lightning activity (which are known to be linked to surface temperature changes and  
516 global weather patterns such as ENSO (Harrison et al. 2011; Williams 1992) will act to increase or decrease  
517  $J_c$ . Variations in the vertical conductivity profile, such as from volcanic eruptions, or even the presence of

518 cloud layers themselves (e.g. Baumgaertner et al 2014, Zhou and Tinsley 2010, Nicoll and Harrison 2009)  
519 also play a role in modulating  $J_c$ . Conductivity variations also occur, more dominantly, through external  
520 factors such as changes in the GCR flux (e.g. decreases in global ionisation rates of up to 10% can occur  
521 during Forbush decreases, typically associated with explosive events on the sun), and also from solar  
522 proton events. Understanding the effects of space weather on the GEC and therefore  $J_c$  should therefore  
523 remain an area of active research, particularly as solar disturbances have the potential to affect cloud  
524 edge charging through the mechanism discussed here (see Nicoll 2014 and Mironova et al 2015 for recent  
525 reviews). There is no doubt that assessing the importance of the role that  $J_c$  plays in modulating cloud  
526 edge charging is difficult to do. Measurements of  $J_c$  are sparse, and because significant variability in cloud  
527 charge profiles exists between individual cloud layers, a larger statistical sample of cloud charge data is  
528 therefore required to properly assess this important factor.

529

## 530 **8. Conclusions**

531 These results represent the first multiple site comparison of non-thunderstorm cloud charge, and the first  
532 study to undertake simultaneous high vertical resolution measurements of cloud droplet and charge  
533 properties in multiple cloud layers and compare them with theoretical charging expectations. The cloud  
534 and charge data demonstrates unequivocally that all stratiform clouds can be expected to contain charge  
535 at their upper and lower boundaries to varying extent, due to vertical current flow in the Global  
536 atmospheric Electric Circuit. On average, charge is found near upper and lower cloud edges and the cloud  
537 edge charge polarity (positive at cloud top and negative at cloud base) agrees with theory, but large  
538 variations in both location and polarity of charge are observed within individual cloud layers, likely due to  
539 dynamical processes within the clouds, which are at present not included in theoretical models of cloud  
540 edge charging.

541

542 The results also demonstrate that a combination of cloud thermodynamics with the background electrical  
543 conditions contribute to the magnitude of cloud edge charging in terms of the “sharpness” of the cloud

544 edge transition zone thickness. This is observed in terms of an asymmetry between cloud top and base  
545 charge, typically with larger charge observed at cloud top ( 32 pCm<sup>-3</sup> compared to 24 pCm<sup>-3</sup> in the cloud  
546 base) due to the more rapid transition from cloudy to clear air which often occurs due to a significant  
547 temperature inversion in the cloud top region.

548

549 Finally, the difference in cloud charge robustly observed between low altitude (<2km) and higher altitude  
550 stratiform clouds (2-5km) demonstrates the important role of cosmic ray ionisation in the cloud edge  
551 charging process, if the other edge properties of clouds remain unchanged. This confirms the theoretical  
552 expectation that layer cloud electrification will be at its greatest for low level clouds.

553

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567 archived through the University of Reading Data repository at <http://researchdata.reading.ac.uk/38/>.

568

569 **Appendix. Effective area of charge sensor**

570 The balloon-borne charge sensor used to provide the in-cloud charge measurements employs an  
 571 approximately spherical electrode connected to a sensitive electrometer, and primarily responds to  
 572 induced displacement currents generated by electric field changes as the sensor moves through the cloud  
 573 layer. A full description of the sensor electronics is given by Nicoll, 2013, with further details of the  
 574 calculation of cloud space charge given here.

575

576 The electrometer circuit comprises a current to voltage converter employing a T-network of resistors to  
 577 synthesise a total effective resistance,  $R = 2.4 \times 10^{11} \Omega$ . The induced current,  $i$ , is related to the final output  
 578 voltage of the electrometer circuit,  $V_{out}$ , by

$$579 \quad i = \frac{V_{out}}{R} \quad \text{A1.}$$

580 To calculate the space charge density,  $\rho$ , first consider the charge,  $Q$ , induced on a stationary sensor  
 581 electrode due to the electric field. This is given by Gauss' law as

$$582 \quad Q = -A_{eff} \varepsilon_0 E \quad \text{A2,}$$

583 where  $A_{eff}$  is the effective area of the electrode i.e. the area of the conductor on which the field lines end,  
 584 which is not necessarily the same as its geometrical surface area. If the sensor is allowed to move, for  
 585 example by ascending vertically through a cloud layer, the electric field will change, and the induced  
 586 charge is no longer constant, causing an induced current,  $i$ , to flow, measured by the electrometer and  
 587 given by

$$588 \quad i = \frac{dQ}{dt} = -A_{eff} \varepsilon_0 \frac{dE}{dt} = \frac{V_{out}}{R} \quad \text{A3.}$$

589 For vertical motion on a free balloon with ascent rate  $w$ , the  $dE/dt$  term in equation A3 is

$$590 \quad \frac{dE}{dt} = \frac{dt}{dz} \frac{dE}{dt} = \frac{1}{w} \frac{dE}{dz} \quad \text{A4,}$$

591 and therefore the current measured by the electrometer is related to the electric field gradient by

$$592 \quad i = -A_{eff} \varepsilon_0 \frac{1}{w} \frac{dE}{dz} \quad \text{A5.}$$

593

594 From equation 1 and equation A5 and substituting for the  $dE/dz$  term, it follows that the space charge,  $\rho$ ,  
595 measured by the charge sensor is related to the measured current by

596 
$$\rho = \frac{i}{A_{eff}w} \quad A6.$$

597 The unknown quantity in equation A6 is the effective area term,  $A_{eff}$ . This can be determined from  
598 experimental calibration using equation A3, by placing the charge sensor in a varying electric field which  
599 is known.

