Title: Possible Effects of Ammonium on Lightning Polarity

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Abstract

This paper assembles evidence to discuss a mechanism that may contribute to the positive Cloud to Ground (+CG) lightning presence over the US Great Plains (GP). Data from EPA reveal maximum ammonium (NH_4^+) concentrations in precipitation over the entire US are observed over the GP. Laboratory experiments show NH_4^+ favors graupel's positive charging, a key-process for +CG enhancement. This study speculates that the spatial coherency between the NH_4^+ concentrations and the enhanced presence of +CG may not be a coincidence but relate to a physical mechanism that acts independently from the established thunderstorm charging processes.

1. Introduction

Perhaps the most interesting and intriguing feature of the US lightning climatology is observed over the region stretching from North Dakota to Oklahoma, trailing the north-central Great Plains (GP). It is over that region where the Cloud-to-Ground (CG) lightning flashes exhibit a distinct positive (i.e., +CG) polarity enhancement. Figure 1 illustrates the relative CG flash counts for both polarities (+CG/-CG ratio) as these were observed from the National Lightning Detection Network (NLDN, Cummins et al., 1998) during 2003-2010. The +CG/-CG ratio enhancement over the GP region has also been shown in numerous past studies (e.g., Lyons et al., 1998; Orville and Huffines, 2001; Koshak et al., 2014 and references therein).

Although still an elusive process, thunderstorm electrification is presently acknowledged as the result of rebounding collisions between graupel and ice crystals in the presence of super-cooled water (Takahashi, 1978; Saunders et al., 2006). Despite some disagreements in the findings or the employed laboratory techniques, the majority of research studies agree that the electrostatic charge exchange between ice crystals and graupel is a multi-parametric process where temperature, water content, drop size distribution and purity of the involved substances play a decisive role (see Jayarante, 2003). It is generally accepted that there is a graupel charging charge-reversal temperature which is a function of the respective water content. Takahasi and Miyawaki (2002) document that at temperatures T<~ -10^{o}C the main limiting factor is cloud liquid water content which, for values less than < ~0.1 g m^{-3} or > ~ 2.0 g m^{-3}, the graupel is charged positively whereas for the same temperature range accompanied by intermediate water content values, the opposite charging occurs. Emersic and Saunders (2010) expanded previous works and observed that the charge transfer between the ice particles (crystals and graupel) is dictated by the saturation of water/ice and the growth speed of the respective particles.

Williams et al., (2005, hereinafter W05) proposed that higher cloud bases over the GP result in larger updrafts that limit entrainment, resulting in higher water content available for the positive graupel charging. Moreover, higher cloud bases also imply lesser warm cloud depth and suppress warm rain process, allowing higher water amounts to be transported into the mixed layer. The latter is expected to invigorate the storm's updraft but also provide more readily available charging particles (i.e., ice crystals) that could lead to enhanced Intra Cloud (IC) lightning. The

latter is further supported by observations in Boccippio et al., (2001) and Medici et al., (2015).

Many follow-on studies supported W05 (Carey and Buffalo, 2007; MacGorman et al., 2008 and references therein). Field observations from the Severe Thunderstorm Electrification and Precipitation Study (Lang et al., 2004 and references therein) also documented that the majority of the +CG-enhanced storms exhibit an "inverted polarity" charge structure, where positively charged graupel is present in the mid-levels while negatively charged ice is present aloft (see Bruning et al., 2014 for an insightful discussion on inverted polarity). In line with W05, Fuchs et al., (2015) found that Colorado storms strongly favored +CG production over the other three regions studied (central Oklahoma, northern Alabama, and Washington, DC) but also featured significantly higher cloud bases and greater instability, which may enable greater liquid water contents in the mixed-phase region.

2. An alternative hypothesis for the +CG enhancement

The GP is a typical example of extensive anthropogenic impact and land-use modification, considering that the region encompasses the most intense agricultural and livestock farming activities across the US (http://www.agcensus.usda.gov/Publications/2012/Online_Resources/Ag_Census_Web_Maps/, Census of Agriculture). According to the Environmental Protection Agency (EPA) reports, these land-use practices are responsible for ~80% of the ammonia (NH_3) emissions across the US, especially from fertilizers and livestock urea (Battye et al., 1994; Konarik and Aneja, 2008).

