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On the contribution of lightning to ultrafine aerosol formation

Fangqun Yu[1] and Richard P. Turco

Department of Atmospheric Sciences, University of California at Los Angeles

Abstract. We propose that locally high concentrations of new ultrafine aerosols may be generated within lightning channels, in which the air is initially highly ionized by discharge currents. In the channel, temperatures are sufficiently elevated to ensure that preexisting condensed materials are vaporized, generating aerosol precursors at concentrations much higher than would be found in ambient air. As the channel expands and cools, ions that survive neutralization attract condensable vapors and grow rapidly. Detailed microphysical simulations indicate that ultrafine particles (>3 nm in diameter) may achieve concentrations of 10^5-10^6/cm^3 in the channel within 10 seconds after a flash, leading to an effective "production index" of 10^{13}–10^{14} particles per meter of channel. While lightning may generate very high local concentrations of ultrafine particles, and occur worldwide, the overall contribution to global aerosol production is estimated to be small.

Introduction

The climatic, health, and chemical effects of aerosols are sensitive to particle size and concentration, which are strongly influenced by formation and growth processes. Atmospheric aerosols are also quite heterogeneous in nature, and it is important to identify their natural and anthropogenic origins. In this paper, we characterize a potentially important local source of ultrafine volatile aerosols associated with lightning.

The air in a lightning channel is heated and highly ionized due to the intense discharge current. As a result, the concentrations of aerosol precursors are expected to be high due to the vaporization of preexisting condensed materials (e.g., sulfates, ammonium, and organics). Moreover, as the air in the channel cools, residual ions can be activated and grow rapidly by attracting the vaporized condensates. The process we envision is quite similar to the formation of ultrafine particles on chemiions in supersaturated aircraft plumes, which has been studied extensively through modeling and field sampling [e.g., *Yu and Turco*, 1997; *Kärcher et al.*, 1998; *Yu et al.*, 1999; *Arnold et al.*, 1999; *Wohlfrom et al.*, 2000]. The detailed physical mechanisms leading from ionization to ultrafine particles are discussed elsewhere [e.g., see *Yu and Turco* 1998, 2000a, b], and a similar computational approach is employed here.

[1] Now at Atmospheric Sciences Research Center, State University of New York, Albany, New York.

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The initial channel

A lightning channel reaches a peak temperature of about 28,000-31,000 K within the first 10-20 μs after the return stroke, when the air in the channel is totally ionized [e.g., *MacGorman and Rust*, 1998]. The initial radius of the channel is only a few centimeters. Within the first 0.05 s after the stroke, however, the radius of the hot core has increased to ~1 m, driven by thermal expansion and shock-wave heating of the surrounding air [*Coppens et al.*, 1998]. Subsequently, due to strong ascending and descending motions and instabilities within the storm cell, intense turbulence—dominated by eddies of a meter and larger—is effective in dissipating the channel. Mixing with ambient air is primarily responsible for cooling the channel (radiative cooling is negligible), with temperatures falling to ambient levels in ~1 s [*Hill et al*, 1980]. The parameterization of these turbulent mixing and cooling processes is discussed in the next section.

Based on an average energy deposition per flash of 6.7×10^9 J and flash length of 7 km [*Price et al.*, 1997], the mean temperature of the initial stabilized lightning channel may be estimated. Assuming a channel radius of ~1.35 m, in the boundary layer the temperature would be ~525 K at 0.05 s. The simulations discussed below are initialized for these conditions. We also assume that preexisting condensed sulfate and ammonium are fully vaporized in the channel. The ambient sulfate mass concentration (SMC) is used to characterize the initial sulfuric acid vapor abundance in the channel.