600

601 The experimental calibration was undertaken by measuring the atmospheric electric field in fog (using an  
602 electric field mill at the Reading University Atmospheric Observatory (RUAO)) and placing the charge  
603 sensor nearby. (The field mill was previously standardised for its form factor using the passive wire  
604 antenna method, Bennett and Harrison 2006a.) Fog typically causes the atmospheric electric field to  
605 increase and become variable thus generating a sufficiently large  $dE/dt$  with which to calibrate the charge  
606 sensor. Figure A1 shows a time series of the rate of change of electric field (measured by the field mill, in  
607 black) and the current measured by the charge sensor during a 3 hour period of fog. The extremely good  
608 correlation between the two traces (Pearson correlation coefficient = 0.75) demonstrates that not only  
609 does the balloon charge sensor respond well to changes in electric field, and thus space charge, but its  
610  $dE/dt$  response is very similar to that of the commercially available field mill. Figure A2 shows the  
611 relationship between the current measured by the charge sensor and  $dE/dt$  measured by the field mill.

612

613 The effective area of the charge sensor electrode can thus be found from a linear fit between  $i$  and  $dE/dt$   
614 (Figure A2) as

615

616 
$$i = -A_{eff}\epsilon_0 \frac{dE}{dt} \quad A7,$$

617 from which it is calculated that  $A_{eff} = 0.0196 \text{ m}^2$ . This can be compared to the approximate surface area  
618 of a perfectly spherical electrode (with radius 0.6cm),  $A = 0.000452 \text{ m}^2$ , i.e. approximately 43 times smaller  
619 than the effective area, which is reasonable considering the likely electric field distortion from the

620 relatively complex geometry of the charge sensor electrode. The final space charge value is then  
621 computed using  $A_{\text{eff}}$  above and equation A6, using the local ascent rate in the cloud as found by the  
622 radiosonde height information.

623

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770 **Figures and Tables**

771

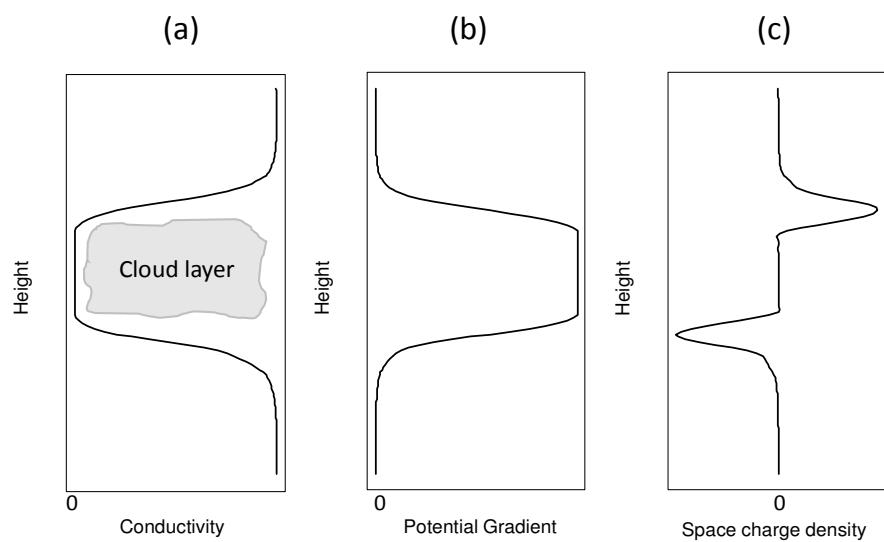
Cloud number	Date	Location	Mean cloud height (m)	Cloud depth (m)	Mean space charge (pCm <sup>-3</sup> )	Max. space charge (pCm <sup>-3</sup> )
1	22/04/13	Reading	1534	446	39±7	204
2	03/07/13	Reading	1846	227	23±6	143
3	04/07/13	Reading	735	640	43±6	253
4	31/07/13	Reading	790	622	30±5	200
5	16/08/13	Reading	720	242	29±6	153
6	16/08/13_2	Reading	1200	90	18±6	151
7	23/08/13	Reading	4553	306	5±1	32
8	31/10/13	Reading	2522	333	9±3	148
9	18/11/13	Reading	1146	328	16±4	174
10	18/11/13_2	Reading	4197	135	2±1	10
11	02/12/13	Reading	1039	174	19±4	97
12	03/12/13	Reading	801	374	21±4	172
13	27/05/14	Hyytiala	4838	163	9±2	75
14	27/05/14_2	Hyytiala	1849	208	16±5	104
15	29/05/14	Hyytiala	3311	120	29±6	113
16	29/05/14_2	Hyytiala	3907	289	11±3	133
17	30/05/14	Hyytiala	2345	86	4±13	52
18	30/05/14_2	Hyytiala	499	178	23±8	127
19	20/02/15	Halley	1011	570	6±1	42
20	21/02/15	Halley	974	577	13±1	92
21	06/03/15	Halley	475	281	5±1	32
22	06/03/15	Halley	3067	1700	4±1	102
<b>Median</b>			<b>1367</b>	<b>285</b>	<b>16</b>	<b>120</b>

