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Role of aircraft soot emissions in contrail formation

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[1] The susceptibility of microphysical properties of young contrails to changes in aircraft soot emissions is studied with a microphysical plume model. Liquid plume and ambient particles compete with exhaust soot particles for the formation of contrail ice particles, assuming that soot particles are activated into water droplets prior to homogeneous freezing. Soot controls ice formation in contrails for high number emission indices including the range of current global fleet values. A fivefold reduction of soot emissions from average levels of $5 \times 10^{14} - 10^{15}$ (kg-fuel)$^{-1}$ approximately halves the initial contrail visible optical depth. Further soot reduction reverses this trend at temperatures well below the formation threshold temperature unless emissions of sulfur and organics are cut substantially. Whether and to which degree reductions in soot emissions help mitigate the contrail climate impact depends on subsequent aircraft wake vortex processing of contrails and their development into contrail cirrus. **Citation:** Kärcher, B., and F. Yu (2009), Role of aircraft soot emissions in contrail formation, *Geophys. Res. Lett.*, *36*, L01804, doi:10.1029/2008GL036649.

1. Introduction

[2] The meteorological conditions describing the formation of aircraft contrails are determined by the moisture and heat budgets of aircraft exhaust plumes that mix with drier and colder ambient air. Thermodynamic considerations suffice to predict contrail formation threshold temperatures T_*, typically below 228 K at air pressures below 300 hPa in an ice-saturated atmosphere for modern jet aircraft [*Schumann*, 1996]. A detailed kinetic treatment of plume turbulence, microphysical and chemical properties of plume particles, and phase transitions is required to describe the number, size, and optical properties of ice particles forming in contrails [*Kärcher*, 1996].

[3] The environmental challenges of aviation operations are well recognized both within the atmospheric sciences [*Sausen et al.*, 2005] and aeronautics [*Green*, 2003] communities. As contrails substantially contribute to aviation-induced climate change [*Forster et al.*, 2007] and demand for air transport is expected to increase strongly [*International Civil Aviation Organization*, 2007], operational and technological options to mitigate contrail formation are under scrutiny, none of which appear to be sufficiently well evaluated as to their efficacy [*Noppel and Singh*, 2007; *Gierens et al.*, 2008]. Technological options to suppress contrail formation include modifications of jet engine emission properties.

[4] Two plume particle types, affected by emissions of water vapor (H_2O), charged molecular clusters (chemi-ions), and condensable sulfur and hydrocarbons (organics), serve as the main contrail ice particle precursors. Ultrafine liquid particles (mean diameters of few nm) mainly forming on chemi-ions in the plume prior to contrails [*Yu and Turco*, 1997; *Kärcher et al.*, 2000] and larger soot particles (mean sizes of few tens of nm) forming during kerosene fuel combustion [*Anderson et al.*, 1998; *Petzold et al.*, 1999]. The latter control contrail ice formation in threshold conditions [*Kärcher et al.*, 1995, 1996]. In the absence of contrails, aircraft soot particles may also trigger or modify cirrus cloudiness by acting as ice-forming nuclei away from the source aircraft; this subject is examined elsewhere [*Kärcher et al.*, 2007].

[5] The effects of ambient temperature and sulfur emissions on the properties of ice particles in the jet regime (up to ~5 s of plume age past emission) have been studied experimentally and theoretically [*Schumann et al.*, 1996, 2002; *Petzold et al.*, 1997; *Brown et al.*, 1997; *Yu and Turco*, 1998; *Schröder et al.*, 1998; *Kärcher et al.*, 1998a]. The susceptibility of contrail ice particle properties to variations in soot emissions has not yet been investigated systematically and cannot be inferred with confidence from measurements [*Schumann et al.*, 2002]. The present modeling work attempts to fill this gap and outlines implications for strategies to suppress contrail formation by changes in soot emissions.

2. Model Description

[6] The simulations have been carried out with a comprehensive parcel model of jet plume aerosol and ice microphysics [*Yu and Turco*, 1998]. It accounts for electrical charge effects on liquid particle nucleation and growth. Soot, liquid (without soot cores), and ice particles are discretized over a fixed size grid and can take variable radii in each size bin. Their chemical components are tracked individually.

