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Observations of particulates within the North Atlantic Flight Corridor: POLINAT 2, September-October 1997

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Abstract. This paper discusses particulate concentration and size distribution data gathered using the University of Missouri-Rolla Mobile Aerosol Sampling System (UMR-MASS), and used to investigate the southern extent of the eastern end of the North Atlantic Flight Corridor (NAFC) during project Pollution From Aircraft Emissions in the North Atlantic Flight Corridor/Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (POLINAT 2/SONEX) from September 19 to October 23, 1997. The analysis presented in this paper focuses on "the corridor effect," or enhancement of pollutants by jet aircraft combustion events. To investigate the phenomena, both vertical and horizontal profiles of the corridor, and regions immediately adjacent to the corridor, were performed. The profiles showed a time-dependent enhancement of particulates within the corridor, and a nonvolatile (with respect to thermal volatilization at 300°C) aerosol enhancement at corridor altitudes by a factor of 3.6. The southern extent of the North Atlantic Flight Corridor was established from a four flight average of the particulate data and yielded a boundary near 42.5°N during the study period. A size distribution analysis of the nonvolatile particulates revealed an enhancement in the <40 nm particulates for size distributions recorded within the flight corridor.

1. Introduction

Project Pollution From Aircraft Emissions in the North Atlantic Flight Corridor (POLINAT 2) was the European Union sponsored program to investigate the contribution from air traffic exhaust emissions to the composition of the lower stratosphere and upper troposphere at altitudes between 9 and 13 km within the North Atlantic Flight Corridor (NAFC) [*Singh et al.*, 1999; *Schlager et al.*, 1997]. Project Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (SONEX) was the NASA-sponsored project dedicated to investigating the NO_x budget, using tracer correlations ($H_2O/N_2O/CH_4/NMHC/CO_2$) with NO_x to identify surface sources, stratosphere sources, aircraft sources, and to additionally investigate the climatology of upper troposphere/lower stratosphere NO_x, O_3, with other ozone precursors and tracers in the North Atlantic. POLINAT 2 consisted of 13 experimental flights between September 13 and October 23, 1997, with an overlap region from October 14 to October 23 with project SONEX.

The University of Missouri-Rolla Mobile Aerosol Sampling System (UMR-MASS) was deployed on the POLINAT 2 experimental platform, a Dassault Falcon, stationed in Shannon, Ireland. The "corridor effect" has been investigated using the five POLINAT 2 flights where the Falcon flight plan called for a constant altitude profile anchored by vertical profiles over Shannon, and a target zone for each flight. The flights were numbered according to the POLINAT 2 program schedule. Details of the flight plan for each day are given in Table 1.

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2. Experiment

The UMR-MASS is a well established platform for the characterization of particulates in a diverse set of sampling conditions including flight campaigns, engine altitude test simulations, and ground-based measurements of jet engine exhaust [*Hagen et al.*, 1992,1994; *Whitefield and Hagen*, 1995; *Howard*, 1996; *Wey*, 1997; *Schumann*, 1997]. Recently, the UMR-MASS has been employed in airborne measurement campaigns at the cruise altitudes of commercial transport aircraft, in particular, the European Union sponsored flight campaign POLINAT 1 and the NASA sponsored flight campaign Subsonic Aircraft Control and Cloud Effects Special Study (SUCCESS) [*Paladino et al.*, 1998; *Hagen et al*, 1998; *Schumann*, 1997].

For project POLINAT 2, the MASS was configured to obtain total particle concentration with a 1 Hz resolution for particles with diameters >5 nm using condensation nucleus counters. Nonvolatile particle concentrations were similarly obtained with the thermal volatilization unit (previously described as a discriminator) set at 300°C, measured by thermocouple situated in the gas flow [*Paladino*, 1997; *Paladino et al*, 1998]. The thermal volatilization unit reduces incoming aerosol to their nonvolatile residues, that is

Table 1. Flight Plan Details

Flight/ Date	Flight Plan	Altitude, m
7 Sept. 18, 1997	Long-range flight from Shannon, Ireland, to 61°N vertical profiles	variable
8 Oct. 14, 1997	Long-range flight from Shannon, Ireland to Tenerife (28°N), constant altitude	8,500
9 Oct. 14, 1997	Long-range flight from Tenerife to Shannon, Ireland, constant altitude	10,600
11 Oct. 20, 1997	Long-range flight from Shannon, Ireland, to Tenerife (28°N), constant altitude	11,200
12 Oct. 20, 1997	Long-range flight from Tenerife to Shannon, Ireland, constant altitude	8,200

to the aerosol material that is nonvolatile at 300°C, measured by a thermocouple placed within the gas stream. Its destruction efficiency for sulfuric acid aerosol was measured to be 95% for particles initially with diameters of 50 nm. Size distributions were obtained by differential mobility analysis, using a differential mobility analyzer, with sample paths that either yielded a total or a nonvolatile particle size distribution.

For typical ambient concentrations, counting statistics yielded an uncertainty in the concentration measurement between 0.9% and 1.1%. The flow uncertainty is small, <1%, due to the use of a critical orifice with the critical pressure differential across the orifice maintained by an electronically controlled pump. The uncertainty in the counter efficiency, associated with effects such as particle size and pressure, is estimated through calibration experiments to be 10%. The probe enhancement uncertainty, due to inertial effects at the probe entrance, is estimated from fluid dynamics calculations to be 5%. The uncertainty due to diffusional losses associated with Brownian motion was calculated to be near 10%. This analysis yields an overall uncertainty in the UMR measurement of 15%. It is noteworthy that this uncertainty did not vary significantly with altitude. The size distributions are characterized by a 10% uncertainty in the width of the size bin due to the transfer function of the differential mobility analyzer.