772

773 Table 1. Details of cloud and charge sensor balloon flights through stratiform clouds from 2013-2015. The  
 774 mean cloud height is the height of the centre of the cloud and the cloud depth is the difference in height  
 775 between cloud base and top. The mean and maximum values of space charge are calculated from the

776 magnitude of the in-cloud space charge for each cloud layer. The uncertainty in the mean charge is 2  
777 standard errors on the mean values which represents the 95% confidence interval. Balloons which  
778 encountered several different layers of stratiform cloud on the same flight are denoted by “\_2” in the  
779 flight date. N.B. all data is from the ascent stage of the flight.

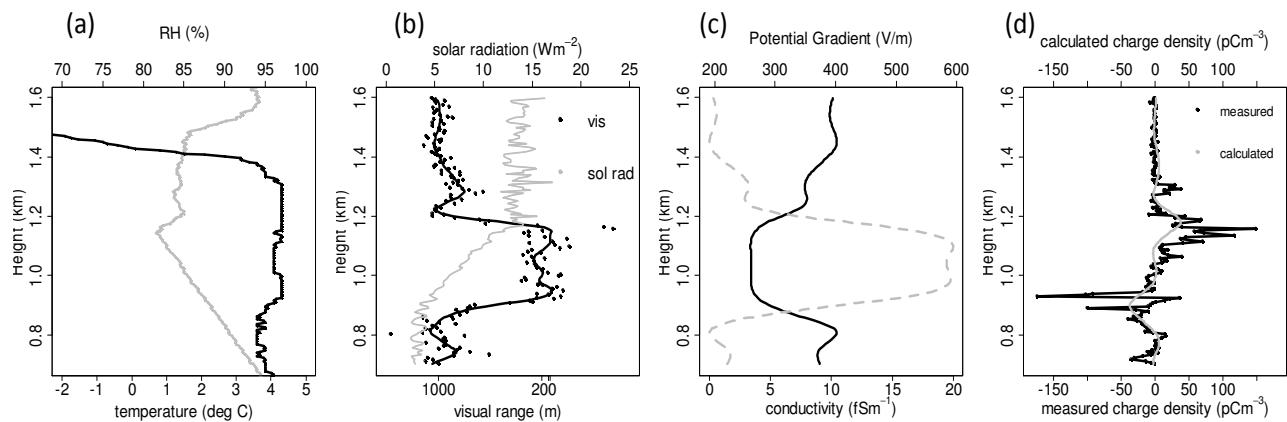
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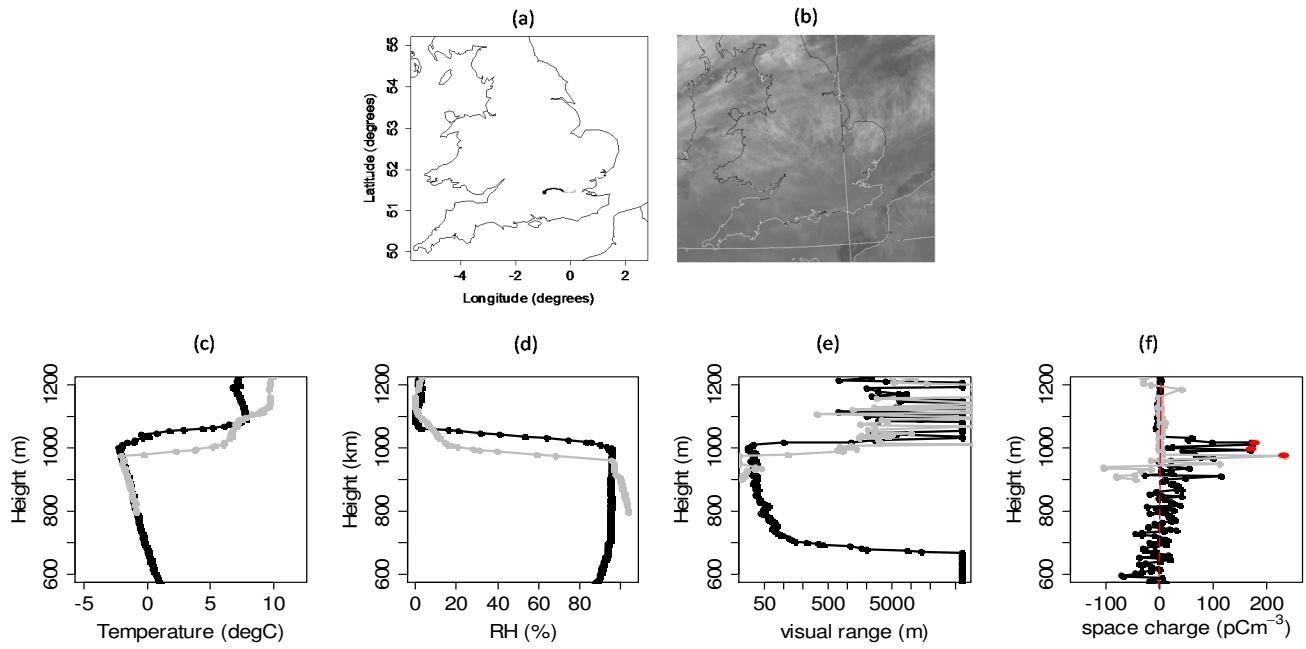
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785 Figure 1. Hypothetical vertical profiles of atmospheric electrical quantities through a horizontally-  
 786 extensive stratiform cloud layer (a) conductivity, (b) potential gradient and (c) derived space charge  
 787 density.