[7] Turbulent plume mixing with ambient air cools the exhaust plume, entrains ambient particles and H_2O, and dilutes plume constituent mixing ratios. Uniform mixing is assumed across the plume cross section, which we regard as a reasonable approach to represent average conditions. Liquid plume and ambient particles consist of water and sulfuric acid (H_2SO_4). Coagulation and condensation lead to an aqueous H_2SO_4 coating around the emitted soot particles, turning them into internally mixed particles. Organic emissions have been omitted because little is known about their chemical nature. However, the mass fraction of condensable organics may become comparable to H_2SO_4 in cruise conditions, increasing the size of ultrafine liquid plume

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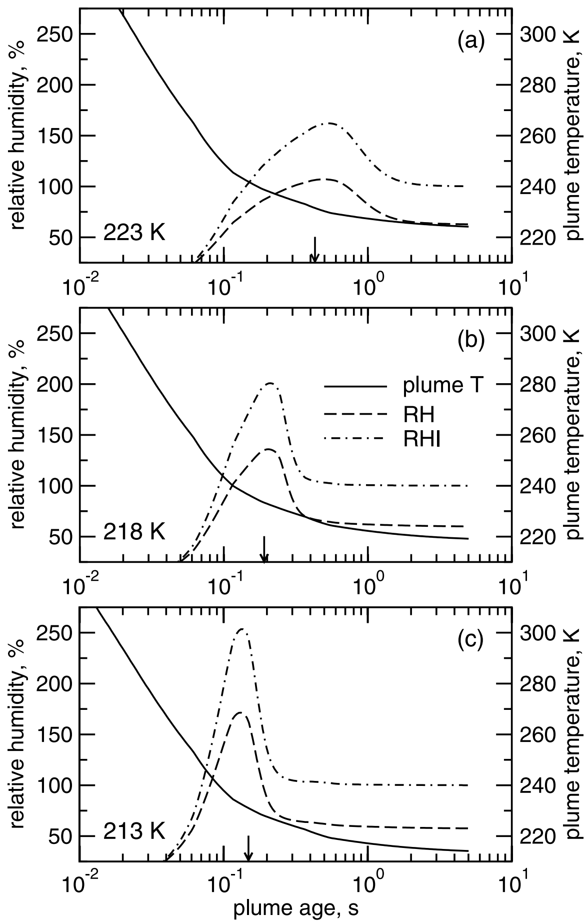

Figure 1. Evolution of plume temperature (solid curves) and relative humidity over water (RH, dashed) and over ice (RHI, dot-dashed) as a function of plume age for ambient temperatures of (a) 223 K close to the contrail formation threshold temperature, (b) 218 K close to the mean temperature at midlatitude upper tropospheric flight levels, and (c) 213 K, a value more commonly found at high latitude flight routes. Arrows in each panel mark the onset time of ice formation in the contrails.

particles [*Kärcher et al.*, 2000]. We point to uncertainties introduced by this omission in sections 3 and 4.

[8] Ice particles in visible contrails mainly form by homogeneous freezing of water-activated soot and liquid (plume and ambient) particles, an assumption that is consistent with available measurements [*Kärcher et al.*, 1998a, Figure 3]. This holds even though aqueous H_2SO_4 and coated soot particles might as well freeze homogeneously prior to water activation. Allowing soot particles to nucleate ice heterogeneously prior to water saturation (comparable to the homogeneous freezing of liquid solutions) would lead to a slightly earlier onset of contrail formation but would not significantly affect our results. More effective ice nucleation of soot particles closer to plume ice saturation contradicts in situ observations. Ice particles formed from these two particle types are tracked individually in the model and compete for available H_2O during depositional growth.

