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## Particle concentration characterization for jet engine emissions under cruise conditions

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**Abstract.** Airborne particle measurements during NASA project SUCCESS have shown that particle concentration profiles serve as good indicators of aircraft exhaust plume encounters. During exhaust plume penetrations there is a strong anticorrelation between the ratio of nonvolatile/total particulates and Nitrogen Oxide (NO) concentrations. An increase in fuel sulfur content was found to increase the total particle emission index, while the nonvolatile emission index remained unchanged. The EI's increased by a factor of 2.6 as the fuel sulfur increased from 70 - 700 ppm. The large particle size distribution (200-400 nm) was seen as a good long term indicator of an aircraft exhaust plume.

### Introduction

Within the last 10 years, the global community has become increasingly aware of man's impact upon the environment. With the discovery of the ozone hole, and the subsequent link to heterogeneous chemistry on the surface of particulates within the polar stratosphere, the role of particulates in the atmosphere has come under intense scrutiny. A major source of anthropogenic aerosols within the upper troposphere and lower stratosphere may be the emissions from jet engines. Hence, the chemical and physical characteristics of turbine engine emissions have come under scientific scrutiny (Ferri 1972; Luther *et al.* 1979; Widhopf *et al.* 1977; Hagen *et al.* 1989, 1990, 1991, 1992, 1993a, 1993b, 1996; Pitchford *et al.* 1991; Baumgardner and Cooper, 1994). Even though a large and diverse subsonic fleet of commercial aircraft has been operating for a substantial time, the impact of their associated engine emissions has not been well defined, nor is their effect on the upper troposphere and lower stratosphere obvious or easily separable from other perturbations. Additionally, aircraft emissions, especially particulates, can influence contrail formation and seed cirrus cloud formation (Schumann *et al.* 1996; Busen *et al.* 1996). Current atmospheric models predict a small climatic response for present contrail coverages (Ponater *et al.* 1996). Model results depend on background concentrations, emission data, mixing assumptions, and various other inputs, unfortunately very few measured data exist to aid in model predictions (Schumann *et al.* 1996).

A recent National Aeronautics and Space Administration (NASA) program was focused on investigating the formation of cirrus clouds, and their contrail precursors: Project SUCCESS (Subsonic Aircraft: Contrail and Cloud Effects Special Study).

This paper describes results from particle characterization measurements made primarily during NASA project SUCCESS, using the University of Missouri Mobile Aerosol Sampling System. The UMR instrumentation was flown on the NASA DC-8 research aircraft for in-situ atmospheric measurements both in aircraft exhaust trails and in the ambient background. This paper focuses on emission sampling from the NASA 757 aircraft, using Rolls Royce RB211-535C engines, which was used extensively as an emission source for this project. The May 3 encounters occurred at 32-35000 feet, where the temperature was -60° with a relative humidity near 70%. The encounter distance varied from behind the 757. A short lived 6-13 km contrail was observed.

### Instrumentation

For NASA project SUCCESS, the UMR Mobile Aerosol Sampling System (MASS) was installed on the NASA DC-8 research aircraft. The MASS facility is described elsewhere (Hagen *et al.* 1992; Hagen *et al.* 1996; Paladino, 1997). It consisted of a variable temperature heated inlet line, and an unheated inlet line feeding various aerosol characterizing devices: particle (CN) counters, three Electrostatic Aerosol Classifiers (EAC), two of which are operated in tandem with a 100% RH water saturator in between them, and a PMS Instruments Laser Aerosol Spectrometer (LAS). One of the CN counters was a TSI 3025 ultrafine CN counter, which counted particles with a diameter of 3 nm or greater, whereas the other counters were Met One counters with a 5 nm cutoff. Size distributions and hydration (CN activation) data can be acquired for particles in the diameter size range 7 nm to 5  $\mu$ m. However, in this study the analysis was limited to particles with a diameter < 400nm. Concentrations of total particles (CN), non-volatile particles, and particles in specific size bands were continuously measured.