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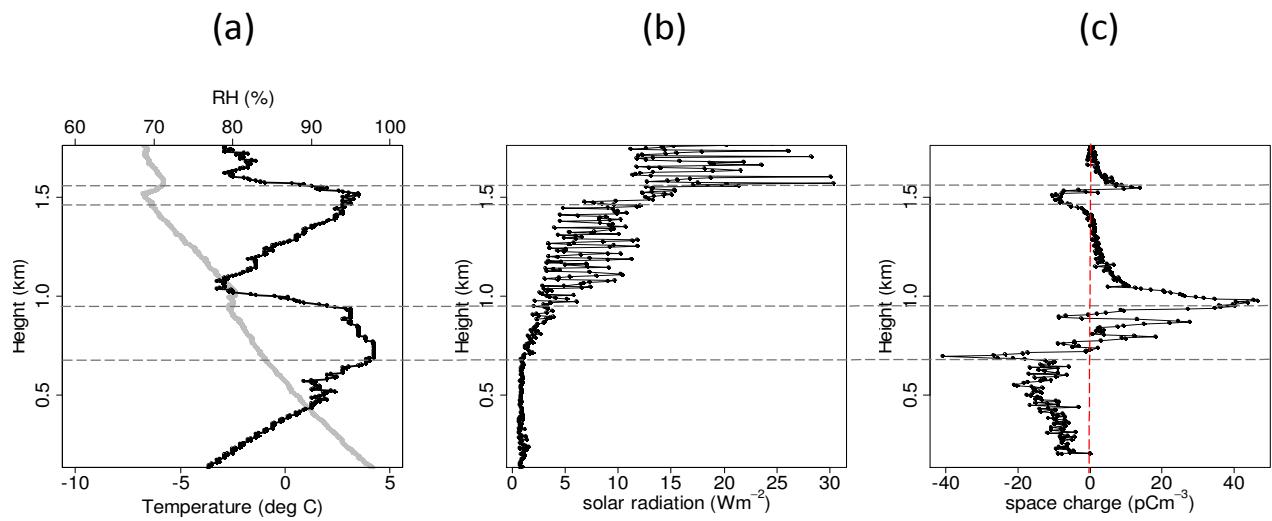
791 Figure 2. Vertical profile through a stratocumulus layer over Reading University Atmospheric Observatory  
 792 (RUAO) from a specially instrumented radiosonde. (a) Temperature (grey) and relative humidity (RH)  
 793 (black) measured by the radiosonde, (b) visual range and downwards solar radiation measured by an  
 794 optical cloud droplet sensor, (c) derived parameters of electrical conductivity (black) and potential  
 795 gradient (grey dotted line) using equations 2, and 4-6 and assumptions outlined in section 3.1 , (d) space  
 796 charge density measured by a charge sensor (black line and data points) plotted alongside the expected  
 797 space charge (grey line) calculated from the cloud droplet sensor measurements using the theory in  
 798 section 3.1.



800

801 Figure 3. Ascent and descent of an instrumented balloon flight through the same cloud layer on 03/12/13.  
 802 (a) shows the flight path of the balloon (ascent in black, descent in grey), launched from Reading, UK. (b)  
 803 demonstrates the presence of a low level cloud layer widespread over most of the UK as measured by the  
 804 AVHRR satellite (IR channel 4) at 10:27am, the same time as balloon launch time. Temperature and RH  
 805 measured by the radiosonde are shown in figures (c) and (d), and data from the cloud and charge sensors  
 806 shown in figures (e) and (f). Note that the visual range data is plotted on a log scale. Black shows the  
 807 ascent, grey the descent through the same cloud layer 105 km from the launch point. Red points in (f)  
 808 denote charge values where the charge sensor saturated therefore these values are a lower estimate of  
 809 the charge in this region.

