Evolution of aircraft-generated volatile particles in the far wake regime: Potential contributions to ambient CCN/IN

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Abstract. We apply a detailed aerosol microphysics model to study the evolution of aerosols in the far wake of an aircraft plume. Constrained by in-situ measurements, our simulations reveal that the largest volatile particles—those most likely to contribute to the background abundance of condensation nuclei—are dominated by ion-mode aerosols formed on chemiions. By tracking the evolution of the aircraft-generated particles from the near wake to the far wake, we estimate the properties of the aircraft particles that are effectively injected into the upper troposphere. The entrainment of ambient vapors and aerosols into the aging plume is taken into account, which significantly affects the outcome. The model calculations reveal that a substantial number of the larger ion-mode volatile particles may survive long enough to act as potential cloud nuclei, thus perturbing the background CCN population. This perturbation is significant during the winter season, and in other locales with low background aerosol concentrations, but is likely to be negligible during the summer season or at locations with heavy aerosol loading. The implications and uncertainties of the results are discussed.

1. Introduction

The large quantities of volatile ultrafine particles formed in aircraft wakes may cause potentially significant climatic impacts [*Friedl*, 1997]. However, the uncertainties regarding the numbers and properties of these aerosols remain large. In particular, a clear understanding of the mechanisms that might create these volatile particles in aircraft plumes, and the tendency of these particles to act as cloud condensation nuclei (CCN) or ice nuclei (IN), has not yet been attained.

Constrained by in-situ measurements, recent detailed numerical studies of plume microphysics reveal that chemiions generated by engine combustors play a crucial role in the formation and evolution of the detectable volatile particles in near-field wakes [*Yu and Turco*, 1997, 1998a,b; *Yu et al.*, 1998, 1999; *Kärcher et al.*, 1998]. This concept is supported by recent ground-based measurements, which have detected large concentrations of heavy negative ions in jet engine exhaust [*Arnold et al.*, 1998]. While the newly formed volatile aerosols in an aircraft plume can have very high concentrations and total surface areas, which may influence heterogeneous chemical processes locally, these particles are typically much too small (<10 nm) to act as cloud nuclei (~ 80 ·nm).

However, if even a modest fraction of these abundant volatile particles were to grow to sizes sufficient to act as CCN or IN, they could have important climatic implications. Self-coagulation and scavenging by preexisting ambient aerosols affect the potential impacts by limiting the total number of aged plume particles [*Turco and Yu*, 1997]. Accordingly, it is critical to understand the fate of aircraft-generated volatile particles as the wake dissipates. In this work, we carry out a detailed numerical study of the evolution of aircraft-generated particles in an aged plume (for times up to 10 days) under typical ambient conditions in the upper troposphere.

2. Mixing of aircraft exhaust with ambient species

2.1. Physical dilution and entrainment

To simulate the combined effects of microphysics and dilution/entrainment on aircraft particulates, we utilize a "trajectory" version of our plume microphysics code [*Yu and Turco*, 1997, 1998b]. In this case, the exhaust is treated as a perturbed parcel of air subject to dilution through mixing with the ambient environment as prescribed along a trajectory confined to the wake; other parameters, such as solar intensity and trace gas concentrations, may be specified as variables along the trajectory. The dilution of an aircraft plume is most simply represented as an increase in the cross-sectional area of the plume, presumably due to the entrainment of ambient air into the plume by various mixing processes. Typically, all of the constituents in the plume are assumed to be uniformly mixed instantaneously across the entire cross section. While this approximation is obviously crude, it may be rationalized as representing the bulk mean properties of the plume.

The concentration of a vapor or particulate species, C, within the plume is affected by entrainment according to,

$$\frac{dC}{dt} = -\frac{1}{A}\frac{dA}{dt}(C - C_{amb}) \qquad (1)$$

where A is the plume cross-sectional area, and C_{amb} is the species' ambient concentration. Equation (1) allows the dilution of emitted constituents, the entrainment of ambient species, and the overall mass balance for each constituent to be calculated explicitly. Here, the plume dilution ratio (and hence the cross-sectional area) is calculated from the equation formulated by *Schumann et al.* [1998].