The particle sampling occurred under ambient pressures with ram pressure probe sampling. The temperature change between ambient atmospheric conditions and the instrument was typically near 60 C, and was expected to remove the water and ice components from the sample flow, but to have only a minimal impact upon aircraft emissions such as soot, and sulfate aerosols which have lower vapor pressures. Particle concentrations were corrected for size, concentration, and pressure inefficiencies in the particle counters. These corrections were made based upon laboratory calibration curves (Paladino, 1997). Diffusional losses through the instruments sampling tubing were calculated based upon the penetration calculations given in Hinds (1982). The compounded uncertainties of the calibrations and corrections total less than 10%. Size distributions, using differential mobility analysis, were made on aerosol samples captured and held in

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a 13 liter sample tank. The coagulation and loss effects during the aerosol residence time within the tank, ( $\sim 100$  sec), were estimated using Gelbard's MAEROS model (Gelbard and Seinfeld, 1978,1980) and were found to be small: 1% at smaller sizes (50nm and lower), and less at larger particle diameters. The NO measurements used in this analysis scheme were those of Wienheimer (1997).

## Results

Particle measurements constitutes one of the best tracers of aircraft effluents. The particles are present in the exhaust immediately upon exiting the engine, and additional aerosols are produced within the expanding exhaust plume via gas-to-particle conversion and nucleation (Schumann, 1997). The UMR-MASS particle concentration measurements consist of both total particulate concentration ( $D_p > 5\text{nm}$ ), as measured by a cloud condensation nucleus counter, and a nonvolatile aerosol concentration with respect to thermal volatilization at  $175^\circ\text{C}$ ; which, in laboratory tests, was found to be greater than 90% efficient in volatilizing sulfuric acid aerosols less than 65nm in diameter, and 95% at 40nm (Paladino, 1997).

The NO concentration serves as another excellent indicator for near field aircraft engine emissions (Schumann, 1997). Both total particle concentration, and non-volatile particle concentration were found to correlate well with NO; thereby identifying exhaust plume penetrations. This is shown in Fig. 1, which presents particle concentrations, total and non-volatile, in conjunction with the concomitant NO concentration as a function of time during the DC-8's 3 May 96 flight, which encountered numerous aircraft plume signatures. Short lived contrails (1 km) were reported, but the measurements were acquired at a distance of 6-13 km behind the subject aircraft. Where both total and non-volatile particle concentrations were available, the ratio of non-volatile particles (NVCN) to total particles (TCN) can be calculated, and this exhibited a strong anticorrelation to the NO signature. This is shown in Fig. 2. This anticorrelation is likely due to gas-to-particle conversion greatly increasing the total aerosol population over the non-volatile concentration (soot) in the plume.

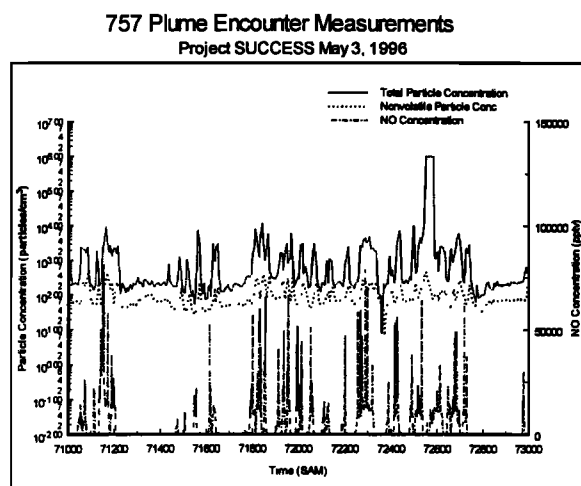


Figure 1. Total particle, non-volatile particle, and NO concentrations vs. time for the DC-8 flight on 3 May 1996 during the 757 exhaust plume penetrations.

Volatile Particle Ratio Anticorrelation with NO Concentration  
Project Success: 5/03/96

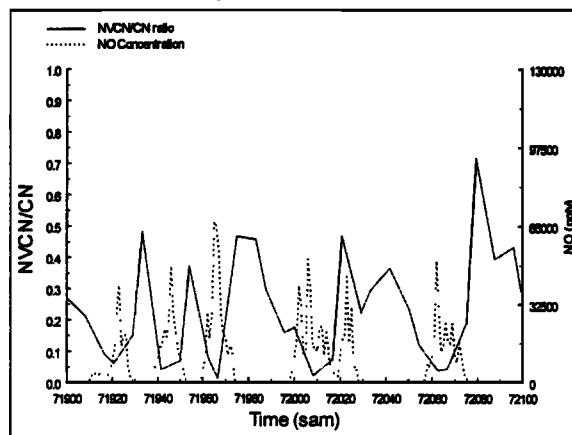


Figure 2. Ratio of non-volatile to total particle concentrations and NO concentration vs. time for the DC-8 flight on 3 May 1996 during the 757 exhaust plume penetrations.