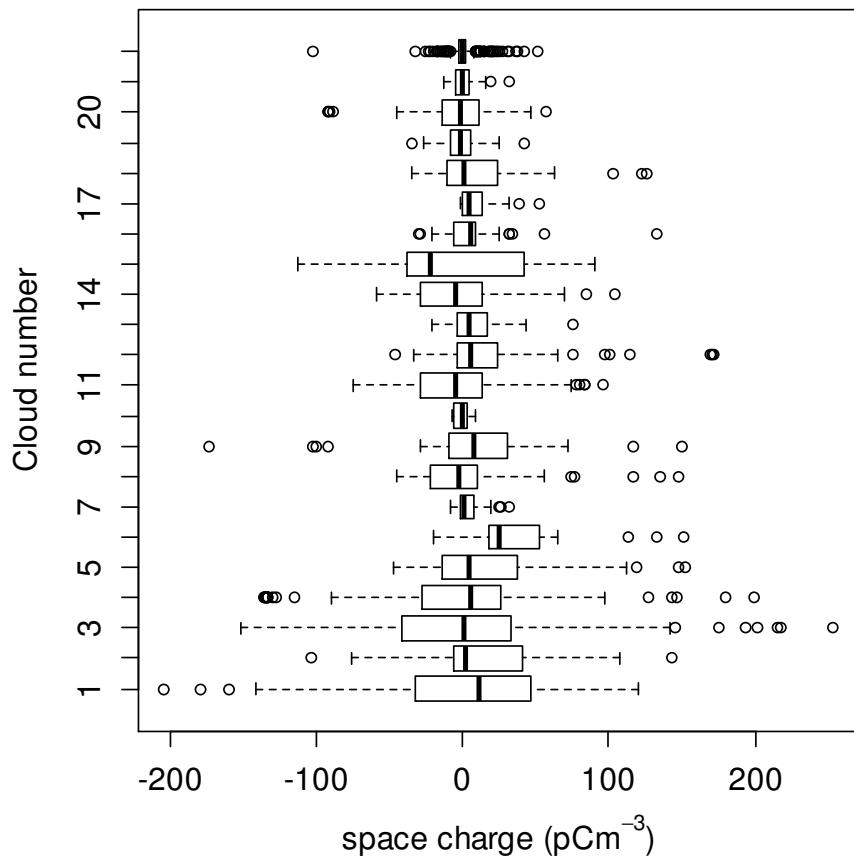
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812 Figure 4. Vertical profile from an instrumented balloon flight through multiple stratiform cloud layers,  
 813 launched from Reading on 08/02/13. (a) temperature and RH measured by the radiosonde, (b) solar  
 814 radiation measured by the passive cloud sensor, (c) space charge measured by the charge sensor. Grey  
 815 dashed lines denote approximate heights of cloud base and top.

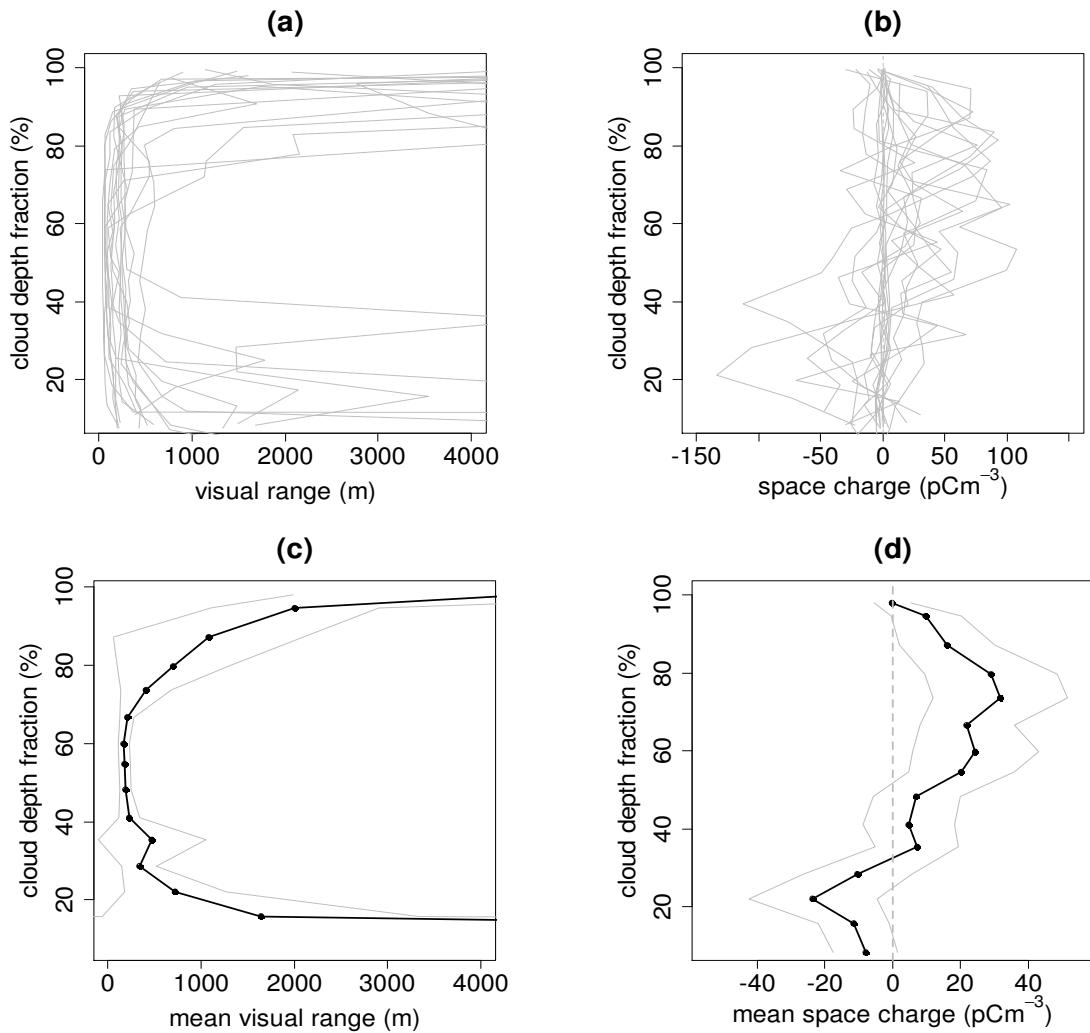
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818 Figure 5. Boxplot of the space charge distributions within each of the 22 studied cloud layers described in  
 819 Table 1. The black solid line denotes the median of the distribution and the whiskers the interquartile  
 820 range.