Upon release into the atmosphere, NH_3 dissolves in cloud droplets and converts to ammonium ion (NH_4^+, Renard et al., 2004; The importance of Ammonia in the Atmosphere, http://www.esrl.noaa.gov/csd/AQRS/reports/ammonia.pdf). According to Stensland (2000), NH_3 vapor and NH_4^+ aerosols are scavenged by precipitation, with the atmospheric NH_4^+ predominately being the form of NH_4^+ sulfate [$(NH_4)_2SO_4$] or NH_4^+ bi-sulfate [$NH_4^+HSO_4^-$] particles that serve as condensation nuclei.

An ammonium wet-deposition network of over 260 sites across the US (National Atmospheric Deposition Program/National Trends Network, NADP/NTN, http://epa.gov/castnet/javaweb/precipchem.html, Konarik and Aneja, 2008) shows the NH_4^+ averaged annual concentrations for years 2003-2010 (Fig. 2). The qualitative comparison between Fig. 1 and Fig. 2 reveals that the highest NH_4^+ concentrations over the US trail part of the GP (Fig. 2). More importantly, one further notes some degree of spatial coherency with the enhanced +CG presence over shown in Fig. 1. [*Note that a "pixel-to-pixel" comparison between Fig. 1 and Fig. 2 should account for the fact that Fig. 2 is a product of interpolation from only ~15-20 NADP/NTN ground observations over the GP, which are subsequently mapped on rectangular grid of the substantially higher spatial resolution of Fig. 1, ~0.5 degrees*]

Although the pattern similarity between +CG enhancement (Fig. 1) and high NH_4^+ concentrations (Fig. 2) cannot establish any causality nor lead to any definitive conclusions, few interesting facts become available if we revisit the earlier discussion on the graupel charging mechanisms (section 1). Graupel is formed by the accretion of super-cooled cloud droplets, which in the "non-sterile" atmospheric conditions, unavoidably nucleate on the ambient soluble aerosol particles. Surprisingly, only a small number of studies have studied the charging properties of graupel in the presence of soluble ionic substances and in particular NH_4^+ (Jayarante et al., 1983; Jayarante et al., 1999; Chelf and Martin, 2001; Nelson and Baker, 2003; Jungwirth et al., 2005; Prakash and Kumar (2010). These studies agree that NH_4^+ demonstrates a pronounced effect on the charge transfer during the ice-graupel collisions. Although the mechanism is described as highly complex, laboratory experiments show that the NH_4^+ ions in the rime favor the positive (negative) charging of graupel (ice crystal) (see Yair, 2008). Jayarante (2003) further argues that although the mechanism has not been fully characterized, a possible key-player is the ionic mobility between the NH_4^+ (cation) residing in the inner graupel layer and the conjugent anion residing in the outer part of the graupel (e.g., SO_4^{-2}, frequently encountered in atmospheric conditions as $[NH_4]_2SO_4$, Stensland, 2000). Consequently, the collisions with ice crystals remove negative charge from the graupel (i.e., leaving it positively charged) while the negative charge is transferred to the ice particle. Chelf and Martin (2001)

further discuss that the presence of NH_4^+ can also effectively lower the ice nucleation temperatures, in agreement with Jayarante et al., (1999) and Prakash and Kumar (2010) who report that the graupel charging sign can be further dependent on the NH_4^+ concentrations and temperature. In particular, Prakash and Kumar (2010) document that positive graupel charging is enhanced at lower concentrations/higher temperatures (-6^{o}C to -15^{o}C) or higher concentrations/lower temperatures (-16^{o}C to -21^{o}C). Further implications have been raised by Jungwirth et al., (2005) and Prakash and Kumar (2010), where the authors discuss that the role of other soluble ions may also explain some of the +CG enhancement that is observed in smoke plumes advected at large distances from wildfires (Lyons et al., 1998; Lang et al., 2006).