2.2. Species variations in the plume

The plume species considered in this study are: volatile aerosols (assumed to be composed of sulfuric acid aqueous solution), nonvolatile particles (soot coated with sulfuric acid), entrained ambient particles, and gaseous H_2SO_4, SO_2, and

H_2O. The size distributions of the plume volatile particles has a distinct bimodal structure, in which there is a distinct ion mode constituting the larger "charge-activated" volatile sulfuric acid particles, and a neutral mode consisting of the residual slowly-growing molecular clusters formed in the highly supersaturated region of the plume (Yu and Turco, 1997).

Gaseous Species: The sources of H_2SO_4 and SO_2 in the plume include: primary emissions of these vapors, secondary chemical production, and entrainment of background material. To calculate the entrained component of sulfuric acid vapor in the plume, we define the diurnally-varying ambient abundance in the upper troposphere as,

$$[H_2SO_4]_{amb} = [H_2SO_4]_{noon} \times Max\left\{0, \sin\left[\frac{\pi}{12}\left(\frac{t}{3600}+t_0-6\right)\right]\right\}$$

(2)

where $[H_2SO_4]_{amb}$ has a maximum noontime concentration of $[H_2SO_4]_{noon}=5\times10^6/cm^3$ [*Arnold et al.*, 1997]. In Eq. (2), t (s) is the age of plume since the time of formation, t_0 (hr) (in this study the plume is assumed to form at 8:00 am, local time; i.e., $t_0=8$).

The most significant continuing source of H_2SO_4 in the aging plume is due to the oxidation of SO_2. The amount of sulfur dioxide emitted by the aircraft is determined by the fuel sulfur content, minus the primary sulfuric acid emissions. The entrained component of SO_2 is calculated here using Eq. (1) with an assumed baseline upper-tropospheric background concentration of 100 pptv [*Arnold et al.*, 1997].

The SO_2 in the plume is oxidized over time by its reaction with *OH* (a rate coefficient of 1.0×10^{-12} cm^3/s is assumed in this study). To simplify the chemical aspects of the problem, and avoid extensive photochemical computations, the *OH* concentration is simply parameterized as,

$$[OH] = [OH]_{max} \times Max\left\{0, \sin\left[\frac{\pi}{12}\left(\frac{t}{3600}+t_0-6\right)\right]\right\}$$

(3)

where the maximum daytime *OH* concentration, $[OH]_{max}$, is taken to be $3.0\times10^6/cm^3$ [*Crutzen and Zimmerman*, 1991] in the baseline simulations. Hence, there is a continuing source of sulfuric acid vapor for condensation in the plume.

Ambient Aerosols: We initialize the background aerosols to either the mean summer (case S) or winter conditions (case W) using the size distributions defined in Table 1, which are based on more than two decades of balloon-borne soundings at Laramie, Wyoming [*Hofmann*, 1993]. Note that these data may be somewhat biased, having been collected at a continental site, where lofting of boundary layer material can occur. In

Table 1. Mean size distributions of the background aerosols in upper troposphere adopted in this study; the bimodal lognormal representation is taken from *Hofmann* [1993].

Case	Ambient modes	Mode number n_i (#/cm^3)	Mean radius r_i (μm)	Standard deviation σ_i
Summer:	1	800	0.02	2.10
S	2	0.2	0.4	1.79
Winter:	1	400	0.01	2.40
W	2	0.006	0.38	1.88

the simulations, following initialization, ambient particles entrained into the plume (as prescribed by Eq. 1), as well as those in the background environment, continue to grow due to condensation and coagulation processes. To keep track of the background particle evolution, and hence the properties of the entrained aerosols over time, we simultaneously model two parallel parcel trajectories—one inside and one outside of the aircraft wake. For the simulations discussed in section 3, the surface area of the background aerosols increases from 12.7 μm^2/cm^3 to 18.5 μm^2/cm^3 after 10 days in case S, and from 2.4 μm^2/cm^3 to 5.7 μm^2/cm^3 after 10 days in case W.

