

## Effects of aircraft on aerosol abundance in the upper troposphere

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**Abstract.** A significant increase in H<sub>2</sub>SO<sub>4</sub> aerosol concentration coincidental with an enhancement in NO was detected above 10 km pressure altitude during a cross-corridor flight out of Shannon on October 23, 1997. The source of this aerosol is ascribed to commercial aircraft operations in flight corridors above 10 km, because (1) a stable atmosphere prevented vertical air mass exchanges and thus eliminated surface sources, (2) air mass back trajectories documented the absence of remote continental sources, and (3) temperature profiler data showed the tropopause at least one kilometer above flight altitude throughout the flight, thus excluding stratospheric sources. Particle volatility identified (60-80)% H<sub>2</sub>SO<sub>4</sub>, ≈20% (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and ≈10% nonvolatile aerosol in the proximity of flight corridors, and (10-30)% H<sub>2</sub>SO<sub>4</sub>, up to 50% (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, and (40-60)% nonvolatile aerosols in air that was not affected by aircraft operations below 10 km. The newly formed H<sub>2</sub>SO<sub>4</sub> particles did not measurably affect surface area and volume of the background aerosol due to their small size, hence did not influence radiative transfer directly.

### Introduction

Aircraft jet engines directly emit aerosols and condensable gases, such as water vapor (H<sub>2</sub>O) and sulfur dioxide (SO<sub>2</sub>) which lead to the formation of new liquid (volatile) sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), particles in the early plume by gas-to-particle conversion (nucleation) processes. Soot aerosol formed during incomplete fuel combustion constitutes part of the nonvolatile particle fraction. The newly formed particles grow by condensation and coagulation amongst themselves and with the background aerosol.

Aviation-produced particles may influence the state of the atmosphere in various ways. For example, they provide surfaces for heterogeneous chemical reactions. Heterogeneous reaction rates depend on the chemical characteristics of aerosols. For example, the reaction probabilities of N<sub>2</sub>O<sub>5</sub> on NH<sub>4</sub>H<sub>2</sub>SO<sub>4</sub>, formed by reaction of H<sub>2</sub>SO<sub>4</sub> from aircraft with ammonia in background air, show a strong dependence on relative humidity and temperature, decreasing with lowering relative humidity and increasing temperature (Mozurkewich and Calvert, 1988). In contrast, measurements of N<sub>2</sub>O<sub>5</sub> hydrolysis on H<sub>2</sub>SO<sub>4</sub> particles show only a weak dependence on temperature and relative humidity (Hanson

and Ravishankara, 1991). Therefore, it is important to determine the enhancement of H<sub>2</sub>SO<sub>4</sub> aerosol by aircraft in relation to the fraction of those particles that are neutralized to (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in order to assess the atmospheric chemical modification potential of aircraft-produced aerosol. Aviation-generated aerosols also form a sink for condensable gases, they absorb or scatter radiation directly, and they change cloud properties which may affect radiation indirectly. In order to assess those effects, aviation-produced aerosol characteristics have to be determined in relation to background aerosol properties.

Model calculations of aircraft emissions indicate that (6-10)% of SO<sub>2</sub> is oxidized to SO<sub>3</sub> and subsequently to H<sub>2</sub>SO<sub>4</sub> to form new small aerosols (Miake-Lye, 1994; Kärcher, 1996; Brown et al., 1996, 1997). Measurements in aircraft wakes, on the other hand, have shown that up to 37% of SO<sub>2</sub> can be oxidized to H<sub>2</sub>SO<sub>4</sub> aerosols (Fahey et al., 1995a,b; Miake-Lye et al., 1998; Pueschel et al., 1998; Anderson et al., 1998; Paladino et al., 1998). These measurements have also shown that the number density of volatile particles in aircraft wakes is proportional to the S-content of the fuel burned, implying that condensable S compounds are responsible for the formation of aerosol in aircraft wakes.