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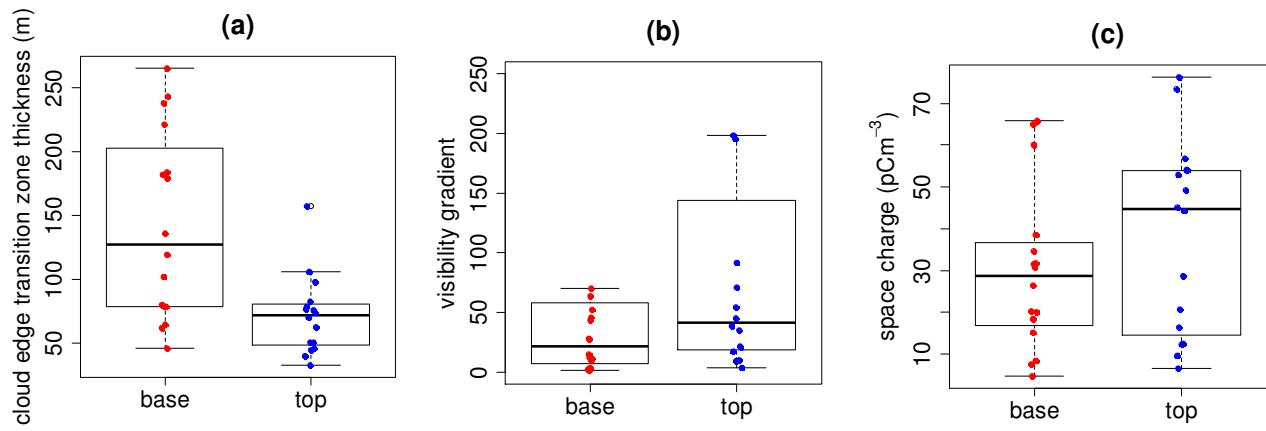


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823 Figure 6. Profiles from 16 cloud and charge sensor balloon flights through stratocumulus clouds with  
 824 height <3km. (a) and (b) individual profiles from flights, (c) and (d) average profiles calculated from the  
 825 16 flights. (a) and (c) visibility derived from cloud sensor, (b) and (d) space charge measured by the charge  
 826 sensor. The y-axis denotes height normalised by cloud depth, found by dividing each cloud layer into 15  
 827 evenly spaced altitude layers, where 0% denotes cloud base and 100% cloud top. The data points in (c)  
 828 and (d) are calculated from the mean of the variables in each of the 15 altitude layers. Grey solid lines in  
 829 (c) and (d) show two standard errors on the mean values.

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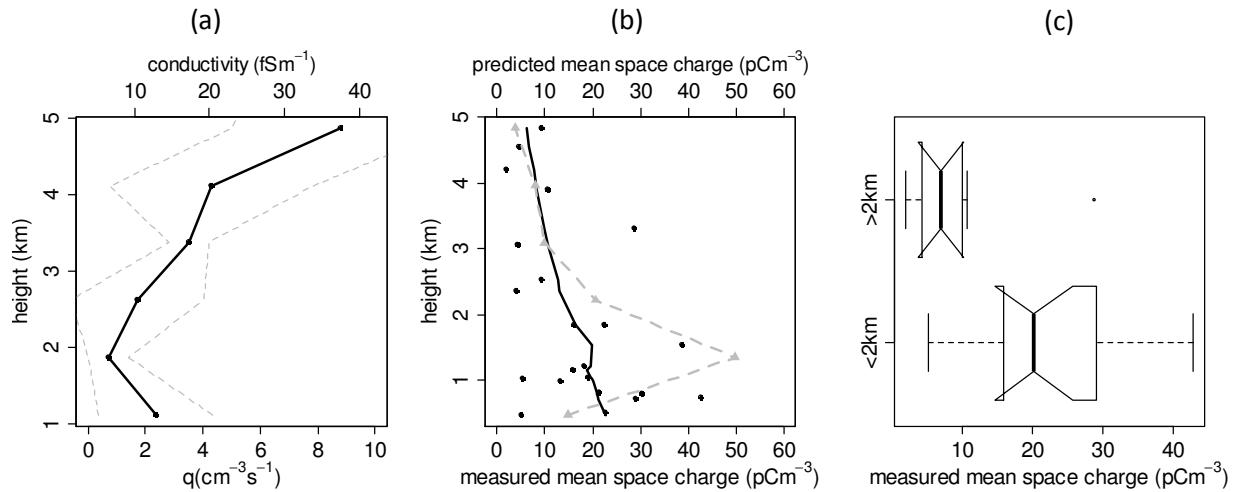
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832