The SMC exhibits large spatial and temporal variations, with maximum mass concentrations typically appearing in the summer season. Based on data compiled by *Adams et al.* [1999], the highest annual-mean SMCs occur in the industrialized areas of central Europe, the eastern United States, and eastern Asia, with values in range of ~4–12 μg/m^3 in the boundary layer. Most areas of Europe, North America, and Asia have average annual SMCs lying between 1.2–4 μg/m^3. Marine boundary layer SMCs generally exceed 0.4 μg/m^3 in the Northern Hemisphere, but are lower in the South Hemisphere. Sulfate mixing ratios are fairly uniform with altitude through the boundary layer, but decrease in the free troposphere, with average mixing ratios at the tropopause ~25% of those in the surface layer [*Adams et al.*, 1999]. Since lightning activity is more intense over land than over the oceans, and during northern-hemisphere summer than winter [*Backer et al.*, 1999], higher sulfate mass loading may be roughly correlated with lightning frequency. Here, we focus on cloud-to-ground lightning and choose a baseline SMC value of 4 μg/m^3. Sensitivity studies using SMCs ranging from 0.6–12 μg/m^3 are carried out to explore the dependence of aerosol formation on this parameter.

While ambient aerosols are vaporized in the initial channel, they are later mixed into the expanding channel. These aerosols provide a competitive sink for the condensing vapors as well as newly formed molecular clusters. In this work, the baseline ambient aerosol is initialized as a log-normal size distribution with a mean diameter of 0.25 μm and a standard deviation of 1.6. The

number concentration of these particles is then normalized to the assumed SMC, with a water content corresponding to equilibrium conditions at the local relative humidity. The background ion concentrations (positive, or negative) are taken to be 1000/cm^3, and the sulfuric acid vapor concentration, 5x10^6/cm^3. For the cases investigated here, the ambient temperature and relative humidity are fixed at 273 K and 90%, respectively, although the results are not highly sensitive to these values.

In the heated channel, the initial ion concentration (that is, at t=0.05 s) is assumed to be 10^8/cm^3. This value is consistent with an ion-ion recombination coefficient of ~2x10^{-7} cm^3/s estimated for a temperature of 523 K at 1 atmosphere pressure. The ions in the channel continue to recombine as the channel expands and cools (corresponding to the growth and coagulation of the ion clusters, referring to the treatment of *Yu and Turco*, 2000b).

Dispersion and microphysics

The evolution of charged and neutral particulate species in the lightning channel is governed by the following equation:

$$\frac{dn_i}{dt} = \left(\frac{\partial n_i}{\partial t}\right)_{mphys} + \left(\frac{\partial n_i}{\partial t}\right)_{mix} = \left(\frac{\partial n_i}{\partial t}\right)_{mphys} - \frac{(n_i - n_i^{amb})}{V_i}\frac{dV_i}{dt} \quad (1)$$

where $n_i = N_i/V_i$ is the concentration of species i, and N_i is number of species i in a volume V_i, defined by a cross section of the channel of unit length. Here n_i^{amb} is the corresponding ambient concentration of species i. The first term on the right-hand side of equation (1) describes the instantaneous microphysical processing of the aerosols within a fixed unit volume of the channel, while the second term defines the effects of mixing on the aerosol concentration in the channel (taking the aerosol as an inactive tracer).

The numerous species generated by a lightning stroke initially occupy a very small cross sectional area (the channel diameter being measured in centimeters). For our purposes, the channel may be considered as an infinite line source, with subsequent mixing and expansion described by Gaussian plume theory. Hence, time variations in the channel width can be expressed in terms of the variance, σ, of the distribution of a passive tracer initially confined to the channel. The variance is essentially the mean width of the tracer concentration profile across the expanding channel. We parameterize the variance as follows:

$$\sigma^2 = \sigma_0^2 + c_1 \frac{T - T_{amb}}{T_{amb}} \sigma_0^2 \ (t - 0.05) + c_2 \ t^p \quad (t \text{ in sec}) \quad (2)$$

where σ_0 is the effective width of the plume at t_0=0.05 s, and T_{amb} is the ambient temperature. The third term in equation (2) describes the power-law regime for turbulent dispersion [e.g., *Gifford*, 1977, 1995]. Based on observations and analysis presented by *Gifford* [1977, 1995], we choose c_2=0.375 (t in seconds) and p=2.2. This term, however, is negligible for $t \leq 1$ sec. The second term in equation (2) takes into account mixing due to buoyancy, and is proportional to the temperature difference between the channel and the local environment. Since, at t=0.05 s, the air in the channel has a relatively high temperature (525 K), buoyant forces initially destabilize and mix the channel. The baseline value of c_1 is chosen to be 20, which allows the channel temperature to approach ambient (within a few tens of degrees) in ~1 s, as suggested by *Hill et al.* [1980]. The sensitivity of our results to the value of c_1 is discussed below.