In this paper we show that increases in H<sub>2</sub>SO<sub>4</sub> aerosol are also detectable in the vicinity of commercial aircraft flight corridors away from dedicated aircraft plumes. The measurements were made during the Subsonic Assessment-Ozone and Nitrogen Oxide Experiment (SONEX). A stable atmosphere on 23 October 1997 separated air masses containing flight corridors above 10 km from air masses below 10 km that were not affected by air traffic. Temperature profiler data determined a height of the tropopause that was above DC-8 flight levels by at least 1 km throughout the flight, thereby eliminating stratospheric intrusions. Furthermore, air mass back trajectories established the absence of long range transport of pollutants from the North American continent. Thus, SONEX Flight No. 7 across commercial airline flight corridors on that day provided an opportunity to determine aerosol characteristics in both aircraft-affected and non-affected air masses in the upper troposphere. Our results document that subsonic aircraft are a source of detectable sulfuric acid aerosol. Even though the particles are too small to measurably affect surface area and volume densities, this finding is an affirmative answer to one of the key questions addressed by SONEX: Can aircraft corridors be detected?

### Results and Discussion

Both the Ames MMS (Meteorological Measuring System) and the Jet Propulsion Laboratory MTP (Microwave Temperature Profiler) established the existence of a sub-adiabatic lapse rate during a cross-corridor flight out of Shannon on 23 October 1997. MTP data show that the lapse rate varied between -6.5 and -7.5 °C/km (against a -9.9 °C/km dry adiabatic lapse rate) during the entire flight. This atmospheric stability prevented vertical air mass exchange. Furthermore, a slight temperature inversion, shown in both the MMS and MTP data, separated air masses above 10 km from those below that altitude. The Florida State University (FSU) air mass back trajectory model showed the confinement of air masses within 25 degrees latitude over the

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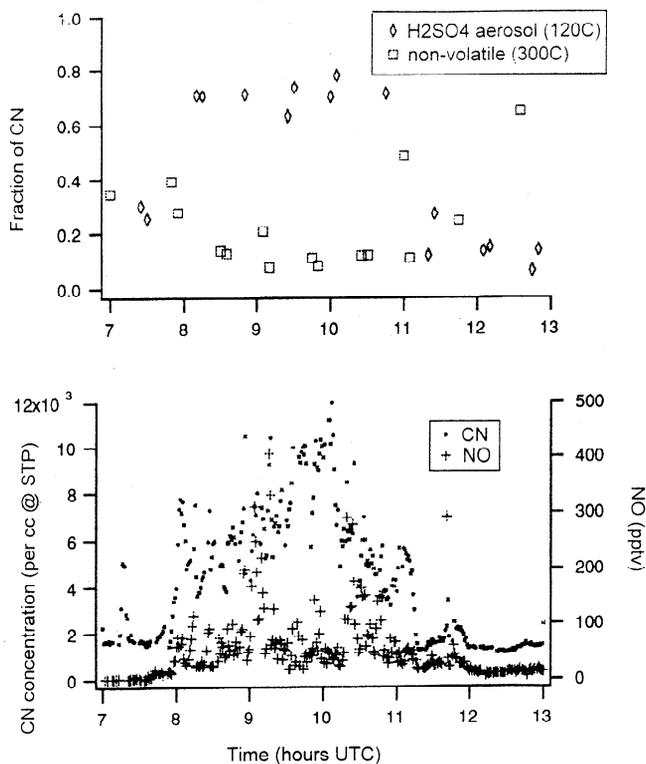
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Atlantic Ocean upwind of the flight track both above and below 10 km pressure altitude for 5 days prior to 23 October. This absence of long-range transport prevented advection of pollutants from North America and permitted accumulation of aircraft emissions in the proximity of corridors upwind of the sampling aircraft. The inhibited transport of aerosols from the Atlantic Ocean's boundary layer, the North American continent and the stratosphere left aircraft flying in corridors above 10 km as the most likely source of particulate pollutants along the flight track.