3. Conclusions

Given the highlighted spatial patterns but also the previously discussed laboratory results, this paper highlights a possible role played by the NH_4^+ on the graupel charging which could explain to some degree the enhanced +CG presence over the GP region. This tentative hypothesis **does not refute or cast doubt to the overwhelming evidence that identifies the storm microphysical (e.g., Emersic and Saunders 2010 and references therein) or dynamical (e.g., W05 and references therein) processes as the major controlling factors of the CG polarity**. Conversely, this study merely questions whether the above speculations could be viewed as an additional mechanism that might have effect on the +CG presence over the GP region. Recently Albrecht et al., (2011) documented evidence that could be viewed as supportive of the speculations herein. These authors observed enhanced

+CG presence over deforested pastures over Rondonia (Brazil), a region that exhibits elevated NH_4+ concentrations from livestock and agricultural activity (Trebs et al., 2006). Nevertheless, in Albrecht et al. (2011) the authors speculate that the dominant mechanism responsible for the enhanced +CG is also in agreement with W05 and Carey and Buffalo (2007).

Currently the author is in search for additional evidence over other regions world-wide (e.g., Kitagawa and Michimoto, 1994; Chronis 2012), although the joint availability of CG polarity as well as NH_4^+ concentrations in rainwater could be restrictive. The author also acknowledges that the extrapolation of laboratory results into the "real" atmosphere requires more extensive research, thereby no conclusions can be drawn as to the degree, spatial or temporal scales at which the NH_4^+ charging mechanism might have an affect on the CG polarity. For instance, Jayarante et al., (1983) and Prakash and Kumar (2010) show a dependence on concentration of NH_4^+ on the graupel charge acquisition (in Normality units) thereby direct comparisons with the ground concentrations of Fig. 2 cannot be made at this moment (personal communication with R. Jayarante). Additional uncertainties stem from the level at which soluble impurities (such as NH_4^+) can compete the control exerted by the saturation of water/ice and the growth speed on the graupel charging processes (see Emersic and Saunders, 2010). Moreover, the speculated mechanism involving NH_4^+

cannot support the enhanced +CG presence over the US west coast (Orville and Huffines, 2001; Ely and Orville, 2005; Orville et al., 2011, also shown in Fig. 1), although Jungwirth et al., (2005) argues that other common soluble aerosol particles over oceanic or coastal environments may also have a similar effect on the +CG production, an argument also recently made by Cooray et al., (2013).

There are additional reasons as to why the hypothesis herein may be proven challenging to test; The most important is the CG year-to-year variation due to storm activity that can overshadow the postulated mechanism (see Koshak et al., 2014). As a result, the comparison between the NH_4+ concentrations and +CG temporal variation may not be adequate to fully corroborate or refute the above speculations. The examination of the similarities and dissimilarities between individual storms (e.g., in terms of thermodynamics or radar observations) over regions of high versus low NH_4^+ concentrations could provide a more complete answer nevertheless such task requires elaborate and extensive data analysis which, at present lies outside the scope of this study.

For the curious minds, in spite of the considerable uncertainties, the previous tentative arguments deserve some consideration. As discussed in Lyons et al., (1998), Kreidenweis et al., (2001) and Lang et al., (2006), we presently know that aerosol chemistry (e.g., pertinent to wild-fires) may be playing a very diverse role (personal communication with Walter Lyons). In this context, the critical question to be answered is whether the aerosol chemistry could effectively modulate thunderstorm charging attributes, independently from the well-established relationships with environmental factors (e.g., liquid water content, temperature etc.). Jayarante (2003)

indicates that, "*The role played by chemical impurities in determining the sign and magnitude of the charge separated during rebounding collisions between ice particles remains a major mystery and so far it has not been possible to explain it in terms of any of the existing theories*". Possible corroboration of the postulated mechanism by further in-depth analysis will break new ground in atmospheric science research and will establish a novel linkage between multidisciplinary science fields.