The initial lightning channel (that is, at 0.05 s) contains very high concentrations of ions and precursor gases. However, the temperature is also greatly elevated, which precludes aerosol nucleation. As the channel mixes and cools, ions are continuously neutralized in response to a temperature-dependent recombination coefficient. The onset of ion nucleation occurs after the air mass has cooled to a temperature at which the precursor gases are sufficiently supersaturated. In this study, we assume H_2SO_4 and H_2O as the primary aerosol precursors. However, any NH_3 vapor in the channel might act to stabilize the embryonic H_2SO_4-H_2O clusters, leading to somewhat earlier nucleation without greatly affecting the growth rates of the particles. We account for the effect of NH_3 on the H_2SO_4 supersaturation using an empirical model for the H_2SO_4-H_2O-NH_3 ternary system [*Korhonen et al.*, 1999; *Kulmala et al.*, 2000].

To investigate the suite of interactions between neutral and charged clusters and particles in a lightning channel, as well as coupling to the vapor phase, we apply an advanced particle microphysics (APM) code [*Yu and Turco*, 1998, 2000b; *Yu et al.*, 1999]. The APM model tracks the compositions and size distributions of different particle types, and treats the entire course of aerosol nucleation and growth as a unified collisional (kinetic) mechanism. A more detailed description of the kinetic approach and the APM model can be found in the publications cited.

Simulations

Figure 1 illustrates the predicted evolution of the concentrations of total ultrafine particles having diameter $d>d_i$ (d_i =3, 10, and 120 nm) as well as the total ion concentration in a lightning channel for the baseline case (i.e., SMC=4.0 μg/m^3; c_1=20). Particles larger than 10 nm are dominated by ambient aerosols, which are mixed into the channel (note that the curves for $n_{d>10\ nm}$ and $n_{d>120\ nm}$ closely overlap). The simulations suggest that new ultrafine particles (d>3 nm) begin to appear at t~0.7 s, with the number concentration increasing sharply between 1 s and 4 s. $n_{d>3\ nm}$, reaches a maximum concentration of 4x10^5/cm^3 at t=5 s (when the channel has a radius of ~ 6.5m), and decreases rapidly thereafter as a result of dilution. The ion concentration initially falls off primarily due to ion-ion recombination, and approaches a value of ~ 10^7/cm^3 at the onset of ion nucleation. Accordingly, our simulated results are not very sensitive to the initial ion concentration

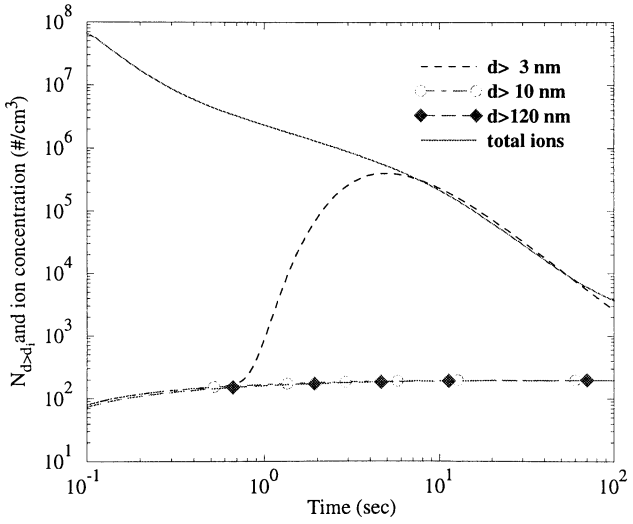

Figure 1. Simulated changes in the cumulative aerosol number concentration, $n_{d>di}$ (d_i = 3, 10, 120 nm), and total ion concentration, in a lightning channel for the baseline case (i.e., SMC=4.0 μg/m^3, c_1=20).