The effect of changing the fuel sulfur content was studied in the 3 May 96 flight, using the NASA 757 as the exhaust source. During the time period 72000 SAM (seconds after midnight) to 72730, a high sulfur content fuel (700 ppm) was burned, and for times beyond 72730 a low sulfur content (70 ppm) was burned. Fig. 3 presents total, non-volatile emission indices for exhaust plume penetrations. The NO valves were controlled to a fixed concentration, so the data represents a plume of approximately the same age and dilution. The average total emission index for high sulfur exceeded that for the low sulfur by a factor 2.6. The corresponding ratio for non-volatile particles was shown to be 1.0 within the statistical uncertainty of the data. This is evidence of the production of aerosols within the expanding exhaust plume by gas-to-particle conversion, possibly from the oxidation and subsequent hydration of  $\text{SO}_2$  to sulfuric acid in the moisture laden environment of the expanding plume. Currently, there is a high degree of uncertainty in the oxidation of sulfur containing species. For example, Arnold *et al.* (1997) made obser-

Particle Emissions for Fuels of Varying Sulfur Content  
Project SUCCESS, May 3, 1996

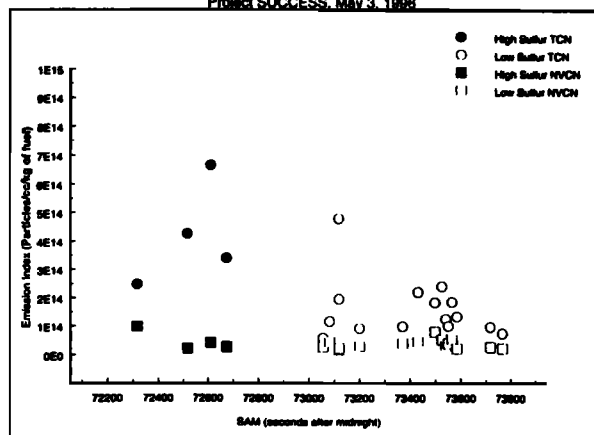


Figure 3. Total particle and non-volatile particle emission indices vs. time, plotted only for plume penetration times, for the DC-8 flight on 3 May 1996 during the 757 exhaust plume penetrations.

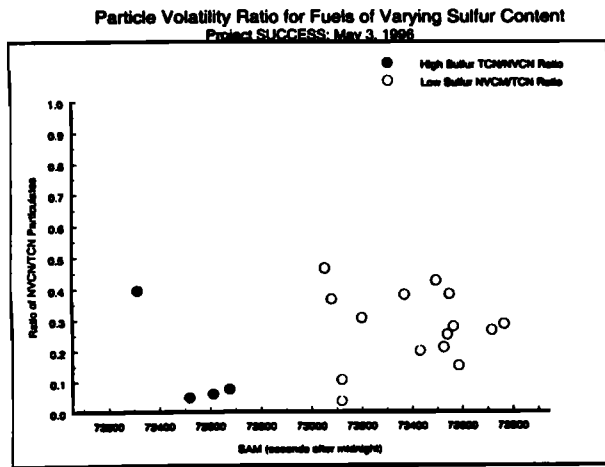


Figure 4. Ratio of non-volatile to total particle emission indices vs. time, plotted only for plume penetration times, for the DC-8 flight on 3 May 1996 during the 757 exhaust plume penetrations.

variations of gas and aerosol phase  $\text{H}_2\text{SO}_4$  in aircraft contrails using the Volatile Aerosol Composition Analysis technique (Curtis *et al.*, 1997) and reported observing good conditions for  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  gas-to-particle production, whereas modeling (Brown *et al.*, 1996a,b) results do not support the development of sulfuric acid particles of sufficient size to be detectable. Fig. 4 shows the non-volatile/total particle emission index ratio for data taken in the plume. Here, the average ratio for low sulfur exceeds that for high sulfur by a factor 1.75. The NVCN/TCN ratio for low sulfur was large since the total particle concentration approached the non-volatile concentration values.