[9] The simulations have been initialized as follows: ambient air pressure and relative humidity over ice 230 hPa and 110%, respectively; plume dilution history for a large aircraft in cruise according to *Yu and Turco* [1998]; average fuel sulfur (S) content 0.2 g/kg; S-to-H_2SO_4 conversion efficiency 3% and chemi-ion emission index 2×10^{17} (kg-fuel)$^{-1}$ consistent with airborne measurements [*Kärcher et al.*, 2000]; bimodal lognormal soot particle size distribution (PSD) with scaled concentrations (fixed ratio of 2615:1 of small versus large mode) and mean diameters D (geometric width σ) 25 nm (1.55) and 150 nm (1.65) estimated from in situ data [*Petzold et al.*, 1997]; lognormal ambient PSD with total number density 600 cm^{-3}, D = 30 nm, and σ = 2.3, representing average upper tropospheric cases. Changing these parameters within reasonable ranges, adding organic emissions, and including applications to smaller aircraft would not significantly alter our results for medium to high soot emission levels.

3. Results and Discussion

3.1. Formation Conditions

[10] The evolution of temperature and relative humidities over supercooled water (RH) and over ice (RHI) in the jet plume are shown in Figure 1 assuming warm (Figure 1a), average (Figure 1b), and cold (Figure 1c) ambient temperatures (T) and a soot emission index $EI_s = 10^{15}$ (kg-fuel)$^{-1}$. In the threshold case at $T_* \approx T$ = 223 K, RH only slightly surpasses water saturation when ice particles start to form. In colder conditions, the plume cooling rates are higher due to faster mixing, leading to a steeper rise of RH and RHI with higher maxima, and an earlier onset of contrail formation. As more ice particles form with decreasing T (see below), RHI becomes more rapidly reduced to saturation despite continuous entrainment of slightly supersaturated air.

[11] To obtain our main results presented in Figure 2, EI_s has been used as a free parameter covering a wide range of values. We report the total number densities n_i of contrail ice particles versus EI_s at a plume age t = 1 s shortly after ice formation (arrows in Figure 1) (Figures 2a–2c) and their PSDs for selected EI_s at t = 5 s at the end of the jet regime after which aircraft wake vortex processes control the subsequent microphysical evolution (Figures 2d–2f).

3.2. Soot-Rich Regime

[12] In the soot-rich regime for $EI_s \geq 10^{15}$ (kg-fuel)$^{-1}$, ice particles mainly form by homogeneous freezing of water around the soot cores (compare solid and dashed curves in Figures 2a–2c). For $EI_s = 10^{15}$ (kg-fuel)$^{-1}$, the fraction of soot particles forming ice (not shown but reflected in the n_i-values) increases with decreasing T, because freezing rate coefficients rise steeply, achieving values 0.09 (223 K), 0.98 (218 K), and 1 (213 K). The frozen soot fraction changes very rapidly near T_*. The n_i-values at t = 1 s increase from ~5000 cm^{-3} (223 K) to ~50,000 cm^{-3} (213 K). These values are 1.5–2.5 times higher at the time of contrail formation because of plume dilution, respectively.

[13] Furthermore, at a given T, n_i rises with increasing EI_s as mixed particles are turned into ice before RHI decreases. The decrease in RHI is mainly forced by dilution (223 K) or by deposition of H_2O on growing particles (218 K). In the latter case, n_i tends to saturate with increasing EI_s indicating that the rate of depletion of supersaturation exceeds the freezing rate. At 213 K, despite an even faster decrease of