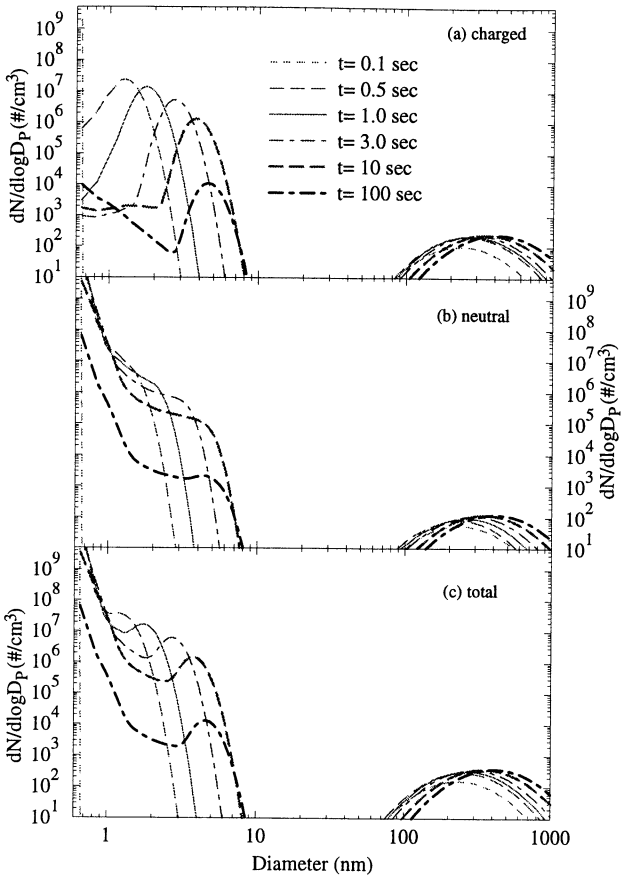

Figure 2. Calculated evolution of the size distributions of: (a) total charged particles (positive plus negative); (b) neutral particles; and (c) total particles (charged plus neutral). The results correspond to the case shown in Figure 1.

for values > 10^7/cm^3. At t=100 s, when the channel has expanded to a radius of about 100 m, most of the newly formed ultrafine particles remain smaller than 10 nm, with a concentration of the order of several thousand per cubic centimeter. By comparison, if SMC=8.0 μg/m^3, $n_{d>3\ nm}$ reaches a maximum concentration of 1.7x10^6/cm^3 at t=2.5 s (when the channel has a radius of ~5 m).

Figure 2 shows, for the simulation depicted in Figure 1, the evolution of the size distributions of the different charged and neutral aerosol species in the channel. In the APM model, the initial, as well as any newly generated or entrained, H_2SO_4 molecules, positive ions, and negative ions, are always inserted into the first size bin of the neutral, positively charged, and negatively-charged particle types, respectively. The size-, charge-, and temperature-dependent kinetic interaction rates [e.g., *Yu and Turco*, 2000b] determine the buildup of charged and neutral clusters and their growth into stable nanoparticles.

Clearly depicted in all of the panels in Figure 2 are the activation and growth of ions into nanoparticles, which occur as soon as a modest supersaturation appears in the channel. At 0.1 s, the temperature is still too high for stable clusters to form. By 0.5 s, however, most of ions that survived recombination and scavenging are activated due to the high concentration of sulfuric acid vapor (~6x10^9/cm^3 at t=0.5 s). These large clusters, which lie below 3 nm at this time, achieve a mean size of ~4 nm by 10 s. Most of the resulting "nucleated" nanoparticles were initially electrically charged, thus having the advantage of accelerated growth due to electrostatic effects. The neutral clusters, by contrast, tend to grow more slowly under the conditions in the channel, and few reach observable sizes. The mode of neutral ultrafine particles seen in panel (b) of Figure 2 is the result of neutralization of charged nanoparticles by entrained small ions, and recombination of oppositely charged clusters and nanoparticles. After 10 s, the concentrations of ultrafine particles in all three panels of Figure 2 decrease rapidly, owing to efficient turbulent dilution of the channel. In panel (a), the charged clusters that appear at very small sizes (<1-2 nm) by t=100 s are associated with small ions either entrained from the ambient environment or generated in situ by galactic cosmic rays (where an ionization rate of 2 ion-pairs/cm^3-s is assumed; *Yu and Turco*, 2000b). The predicted slow growth of entrained background aerosols (the largest size mode in Figure 2) is partly due to the collection of precursor gases and small molecular clusters, and partly due to the increase in relative humidity as the air mass in the lightning channel cools.