Acknowledgements

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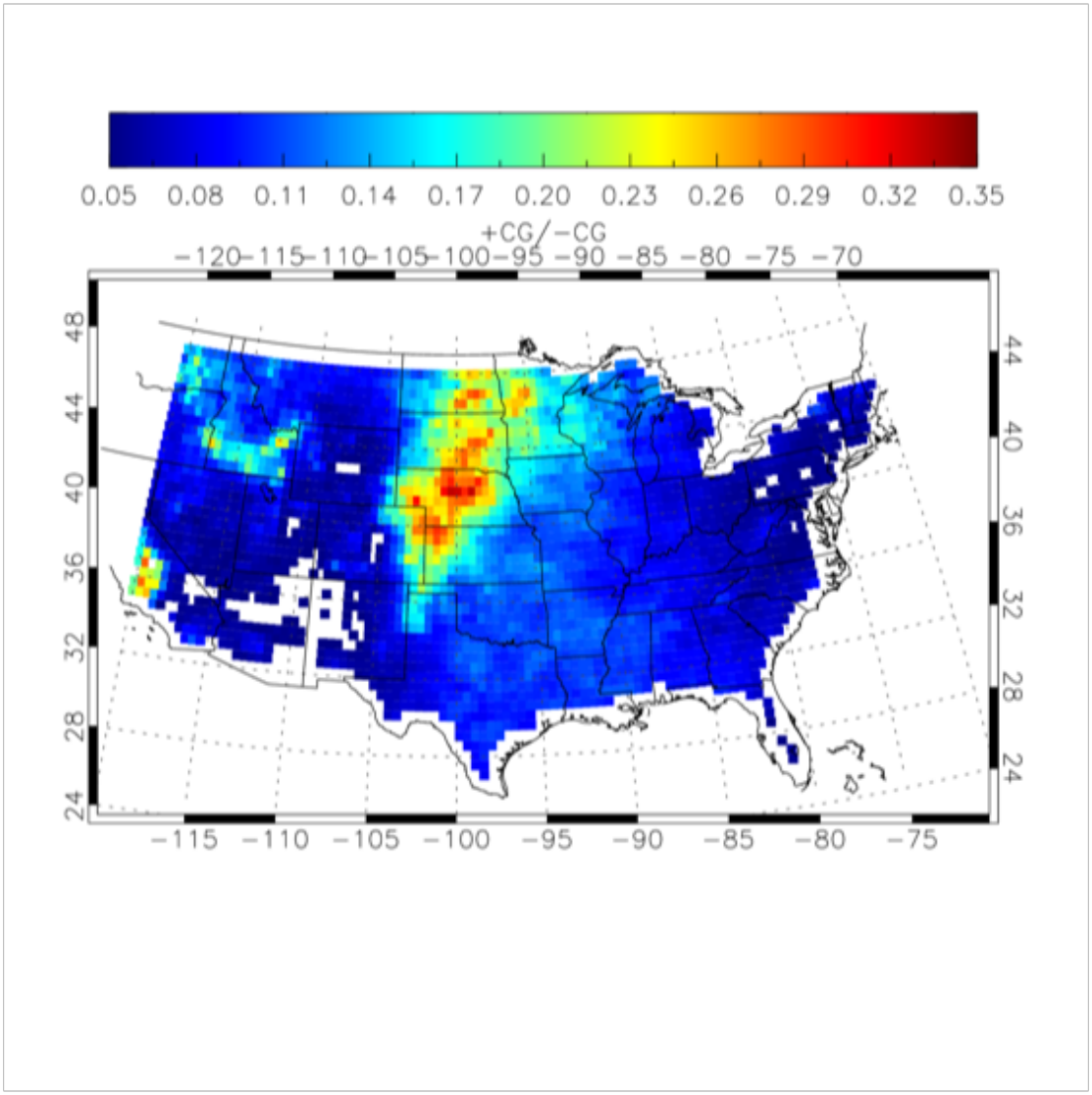
Figures Captions

Figure 1: The National Lightning Detection Network (NLDN) +CG/-CG ratio. The map is drawn on a ~0.5 degrees rectangular grid. The ratio is computed for each grid-box from on the counts of CGs flashes (both polarities) that were observed form 2003-2010.

Figure 2: Average (2003-2010) NH_4^+ precipitation concentrations (mg L^{-1}) from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) Crosses indicate the NADP/NTN sampling stations around the GP region. Data were initially acquired in GeoTiff format in Albers projection of ~3 km resolution. The annual composite precipitation surfaces are derived from an adapted version of a high resolution precipitation model developed by the PRISM Climate Group, and supplemented with NADP precipitation observations (see http://nadp.sws.uiuc.edu/data/mapProcess.aspx). The weighted values are calculated using a combination of the PRISM modeled precipitation data, and the NADP observed precipitation values (crosses). The weighting function was established so that as one approaches the edge of the 30 km radius the values of the weighted grid cells approached that of PRISM. Outside of the 30 km radius the annual precipitation grid cells were populated using only PRISM data.