3. Simulations and Results

To study the properties of aerosols in the far wake, we have extended our earlier simulations of near-wake microphysics [*Yu et al.*, 1998; *Kärcher et al.*, 1998]. The present simulations are initialized using concentrations of plume species (H_2SO_4, SO_2, H_2O, and size-resolved volatile, soot, and mixed aerosols) determined from detailed kinetic simulations of wake evolution during the first 100 s, including the effects of chemiions. The resulting near-wake composition depends, of course, on the assumed fuel sulfur content (FSC) and fraction (η) of fuel sulfur oxidized to $SO_3+H_2SO_4$ shortly after emission. In the present application, calculations of subsequent particle-particle and particle-vapor interactions are based on a "classical" (rather than "kinetic") microphysical treatment, which is appropriate for the time scales of interest (refer to *Yu and Turco*, 1997, 1998b).

At $t=100$ s, the plume aerosols exhibit two distinct modes—the ion mode, and a neutral mode consisting of extremely small homogeneously nucleated particles [*Yu and Turco*, 1997]. Since the purpose of this study is to investigate the fate of aircraft-generated volatile particles under typical upper tropospheric conditions, the possible formation mechanisms for aerosols in the background atmosphere are neglected. The quantitative contribution of aircraft-emitted organic species to the long-term growth of the plume aerosols is most likely negligible [*Yu et al.*, 1999] and is also neglected in this study.

As a baseline case, we assume $\eta=2\%$ for high sulfur fuel (HS, FSC=2700 ppmm), and $\eta=55\%$ for low sulfur fuel (LS, FSC=20 ppmm), which results in an ultrafine plume aerosol having properties consistent with CN measurements in near field [*Yu et al.*, 1998; *Kärcher et al.*, 1998]. It turns out that the latter value of η reflects, in a simplified manner, the potential role of organic materials when fuel sulfur levels are very low [*Yu et al.*, 1999]. It should be noted that the total amount of sulfur emitted under those circumstances is negligible in comparison to the quantity of entrained ambient sulfur, and particle growth in the far wake is not very sensitive to η in such cases.

Figure 1 shows the simulated evolution of "apparent" emission indices (*AEI*'s) corresponding to the high sulfur (HS) and low sulfur (LS) SULFUR-5 experiments for the summer baseline case (case S, Table 1). An *AEI* gives, for each kilogram of fuel burned, the total number of particles larger than a specific threshold size that would be present in the plume. The *AEI* is therefore equivalent to the number that would be measured by a particle (CN) counter with the same size cutoff (integrated across the plume cross section at the time considered). Accordingly, *AEI*'s are a convenient measure for estimating the po-
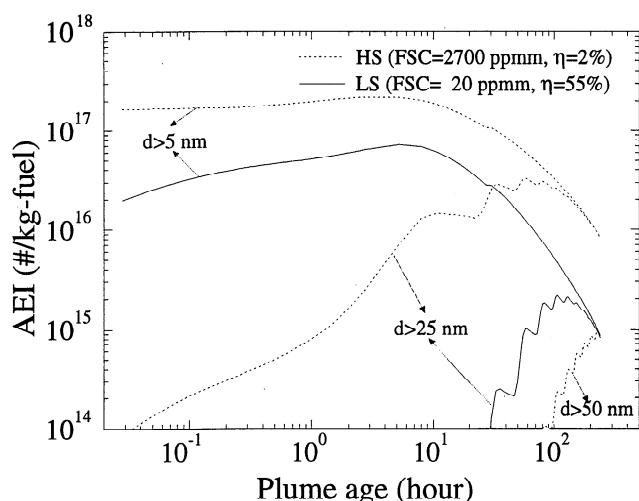
Figure 1. Calculated time evolution of the "apparent" emission indices (AEI) of aircraft-generated volatile particles for several assumed particle measurement threshold sizes, under summertime background aerosol conditions in the upper troposphere (case S in Table 1). Results are shown for both high sulfur and low sulfur fuel simulations.

tential aerosol perturbation associated with a fleet of aircraft. Note, however, that the AEI's corresponding to different particle sizes change as the plume evolves, which in itself provides information on the growth and coagulation of the aircraft-generated particles. The AEI for a specific threshold size initially increases with time, as fresh particles grow past the cutoff. Later, the AEI's decrease owing to coagulation scavenging by the background aerosols. A small diurnal variation in the AEI's (for cutoff sizes, d, exceeding the mean mode diameter, $\bar{d}$, of the evolving aerosols) is related to the diurnal variation of the H_2SO_4 in the plume.