It is clear from Figures 1 and 2 that, in a lightning channel, ultrafine particles are formed at very high concentrations within the first 10 seconds. It is important, however, to estimate the total particle source strength associated with a lightning stroke. Accordingly, we define the production index (*PI*) as,

$$PI_{d>d_i} = A \times (n_{d>d_i} - n^{amb}_{d>d_i}) \quad (3)$$

where A is the cross sectional area of the lightning channel, and $n_{d>d_i}$ and $n^{amb}_{d>d_i}$ are the number concentrations of particles larger than d_i inside and outside of the channel, respectively. The *PI* gives the apparent number of ultrafine particles that are formed per unit length of the lightning channel (i.e., number/m) at any given time. Note in Figure 2 that, beyond 10 s, the ultrafine particle concentration falls off mainly because of dilution; the total number of particles originating in the channel (*PI*) remains relatively invariant beyond this time (see below).

Figure 3 illustrates the time evolution of $PI_{d>3nm}$ for different ambient sulfate mass concentrations (SMC ranging from 0.6–12 μg/m^3), with the baseline dispersion parameters (and, hence, temperature and cross sectional area). The ultrafine particles (d>3 nm) appear sooner with higher SMC values since the channel becomes supersaturated earlier, resulting in a faster growth rate. With the baseline SMC, $PI_{d>3\ nm}$ achieves an asymptotic value of ~7x10^{13}/m. It is not surprising that higher ambient sulfate mass

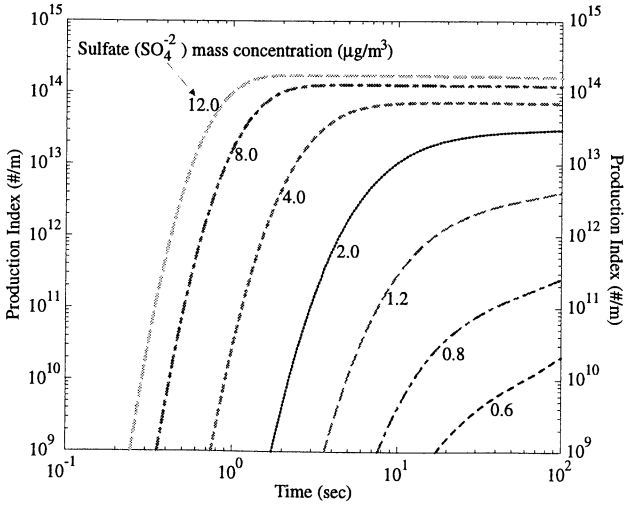

Figure 3. Time histories of the apparent production index for particles larger than 3 nm (i.e., $PI_{d>3nm}$) for different ambient sulfate mass concentrations (from 0.6 μg/m^3 to 12.0 μg/m^3).

concentrations lead to enhanced aerosol production. However, at the largest SMCs, $PI_{d>3\ nm}$ is limited by the availability of ions surviving recombination. For sulfate mass loadings less than ~4 $\mu g/m^3$, $PI_{d>3\ nm}$ decreases quickly; e.g., by more than a factor of ten as SMC is reduced from 4 to 1.2 $\mu g/m^3$.