Figures


0.05 0.08 0.11 0.14 0.17 0.20 0.23 0.26 0.29 0.32 0.35
+CG/−CG
−120 −115 −110 −105 −100 −95 −90 −85 −80 −75 −70
48 44 40 36 32 28 24
44 40 36 32 28 24
−115 −110 −105 −100 −95 −90 −85 −80 −75

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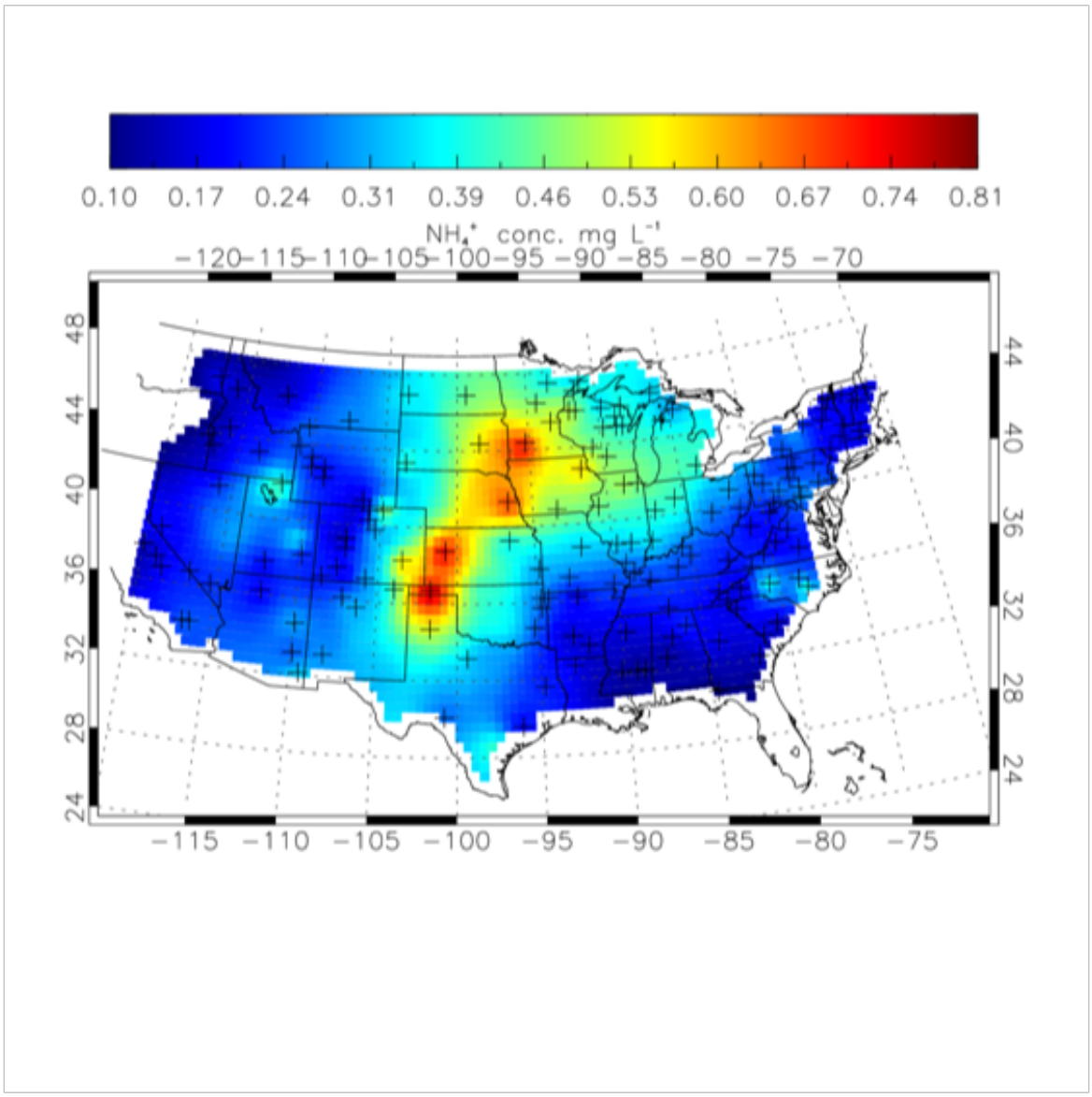
0.10 0.17 0.24 0.31 0.39 0.46 0.53 0.60 0.67 0.74 0.81
NH4+ conc. mg L-1
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48 44 40 36 32 28 24
44 40 36 32 28 24
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