833 Figure 7. (a) vertical extent of transition region between cloudy and clear air, (b) gradient in visibility at  
 834 cloud edge measured by cloud sensor, (c) magnitude of space charge measured by charge sensor. Left  
 835 hand box plots (left) represent cloud base (red) and right hand plots cloud top (blue). Data are  
 836 averages over cloud top/base region, where each point represents one cloud edge and only include  
 837 cloud layers <3km altitude. Cloud base regions are selected on the basis of when the optical cloud  
 838 droplet sensor voltage was increasing, and cloud top when it was decreasing.

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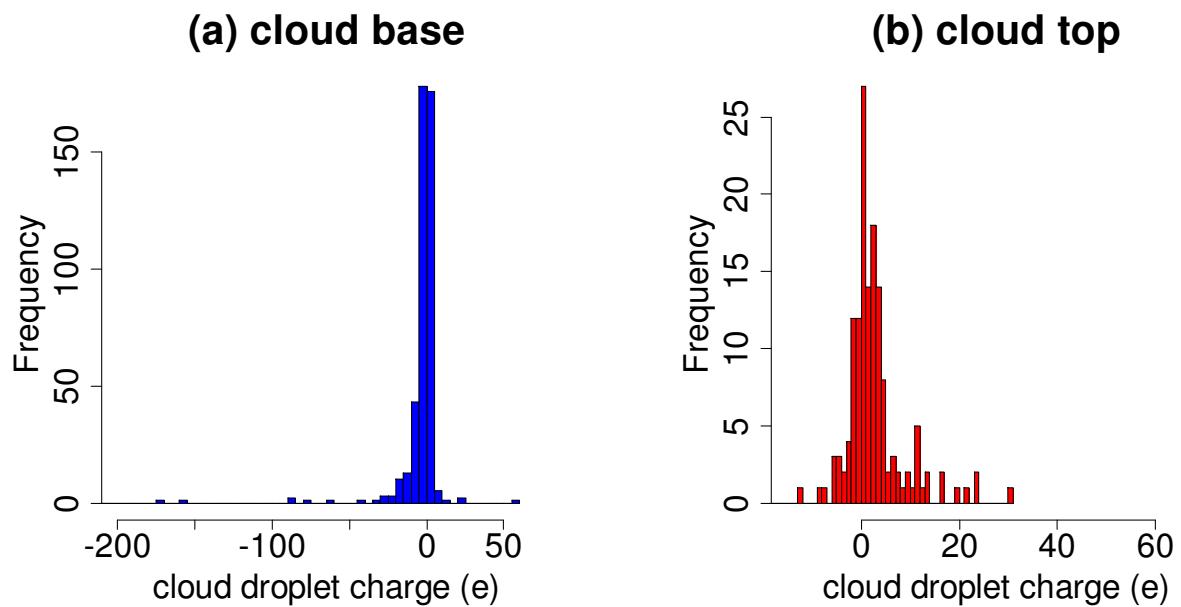


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841 Figure 8. (a) average vertical profile of ionisation rate measured by a balloon borne Geiger sensor and  
 842 corresponding conductivity profile calculated from equation 6 for Reading, Hyytiala and Halley between  
 843 2013 and 2015. Grey lines denote 2 standard errors on the mean values. (b) magnitude of the mean in-  
 844 cloud space charge (grey dotted line), predicted as a function of height, using the average cloud droplet  
 845 profile of the case in Figure 2, and the ambient conductivity profile in Figure 8 (a), and measured mean  
 846 space charge (absolute values) from all 22 cloud layers at Reading, Hyytiala and Halley (black points and  
 847 black line, which shows a lowess fit to the data points). (c) boxplot of measured mean space charge values  
 848 separated into two cloud height ranges, with mean cloud height <2km (14 cloud layers) and >2km (8 cloud  
 849 layers). The edges and line in the centre of the boxes show the upper and lower quartiles and the median.  
 850 Notches indicate the 95% confidence limits on the median and whiskers extend to 1.5 times the inter-  
 851 quartile range.