Large particle characterization ( $D_p > 200\text{nm}$ ) was performed with a PMS Instruments Laser Aerosol Spectrometer (LAS), which uses a laser scattering technique to count and size the particles. Whereas the peak in the size distributions for most jet engine emissions lies within the range of 20–60nm, there is also a considerable amount of ambient background aerosol at these sizes (Howard *et al.* 1996). However, the larger diameter particles are fewer in the ambient atmosphere, and so monitoring the population of larger

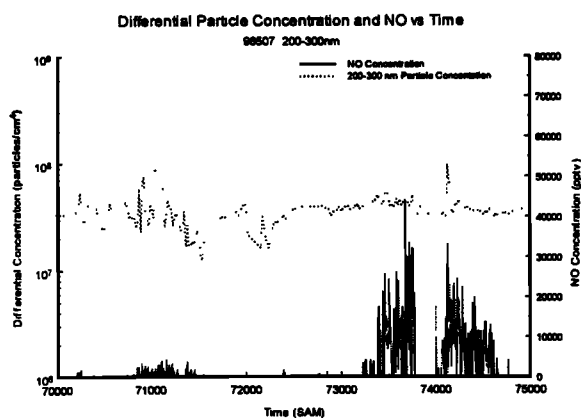


Figure 5. Differential concentration of 200–300nm particles during the May 7, 1996 incursions into the exhaust plume of the B757.

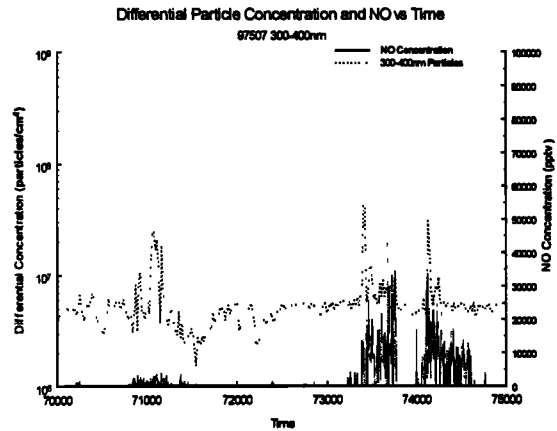


Figure 6. Differential concentration of 300–400nm particles during the May 7, 1996 incursions into the exhaust plume of the B757.

particles may be an efficient way of identifying aircraft exhaust plumes.

Fig. 5 shows the 200–300nm particle profile for a segment of the May 7, 1997 chasing of the NASA B757. The y-axis is the differential concentration of particles for a size bin. The actual concentration may be obtained by multiplying the differential concentration by the width of the size bin, which in this case is 100nm. For reference, the NO trace is also provided. There are two distinct types of encounters represented by this data. The first, typified by the large NO signal at 73000–74500, was an incursion into a newly formed exhaust plume, where only slight amounts of plume processing and dilution have occurred. The second type of encounter, as is found at 71000, was an incursion into an aged exhaust plume, where significant diffusion and processing has occurred. The implication of Figure 5 is that the large particle concentration provides a long lived tracer for aircraft emissions. Figure 6 presents the analogous data for 300–400 nm particulates, and the same trend is evident.

## Summary

Aerosol measurements were found to serve as excellent tracers for jet engine emissions into the atmosphere. Peak CN concentrations (peaks in the CN vs. time profiles) exceeded background levels typically by factors of five. Typical background volatility ratios (ratio of total particle concentration to non-volatile concentration) were less than 3:1, whereas ratios as high as 80:1 (high sulfur case) were observed in exhaust plumes. When total particle concentration and non-volatile concentration were available, the ratio of non-volatile particulates to total particulates was calculated and proved to serve as another excellent indicator for aircraft exhaust; additionally, it strongly anticorrelated with the NO concentration. In the fuel sulfur experiment, the sulfur was found to enhance the total particle population, while the nonvolatile population remained constant, supporting the idea of production of aerosols by gas-to-particle conversion.

Larger particles, those greater than 200nm, were also found to offer an efficient way to identify aircraft emissions. The large particle population of an aircraft exhaust plume was seen to persist in time. Thus, large diameter particles may offer a means for identifying aged exhaust plumes even after

the gaseous emissions have diffused to near background levels.

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