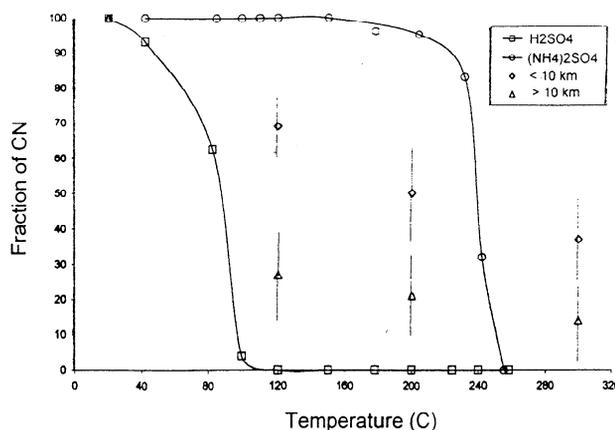
A combination of fine-particle and NO measurements constitutes one of the best tracers of aircraft effluents (Paladino et al., 1998; Schumann, 1997). Fig. 1 shows in the lower panel the ambient fine particle (CN) concentration as dots and NO concentration as plusses, and in the upper panel the fraction of particles that remain at 120C and 300C, respectively, as function of UTC time in hours for all of the flight. CN particles were detected by growing particles  $> 0.003 \mu\text{m}$  diameter at 400% supersaturation in the cloud chamber of a TSI Model 3025 ultrafine CN counter to optically detectable sizes. A heater upstream of the CN counter permitted differential heating of the air up to 300C prior to entering the cloud chamber.

Noticeable in Fig. 1 is a significant increase in fine particle and NO abundance at 8:00 hours UTC when the aircraft ascended through 10 km pressure altitude, and a corresponding decrease at 11:08 hours UTC while passing through 10 km pressure altitude during descent. Simultaneous gas measurements by other investigators yielded  $\approx 40$  ppbv  $\text{O}_3$  and 60-70 ppbv CO, both indicative of tropospheric air, which were not affected by aircraft. NO was the only gas that was enhanced above 10 km proportional to the increase in CN.

Figure 2 shows the fraction of particles that remain at 120C, 200C, and 300C temperature in relation to the thermal volatility



**Figure 1.** Lower panel: Ambient ultrafine CN (dots) particle and NO (plusses) concentrations. Upper panel: Fractions of particles remaining at 120C (diamonds), 200C (triangles) and 300C (squares). Abscissa in both panels is flight time in UTC hours. Date is 23 October 1997.



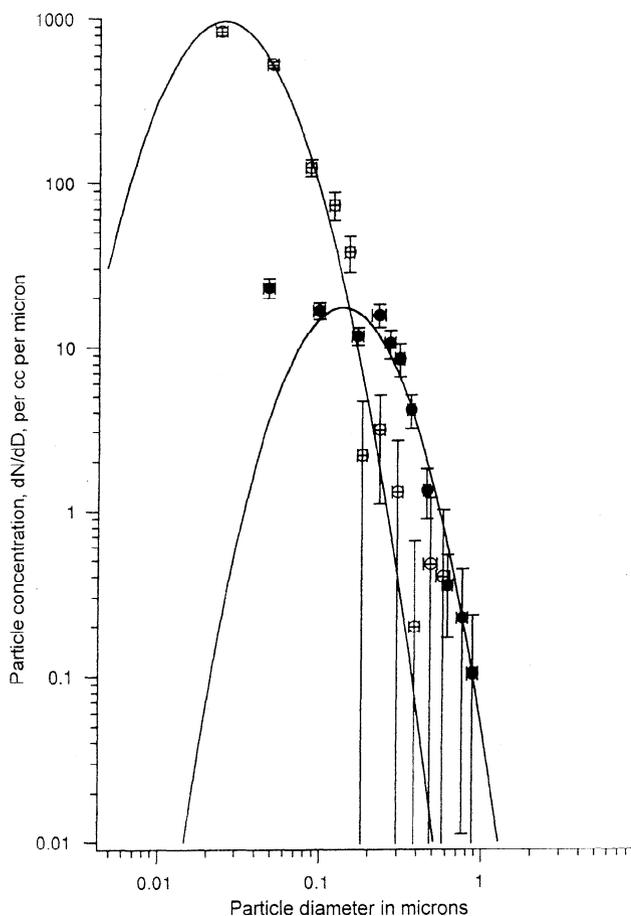
**Figure 2.** Thermal decomposition spectra of sulfuric acid (squares) and ammonium sulfate (circles). The diamonds and triangles are averages of measurements below 10 km and above 10 km pressure altitude, respectively on 23 October 1997.