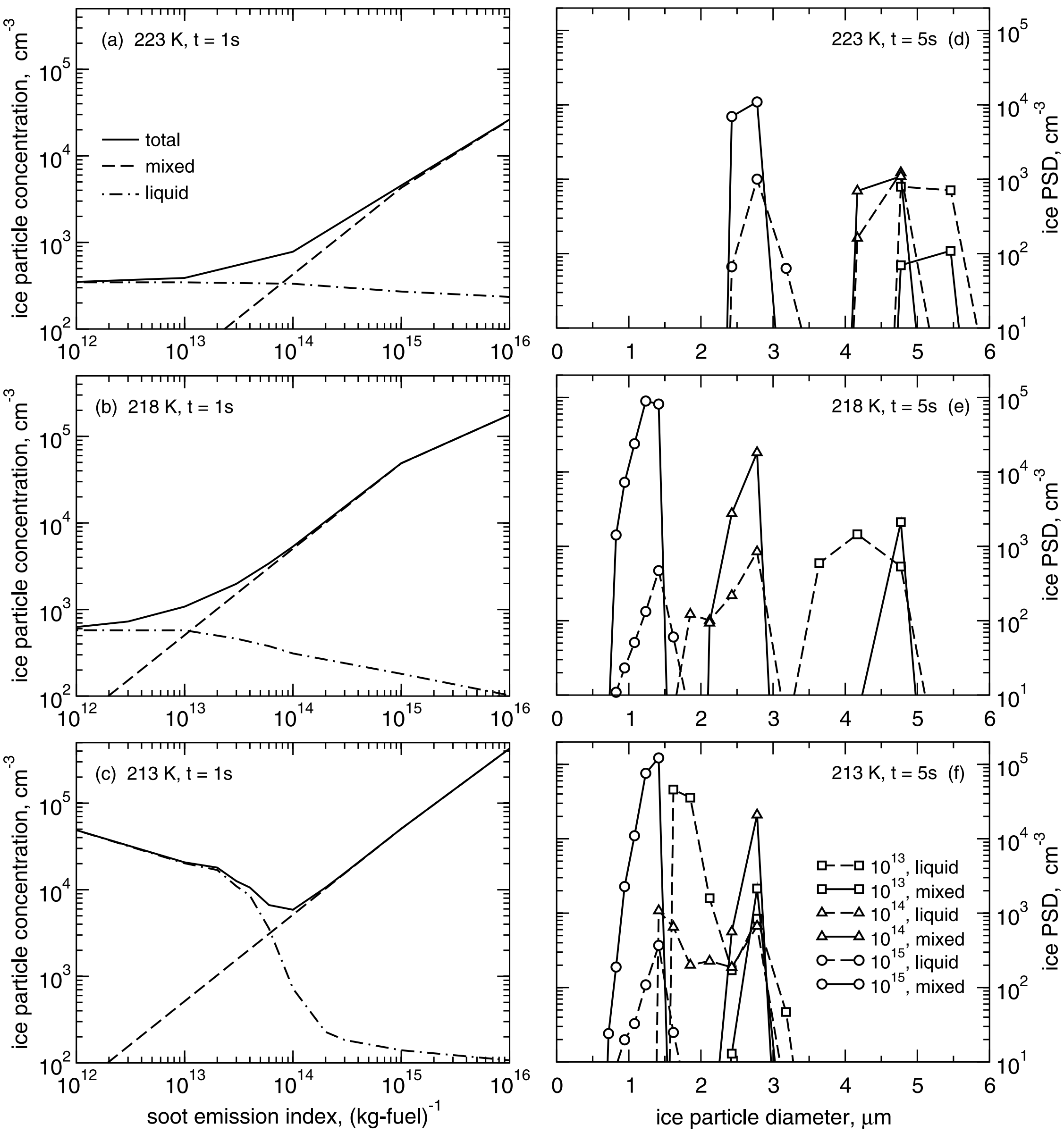

Figure 2. Ice particle concentrations n_i at a plume age of $t = 1$ s and ice particle size distributions $dn_i/d\log(D_i)$ at $t = 5$ s for ambient temperatures (a, d) 223 K, (b, e) 218 K and (c, f) 213 K. Total n_i (solid curves) and respective contributions from liquid plume and ambient (dot-dashed) and mixed soot-containing particles (dashed) are shown as a function of EI_s. The ice PSDs originating from freezing of liquid (dashed) and mixed particles (solid) are shown for selected values of EI_s (see legend). At $t = 1$ s, $n_i = 5 \times 10^4$ cm^{-3} corresponds to a dilution-corrected value of $\sim 10^{15}$ (kg-fuel)$^{-1}$, staying constant in the jet regime.

RHI, n_i rises again strictly in proportion to EI_s because freezing rates dominate over the water vapor depletion rates.

[14] The contribution of liquid (soot-free) particles to contrail formation is negligible (dot-dashed curves in Figures 2a–2c). Concentrations of ice particles forming on entrained ambient particles reach 200-300 cm^{-3}, depending on T. The plume supersaturation ratios are too small to activate the ultrafine liquid plume particles into water droplets for subsequent homogeneous freezing.