During the day, the tendency in $AEI_{d>\bar{d}}$ is to decrease because of scavenging, and to increase because of condensation. At night, the AEI always decreases since condensational growth is essentially absent (and heterogeneous chemical processes that might lead to growth are assumed to be negligible). For Case S, as defined in Table 1, ambient aerosols quickly scavenge most of the aircraft-generated volatile particles. The fraction of volatile plume aerosols surviving at $t=10$ days is only ~10% with HS, and ~1% with LS (Fig. 1). The fact that the abundance of volatile aircraft particles with $d\geq25$ nm is roughly a factor of 10 lower after 10 days in the simulations with low sulfur fuel (LS versus HS in Fig. 1) supports an earlier suggestion that reductions in FSC, while not likely to reduce the total number of volatile particles emitted (in the near field), will decrease the size of ion-mode particles and hence their ability to form CCN [Yu et al., 1998]. Indeed, the relatively high background aerosol surface area concentration in summer (~15 $\mu m^2/cm^3$) inhibits the subsequent growth of the ion-mode plume particles. As a result, even after 10 days, only a very small number (<10^{13}/kg-fuel) of the initial ion-mode particles (~10^{17}/kg-fuel) have grown to sizes large enough to act as CCN (taking a minimum CCN diameter as ~80 nm). In fact, for the HS case, $AEI_{d>80\ nm} \simeq 9\times10^{12}$/kg-fuel at $t=10$ days, and with LS, $\simeq 5\times10^{10}$/kg-fuel.

The evolution of the AEI's for winter conditions (case W, Table 1) is illustrated in Figure 2. The aerosol surface area is a factor of 5 smaller than in summer, and the scavenging effi-

ciency of the background aerosols is greatly reduced. Accordingly, at 10 days, many more of the volatile ion-mode particles have survived, and they are much larger than under summertime conditions (compare Fig. 1). The winter AEI for CCN ($d\geq80$ nm) is ~1×10^{16}/kg-fuel with LS and 2×10^{16}/kg-fuel with HS, both values being greater by about one order of magnitude than the emission index for total soot particles. Importantly, the reduction in FSC between HS and LS does not effectively reduce the number of aircraft particles capable of acting as CCN in typical winter situations. The smaller concentration and surface area of the background aerosol in winter allow vapors to condense more effectively on the aircraft ion-mode particles, and at the same time reduce the scavenging of the plume aerosols. As a result, even the smallest ion-mode exhaust particles generated by low sulfur fuel have an opportunity to grow substantially prior to being scavenged. Note that, in all cases, most of the sulfur mass carried by the aircraft-generated volatile particles at 10 days is derived from H_2SO_4 and SO_2 entrained into the plume. In the HS case, the apparent increase of the $AEI_{d>5\ nm}$ within hours after emission is due to the growth of the neutral mode particles. Nevertheless, the largest particles that evolve in the plume are dominated by ion-mode aerosols.

If the total number of background particles is reduced by 50% in case W (with other parameters unchanged), which may represent relatively clean upper tropospheric air, then $AEI_{d>80nm}$ increases to 5.6×10^{16}/kg-fuel at 10 days for HS, and 4.0×10^{16}/kg-fuel for LS. This underscores the significant sensitivity of aircraft CCN production to the ambient aerosol burden. In "clean" air, a high yield of aircraft CCN is possible.

4. Summary and Discussion

In this study, we have focused on the microphysical evolution of aircraft plume volatile aerosols over periods of a week or longer. On this extended time scale the oxidation of SO_2 emissions, and the entrainment of ambient sulfur vapors (H_2SO_4 and SO_2) and particles are important. We have attempted to include in our analysis all of the key processes that affect the growth of plume aerosols in the upper atmosphere.