spectra (Clarke 1991) of  $\text{H}_2\text{SO}_4$  (curve dropping off near 100C) and  $(\text{NH}_4)_2\text{SO}_4$  (curve dropping off near 240C). It follows from Fig. 2 that 73% by number of fine particles in the air mass above 10 km near commercial flight corridors evaporated at 120C, hence consisted of sulfuric acid, while 14% could be classified as "non-volatile" aerosols (defined as those that exist at 300C) and 13% showed thermal characteristics that were typical of  $(\text{NH}_4)_2\text{SO}_4$ . In contrast, below 10 km, only 31% of the aerosol showed volatility that is typical of sulfuric acid, 37% resembled nonvolatile aerosols, and 32% were  $(\text{NH}_4)_2\text{SO}_4$ -like. Thus, the sulfuric acid aerosol abundance above 10 km increased almost tenfold, while the total aerosol increased approximately fourfold above the concentration below 10 km.

The increase in the concentration of an inert passive tracer in the aircraft wake above its background concentration is  $\Delta c_i = EI_i/N$ , where  $EI_i$  is the emission index and  $N$  the dilution ratio (Schumann et al., 1998). From Fig. 1 it follows that  $\Delta(\text{NO}) = 50$  pptv. Assuming an emission index  $EI_{\text{NO}} = 10 \text{ g/kg}_{\text{fuel}} = 0.16 \text{ g/kg}_{\text{air}}$  (at a fuel-to-air ratio of 0.016), the dilution ratio amounts to  $N = 3.2 \times 10^6$ . Schumann et al. (1998) established a relationship between dilution ratio  $N$  and time after emission of effluents as  $N = 7e3(t/t_0)^{0.8}$  (where  $t$  is in seconds and  $t_0$  equals 1 sec). Hence it follows that, on the average, the plumes were 0.6 hours old.

The CN emission index was calculated from the relationship  $EI_{\text{CN}} = EI_{\text{NO}} \times (\Delta\text{CN}/\Delta\text{NO}) \times (M_{\text{air}}/M_{\text{NO}} \times \rho)$ , where  $EI_{\text{NO}} = 10 \text{ g/kg}_{\text{fuel}}$  is the NO emission index,  $M_{\text{air}}$  and  $M_{\text{NO}}$  are the molecular weights of air and NO, respectively, and  $\rho$  is the density of air. From Fig. 1 it follows that  $\Delta\text{CN} = 6e3 \text{ cm}^{-3}$  at STP, corresponding to  $\Delta\text{CN} = 1.6e3 \text{ cm}^{-3}$  at 10 km altitude, and  $\Delta\text{NO} = 50$  pptv. Hence,  $\Delta\text{CN}/\Delta\text{NO} = 32 \text{ cm}^{-3} \text{ pptv}^{-1}$  and  $EI_{\text{CN}} = 8e17 \text{ particles/kg}_{\text{fuel}}$  which approaches the upper bound of values reported by Andersen et al. (1999).