[15] The PSDs at $t = 5$ s (Figures 2d–2f) reflect the partitioning of contrail ice between liquid and mixed particles as discussed above. The mean diameters D_i of ice particles from ambient liquid particles (dashed) are slightly larger than those from the more abundant soot-containing particles (solid) since the former are initially larger and therefore more efficient in freezing. The ice PSDs contain particles with $D_i \approx 3$ μm (223 K) and ~ 1.5 μm (218 K and 213 K).

3.3. Soot-Poor Regime

[16] In the soot-poor regime, most contrail ice originates from liquid particles without soot cores. Figures 2a–2c show that this regime is realized for $EI_s \leq 10^{13}$ (kg-fuel)$^{-1}$ in the cases 223 K and 213 K (for different reasons) but only below 10^{12} (kg-fuel)$^{-1}$ at 218 K.

[17] In threshold conditions, total n_i is determined by the number of entrained ambient particles available for

freezing at the time of maximum supersaturation. Somewhat more particles freeze ($\sim$350 cm^{-3}) than in the soot-rich regime because soot particles acting as condensational sinks for H_2O are lacking. At lower T, n_i increases and becomes controlled by freezing of the liquid particles that nucleated and grew in the plume prior to ice formation. Only a small fraction of these copious, nm-sized particles can be activated. This fraction increases sharply between 218 K and 213 K, resulting in very high n_i similar to levels in the soot-rich regime.

[18] Less abundant engine metallic particle emissions, from burning of traces of metal compounds contained in kerosene or from engine erosion, would contribute to contrail ice [*Kärcher et al.*, 1998a] but would not be noticeable in Figures 2a–2c. Uptake in aqueous plume particles of nitric acid formed via rapid oxidation of emitted nitrogen oxides [*Kärcher*, 1996] and emissions of condensable organics from unburned fuel hydrocarbons [*Yu et al.*, 1999] would further increase the fraction of freezing liquid plume particles by increasing their size and activation probability. Therefore, our n_i-values in the soot-poor regime are considered lower-limit estimates, especially near and below 218 K.

[19] The simulated ice PSDs (Figures 2d–2f) confirm that ice particles originating from liquid particles are more abundant than those from soot. Furthermore, D_i is larger since the total number of particles formed is smaller and they share the same amount of available H_2O. At 223 K, both liquid and mixed ice modes have $D_i \approx 5.5$ μm. At 218 K, these two modes begin to split up into a smaller liquid ($\sim$4.2 μm) and larger mixed ($\sim$4.8 μm) mode. At 213 K, this bimodality is even more distinct, the liquid mode itself being separated into a first mode ($\sim$3 μm) originating from freezing of ambient particles and a second mode ($\sim$2 μm) from plume particles.

3.4. Intermediate Regime

[20] Total n_i decreases from the high values in the soot-rich regime to the comparatively low levels in the soot-poor regime (Figures 2a–2c). The crossover at which ice formation from liquid and mixed particles becomes equal shifts from 10^{14} (kg-fuel)$^{-1}$ at 223 K to 10^{13} (kg-fuel)$^{-1}$ at 218 K and back at 213 K. In the latter case, n_i takes a minimum near 10^{14} (kg-fuel)$^{-1}$, minimizing the susceptibility of n_i as a function of EI_s. As the simulated n_i-values in the soot-poor regime are lower-limit estimates, this minimum would already develop near 218 K had we included condensable organic emissions and nitric acid in the simulations. The mean diameters of contrail ice particles vary in the range 2.5–4.5 μm from low to high T.

[21] At each T, decreasing EI_s from 10^{15} to 10^{14} (kg-fuel)$^{-1}$ causes a tenfold decrease in n_i and roughly a twofold increase in D_i; including changes in the scattering efficiency of visible light, this reduces the contrail optical depth by less than 30–40% at t = 5 s.