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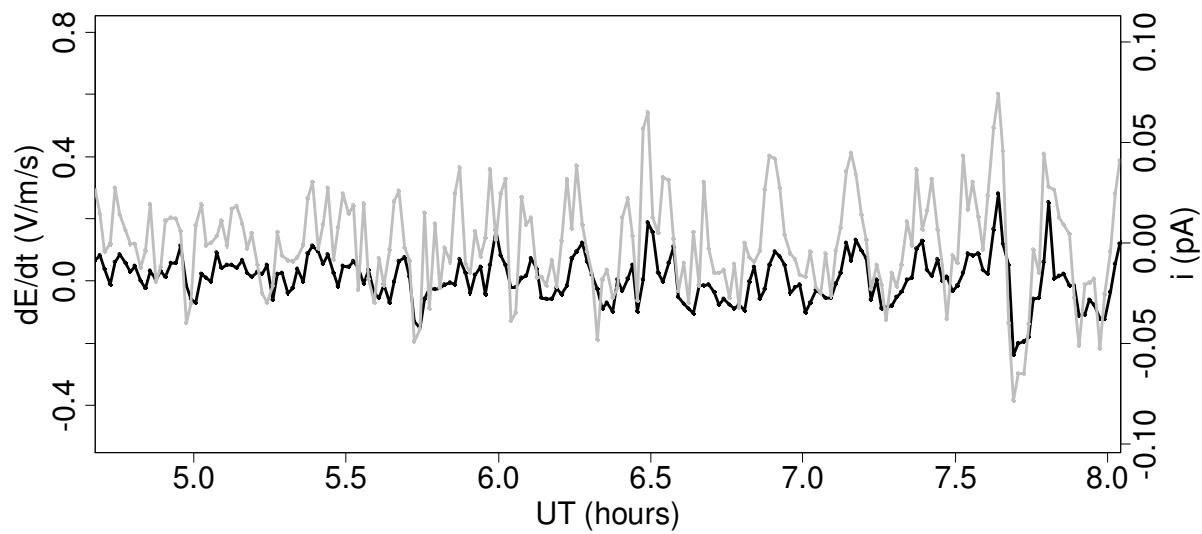


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855 Figure 9. Histograms of individual cloud droplet charges (in units of elementary charge, e), calculated from  
 856 measured space charge and calculated equivalent cloud droplet number concentration (assuming cloud  
 857 droplets are monodisperse with  $10\mu\text{m}$  diameter), for (a) cloud base and (b) cloud top for 16 stratiform  
 858 cloud layers  $<3\text{km}$  altitude. (a) contains 444 data points and (b) 147 points, since the cloud top region is  
 859 typically much shallower than the base.

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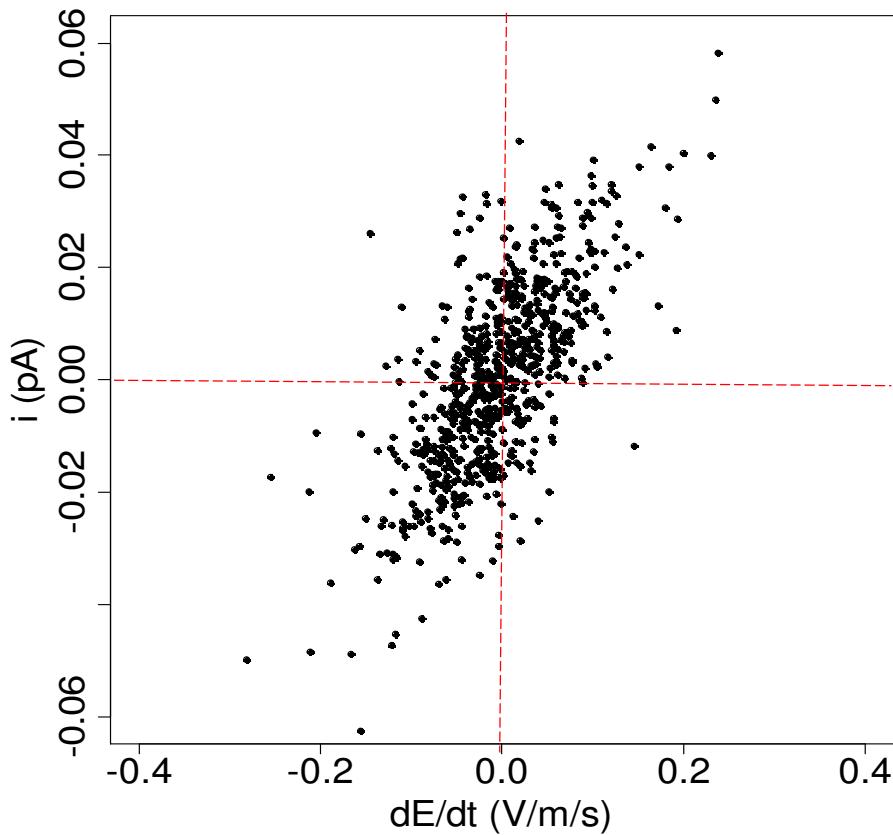
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862

863 Figure A1. Time series of data from calibration of balloon borne charge sensor in fog on Reading University  
864 Atmospheric Observatory (RUAO). The electric field change ( $dE/dt$ ) measured by a commercial field mill  
865 (JCI 131) is shown in black and the corresponding current measured by the charge sensor as a result of  
866 the electric field changes shown in grey.

867



868

869 Figure A2. Calibration of balloon borne charge sensor during fog-induced electric field changes. Sensor  
870 electrode current,  $i$ , is plotted against simultaneously measured rate of change of electric field ( $dE/dt$ )  
871 measured by a commercial field mill.

872