The aerosol characteristic that determines both heterogeneous reaction rates and light scattering properties is the particle surface area density. Single-instrument CN measurements do not permit a determination of particle surface area, because they constitute a bulk particle density without significant information on particle size. The only information about particle size that is available from CN measurements is a detectable minimum size which depends on the supersaturation at which CN counters are being operated. We determined particle number, surface area and volumes independently from CN measurements by collecting particles by impaction on wires. This sampling technique (e.g., Pueschel et al., 1992) by inertial impaction is fundamentally different from CN measurements. The collection efficiency of particles on wires



**Figure 3.** Particle size distributions measured above 10 km at 8:59-9:01 UTC (open circles) and below 10 km at 7:09-7:12 UTC (solid circles) pressure altitudes on 23 October 1997.

at nominal jet aircraft speeds drops off rapidly at 0.05  $\mu\text{m}$  particle diameter (Lem and Farlow, 1979). Impactor sampling has a distinctive advantage over other techniques, however, by allowing morphological and chemical analysis of those particles that were collected, thereby offering the capability of a distinction between soot and other particles (Pueschel et al., 1992).

Figure 3 shows particle size distributions from samples collected on 23 October below and above 10 km pressure altitude. Least square curve fits to the data yield log-normal characteristics  $N_{01}=4.0 \text{ cm}^{-3}$ ,  $r_{01}=0.09 \mu\text{m}$  and  $\sigma_1=1.8$  for the lower, and  $N_{02}=40 \text{ cm}^{-3}$ ,  $r_{02}=0.02 \mu\text{m}$  and  $\sigma_2=2.0$  for the higher altitude, respectively. From those log-normal parameters follow particle surface area densities  $A_1 = 0.81 \mu\text{m}^2 \text{ cm}^{-3}$  and  $A_2 = 0.60 \mu\text{m}^2 \text{ cm}^{-3}$  and particle volume densities  $V_1 = 0.057 \mu\text{m}^3 \text{ cm}^{-3}$  and  $V_2 = 0.014 \mu\text{m}^3 \text{ cm}^{-3}$  for air sampled below and above 10 km pressure altitude, respectively. The importance of this result is the fact that aircraft operations do not enhance either particle surface area or particle volume of aerosol larger than 0.05  $\mu\text{m}$  radius, in spite of a significant increase of fine-particle concentration.

Particle concentration, surface area and volume densities have been derived independently from log-normal curve fits by integrations over number (shown in Fig.3), surface area and volume size distributions. The results are shown in Table 1.

The differences between results from the log-normal curve fits and those derived by numerical integration lie within experimental error. Both data sets show that particle concentration is the only aerosol characteristic that is significantly enhanced in air masses that are affected by aircraft operations, whereas surface area and volume densities are not. Fig.3 indicates that those aircraft-produced particles are smaller than 0.1  $\mu\text{m}$  diameter. A comparison of the results of Fig.3 with those of Fig.1 suggests that the majority of those particles are smaller than 0.05  $\mu\text{m}$ , hence fall in the (0.003-0.05)  $\mu\text{m}$  size range. It is this small size that renders aircraft-generated sulfuric acid particles unimportant for either aerosol surface area or volume. Their coagulation with other aerosols, however, may affect the surface characteristics of those aerosol particles by (partially) coating them with sulfuric acid. Thus it is possible that particles that are initially hydrophobic, such as soot, are converted to effective cloud condensation nuclei to potentially affect the cloud particle sizes and concentrations and thus cause indirect effects.

The Ames Wire Impactor samples were also analyzed for soot aerosol which is identified morphologically by the fractal nature of soot particles, thereby distinguishing them uniquely from other aerosols (e.g., Pueschel et al., 1992). Those results are also shown in Table 1. From this data it follows that, in contrast to a significant effect on the sulfuric acid fine-particle abundance, aircraft leave no measurable imprint on the soot aerosol characteristics. In fact, contrary to what applies to  $\text{H}_2\text{SO}_4$  fine particles, soot aerosol above 10 km is less abundant than below 10 km altitude. Of particular note is the fact that the morphologically determined soot aerosol comprises a mere few percent of the total aerosol. In contrast, the non-volatile aerosol fraction identified by preheating air prior to entering a CN counter (Fig.1 and Fig.2) amounts to up to 13%. In order to resolve this discrepancy, one has to assume that either CN counters detect refractory aerosols other than soot, including only partially evaporated  $(\text{NH}_4)_2\text{SO}_4$ , and/or chemions, or that the majority of soot particles are too small ( $<0.05 \mu\text{m}$ ) to efficiently be collected by inertial impingement on wire impactors. The latter assumption is doubtful, because electron-microscopic analysis of soot fractals shows that individual soot spherules are large enough to be collected by wire impactors. Because the supersaturations in CN counters are extremely high, the possibility exists that they possibly count chemions which are generated in aircraft jet engines (Yu and Turco, 1997). We, therefore, caution against an unqualified interpretation of non-volatile CNs as soot particles.