4. Summary and Implications

[22] Salient points of previous model results [*Kärcher et al.*, 1998a] obtained for fixed soot emissions have been confirmed by an independent model approach, building confidence in our ability to model the microphysical processes controlling persistent contrail formation. The present study, representative for extratropical upper tropospheric flight levels, confirms that (1) it is not possible to reduce the occurrence frequency of, or entirely avoid, contrails by changing or eliminating soot emissions; (2) total n_i increases and D_i decreases with decreasing T; (3) at ambient temperatures below but near T_*, a large fraction of unactivated soot particles remains as interstitial aerosol in the contrail.

[23] Furthermore, we have shown that the majority of contrail ice particles originates from aircraft soot particles for $EI_s > 1–2 \times 10^{14}$ (kg-fuel)$^{-1}$, including the range $\sim 3 \times 10^{14} - 2 \times 10^{15}$ (kg-fuel)$^{-1}$ from the current fleet [*Petzold et al.*, 1999]. Total n_i is not sensitive to H_2SO_4 and chemi-ion emissions and increases (D_i decreases) with increasing EI_s. Detailed knowledge of soot PSDs and ice nucleation properties is only needed to predict visible contrail formation accurately near but slightly below T_* when the plume barely overshoots water saturation, or to predict (initially) subvisible contrail formation near but slightly above T_* when the plume becomes ice- but not water-supersaturated.

[24] For $EI_s \leq 10^{12} - 10^{13}$ (kg-fuel)$^{-1}$ depending on ambient temperature, contrail ice formation is dominated by ambient and liquid plume particles, with T determining their relative contributions. These contributions can only be predicted with confidence if their PSDs and ice nucleation properties are known. Total n_i decreases to ambient levels and D_i increases with decreasing EI_s, but only for $T > 218$ K as plume cooling rates and hence supersaturation are insufficient to activate and freeze the small acidic liquid plume particles. In colder conditions, the situation reverses, and predictions of n_i depend on chemi-ion and condensable sulfur, nitric acid, and organics levels available at emission. Changes in concentrations of condensable species result in changes in the size of liquid particles and hence their water activation properties [*Kärcher*, 1996; *Yu and Turco*, 1997].

[25] Our work relates to technological efforts aiming at reducing soot emissions using jet fuel additives [*Liscinski et al.*, 2001; *Bae and Avedisian*, 2004; *Montgomery et al.*, 2005]. Reducing EI_s from current average levels of $\sim 5 \times 10^{14} - 10^{15}$ (kg-fuel)$^{-1}$ by a factor 5 would lower n_i by approximately the same factor and less than double D_i. This result highlights the potential to alter nascent contrail properties. Further reduction of EI_s would push n_i down to ambient levels in warm ($T > 218$ K) conditions, especially near T_*. This is the largest possible effect, achievable only for very efficient removal (>90%) of soot particle emissions, or when using non-sooting fuel.

[26] However, in the soot-poor regime, no further reduction of n_i, even the opposite effect, is realized at average and cold temperatures because ultrafine liquid plume particles then act as ice forming agents despite their small sizes. With today's use of kerosene, a concomitant removal of fuel sulfur would not alter this conclusion, as those soot-free particles still form by ion-induced nucleation and condensation of emitted organic vapors according to analyses of in situ observations [*Kärcher et al.*, 1998a, 1998b; *Yu et al.*, 1998]. Any organic compound present in soot-reducing fuel additives might increase the size of liquid plume particles and thus heighten their role in contrail ice formation. This implies that it might be more beneficial to reduce EI_s rather than to entirely remove soot emissions.

[27] Our simulations suggest a significant potential to lower the initial contrail visible optical depth. Subsequent microphysical processing of ice particles after the jet regime might enhance or alleviate such changes. Therefore, it remains open if and to which degree the differences in contrail properties resulting from reductions in soot emissions propagate into the further evolution of contrails and affect their radiative properties. This issue, however, is crucial for assessing the potential of mitigating the contrail climate impact based on reductions in soot emissions.

[28] Future atmospheric research into persistent contrails should investigate their development into cirrus as a function of the factors controlling contrail ice formation. Similar efforts are needed to study contrail cirrus induced by aerodynamic effects [*Kärcher et al.*, 2008] or forming from alternative fuels such a liquid hydrogen [*Ström and Gierens*, 2002].

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