Simulations indicate that the fraction of aircraft-generated volatile particles evolving into potential CCN/IN depends
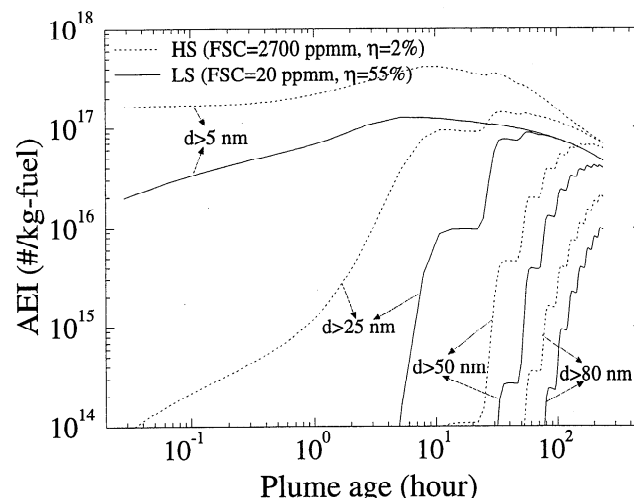

Figure 2. Same as Figure 1 except under wintertime background aerosol conditions (case W in Table 1).

strongly on the concentration and surface area density of the background aerosol. Under mean summer conditions [Hofmann, 1993], the lifetimes of ultrafine particles in the upper troposphere are quite short and very few ($<10^{13}$/kg-fuel) reach sizes large enough to be classified as CCN ($\geq$80 nm), even after 10 days. However, under winter conditions, which are cleaner [Hofmann, 1993], a significant number (~1-2×10^{16}/kg-fuel) of the aircraft particles may evolve into CCN. The effective emission index for these climatically significant particles (potential CCN/IN) can approach ~1-5×10^{16}/kg-fuel, which exceeds the soot emission index by roughly one order of magnitude.

Based on the calculated values of $AEI_{d>80\ nm}$, the likely perturbations of background CCN by aircraft emissions can be estimated for typical upper tropospheric states. Using 1992 aviation fuel consumption statistics (3.73×10^8 kg/day; e.g., Friedl, 1997) and assuming that 50% of the fuel is burned within the 9-12 km height interval (where cirrus clouds typically form), we estimate that flight emissions might enhance global upper-tropospheric CCN concentrations by <0.01/cm^3 during the summer season (i.e., case S in the text, and assuming a 10-day average residence time for the aircraft CCN). In the winter season (case W), the CCN perturbation may reach ~5/cm^3. Within the North Atlantic flight corridor the CCN enhancement could be 7 times larger than that determined as a global average [Friedl, 1997]. Accordingly, enhancements of as little as <0.07/cm^3 during the summer season and as great as ~35/cm^3 during winter season might be projected for the North Atlantic flight corridor. These estimates do not include a possible increase in the ambient CCN activity owing to the scavenging of excess aircraft exhaust sulfur by the background aerosol.

Measured background CCN concentrations (at 1% supersaturation) at ~10 km over Laramie, Wyoming are ~100/cm^3 during summer and ~40/cm^3 during winter [Delene et al., 1998]. Accordingly, the aircraft perturbations predicted here are likely to be significant during the winter season, as well as in other locales with low background aerosol concentrations, but not during the summer season or at locations with heavy aerosol loading. It is also noteworthy that, for relatively clean upper tropospheric conditions, reductions in fuel sulfur content may not effectively reduce the number of climatically significant particles injected into by aircraft.

Uncertainties in our results are associated with variations in the background concentrations of SO_2, OH, H_2SO_4, and aerosols, in the long-term course of aircraft plume mixing and dispersion, and in the contribution of other species to the growth and activity of CCN. The predictions already shown are clearly sensitive to the ambient aerosol concentration (compare cases S and W). In a series of sensitivity calculations associated with this work, we find that the mean size of plume volatile particles (hence, the AEI's) at 10 days are also quite sensitive to the rate of sulfuric acid vapor generation in the plume. For example, in case W, a reduction [SO_2] by half shifts the mean diameter of plume volatile particles (at day 10) from 68 nm to 49 nm, although the total number of these particles changes only marginally (from 4.6×10^{16}/kg-fuel to 4.5×10^{16}/kg-fuel). Additional sensitivity studies will be discussed elsewhere.

Contrail formation, and cirrus cloud processing, which are not considered in this study, can have a profound effect on the sizes and properties of CCN/IN in the upper atmosphere [Yu and Turco, 1998a]. A comprehensive assessment of aircraft impacts on background aerosol populations also requires a clear understanding of the origin of the background tropospheric aerosols [Brock et al., 1995; Turco et al., 1998]. To reduce the remaining uncertainties, more detailed far wake observations and simulations are clearly needed, as are more advanced cloud microphysical processing studies.

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