**Summary**

A sub-adiabatic lapse rate, amplified by several inversion layers at various altitudes, prevented vertical exchange of air masses during a cross-corridor flight out of Shannon on 23 October 1997. Air mass back trajectories showed that air parcels reaching the aircraft had been over the Atlantic Ocean for at least 5 days

**Table 1:** Particle number, surface area and volume densities above and below 10 km pressure altitude.

Altitude	< 10 km		> 10 km	
	Aerosol	Soot	Aerosol	Soot
N ( $\text{cm}^{-3}$ )	4.6±0.3	(1.9±1.0)e-1	40±1.4	(9.6±3.4)e-2
A ( $\mu\text{m}^2 \text{ cm}^{-3}$ )	(7.7±0.9)e-1	(3.7±1.9)e-2	(6.0±0.9)e-1	(2.4±1.2)e-2
V ( $\mu\text{m}^3 \text{ cm}^{-3}$ )	(4.8±0.9)e-2	(2.3±1.1)e-4	(1.6±0.8)e-2	(1.9±0.9)e-4

prior to sampling. Microwave Temperature Profiler data showed the absence of stratospheric-tropospheric exchanges. In spite of this inhibited vertical and horizontal transport, a significant enhancement of fine particle and NO concentrations was detected above 10 km pressure altitude in relation to concentrations at lower altitudes. Aircraft operated in flight corridors above 10 km were the most likely source of this fine-particle abundance. From measured NO enhancement and assumed  $EI_{NO}=10 \text{ g/kg}_{fuel}$  we calculated an average plume dilution ratio  $N=3.2e6$  and an average time between emission and measurement of 0.6 hours. The measured  $\Delta CN/\Delta NO=30 \text{ cm}^{-3} \text{ ppt}^{-1}$  ratio yielded a CN emission index  $EI_{CN}=8e17 \text{ particles/kg}_{fuel}$ . In contrast to fine-particles and NO, the non-volatile and soot component of the aerosol was not measurably affected by aircraft.

Volatility measurements identified sulfuric acid as the predominant compound of the aircraft-generated aerosol, raising the sulfuric acid fraction of the total aerosol to >70% of  $8e3 \text{ cm}^{-3}$  STP particle density from <30% of  $2e3 \text{ cm}^{-3}$  STP density. The rest was approximately equally divided between  $(NH_4)_2SO_4$  and a non-volatile component, a small fraction of which was soot aerosol. In contrast to volatile CNs and NO, an enhancement near flight corridors of non-volatile CNs and soot particles was not observed. This finding suggests that we sampled aircraft effluents that were sufficiently diluted to completely mix the non-volatile CNs and other gaseous effluents (with the exception of NO) with the ambient background aerosol.

Ames wire impactor samples, collected concurrently with the fine particle measurements, showed that only particle number density was influenced by aircraft. Neither particle surface area nor volume densities were affected because of the small size ( $0.003 < R < 0.05 \mu\text{m}$ ) of the aircraft-generated particles. Thus, a direct radiative effect of aircraft-produced aerosol is not to be expected. However, coagulation with the aircraft-generated sulfuric acid particles could render hydrophobic aerosols, such as soot, to become cloud condensation nuclei and thereby constitute an indirect effect.

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