Sensitivity studies were also carried out to investigate the effect on $PI_{d>3\ nm}$ of the initial mixing rate of the channel (i.e., c_1 in equation 2) The simulations indicate that, while c_1 affects the timing of the nucleation event and the growth rate of the particles, $PI_{d>3\ nm}$ at t=100 s is effectively insensitive to its value. For example, by varying c_1 between 1–100 (with other baseline parameters fixed), $PI_{d>3\ nm}$ changes only from $3.5x10^{13}$/m to $8x10^{13}$/m. This low sensitivity is mainly a result of the dependence of the expansion rate of the channel on the temperature contrast (equation 2). Faster mixing acts to reduce the temperature gradient more quickly, thereby damping the buoyant forces such that the integrated effect over time is similar regardless of the value of c_1.

Summary and Discussion

The detailed evolution of charged and neutral molecular clusters, and the creation of nanoparticles following a lightning discharge, are simulated using an advanced particle microphysics (APM) model. The air in a lightning channel contains high concentrations of ions and aerosol precursor gases, owing to ionization by the intense discharge current and vaporization of preexisting particles by extreme heating. As the air in the channel cools, the ions that survive recombination collect condensable vapors and grow very rapidly. The processes that control the mixing of the channel with ambient air have been considered, and the dependence of lightning-generated particle properties on ambient sulfate mass concentrations (SMCs) has been investigated.

We predict ultrafine (d>3 nm) particle abundances as high as 10^5-$10^6/cm^3$ in a lightning channel within 10 seconds after a flash for typical sulfate mass loadings (SMCs of 1.5–8 $\mu g/m^3$). The high concentrations of both ions and precursor gases that exist in the channel are essential in generating these particles. By 100 s following the flash, when the channel has expanded to a radius of roughly 100 m, the concentration of newly formed ultrafine particles still approaches several thousand or more per cc. Considering the multi-stroke and multi-branch aspects of common cloud-to-ground (CG) lightning flashes, such events are likely to generate high concentrations of ultrafine particles locally within and above the boundary layer.

The production index (*PI*) of lightning-generated ultrafine particles (d>3 nm) is in the range of 10^{13}–10^{14} particles per meter of lightning channel for typical values of SMC (1.5–8 $\mu g/m^3$). Assuming a global CG flash rate of 20-30 flashes/s and a mean CG channel length of 7 km [*Price et al.*, 1997], we estimate that $1.4x10^{18}$–$2.1x10^{19}$ ultrafine particles are generated each second worldwide. Assuming that these particles are evenly distributed within the first 5 km of the atmosphere, an average global production rate of ~$5x10^{-7}$–$8x10^{-6}/cm^3s$ is found. This rate is very small compared with other apparent sources of aerosols observed in the lower atmosphere [e.g., *Yu and Turco*, 2000a].

While this study has focused on ultrafine particle formation in cloud-to-ground lightning, similar processes are certain to accompany intra-cloud lightning, which is more common. Accordingly, locally high concentrations of ultrafine particles might be expected in certain outflow regions, or inside convective cells experiencing strong electrical activity. One line of evidence suggesting such a connection is a high correlation between elevated CN concentrations and high NO/NO_y ratios in plumes associated with convective systems [*Wang et al.*, 2000]. In the present work, we have assumed that sulfuric acid evaporated from existing sulfate aerosols is the key precursor leading to ion activation and growth in a lightning channel. The potential influence of NH_3 in lowering the H_2SO_4 saturation vapor pressure is also considered. In reality, other chemical processes (for example, the oxidation of SO_2 in the channel) might conceivably contribute to aerosol production. Further, species such as HNO_3, and organics, may play a role, especially with low background sulfate levels. Nevertheless, despite the uncertainties cited above, we do not expect lightning discharges to represent a primary global source of new tropospheric aerosols.

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R. P. Turco, Department of Atmospheric Sciences, University of California, Los Angeles, CA 90095-1565. (turco@atmos.ucla.edu)

F. Yu, Atmospheric Sciences Research Center, State University of New York at Albany, 251 Fuller Road, Albany, New York 12203. (yfq@asrc.cestm.albany.edu)

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