3. Results

The data presented here have been analyzed specifically with respect to identifying corridor effects. Particle number concentrations are given with respect to the operating conditions of the MASS, i.e. ambient pressure and 20°C.

3.1. Altitude Profiles

Figure 1 represents the altitude profiles taken at 61°N, a latitude anticipated to be beyond the northern extent of the NAFC, and at 52.5°N over the Shannon Air Traffic Control(ATC) zone. The vast majority of air traffic flies between 9500 and 12000 m, or flight levels, 310-390. The vertical aerosol profile over the Arctic region (61°N) shows a nearly unperturbed distribution of total particles, while the profile over the Shannon region shows a two-fold average

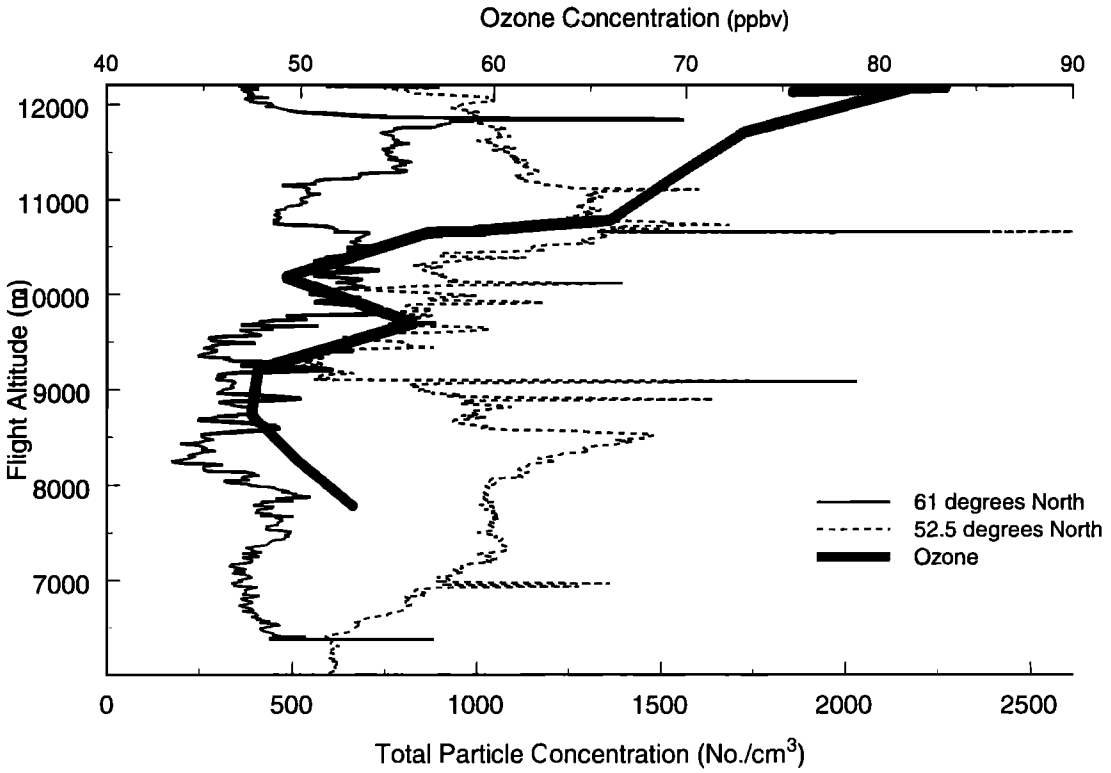

Figure 1. Altitude profiles of total condensation nuclie (CN) concentration at 61°N and 52.5°N during flight 7.

increase of particles in the flight zone. Total aerosol population can be a deceiving indicator of air traffic influence; as seen in Figure 1, there is an enhancement of the aerosol population below standard flight altitudes for the 52.5°N profile. This enhancement is unlikely to be due to aircraft emissions since it occurs well below typical flight altitudes and is clearly tropospheric in origin as indicated by the shape of the ozone profile plotted in Figure 1, where a sharp increase in ozone concentration is indicative of stratospheric penetration. This enhancement, however, may be a result of environmental processes, or uplift of aerosol from the boundary layer, thus contaminating the measurement with respect to combustion aerosols. The variability of total tropospheric aerosol concentration can be quite dramatic (ranging over 1 order of magnitude). Thus, for corridor studies, it is more meaningful to investigate the nonvolatile particulate contribution to the total aerosol population, as ambient cloud forming processes are more likely to produce volatile particulates. Additionally, studies made above the tropopause are less likely to be interfered with by uplift of maritime or continental aerosols. The variability of nonvolatile, stratospheric aerosols is quite low, though tropospheric folding during thunderstorm activity can cause localized perturbations. Figure 2 is a similar plot to Figure 1, but in this case the nonvolatile particle population for both altitude profiles are given. The particle populations are comparable with the exception of the flight zone where a factor of 2.2 enhancement is observed over Shannon. From 10,500 m to11,900 m, or flight levels 350-370, the nonvolatile particle population within the NAFC exceeds that measured at 61°N by a factor of 2:1.

In order to properly interpret data of this type, it is important that the meteorological history of the sampled air mass be understood. Meteorological data were provided by the Koninklijk Nederlands Meteorologisch Instituut (KNMI) and, are part of the POLINAT database [*Schumann*, 1997]. Backward trajectories indicate that the air parcel sampled during the arctic vertical profile likely had its origins in the Atlantic lower troposphere and the marine boundary layer below 700 hPa, and had been uplifted within the past 36 hours. Thus it is unlikely that the air parcel was contaminated with prior air traffic pollutants.

Figure 3 shows the nonvolatile altitude profiles recorded during the morning flight of October 14 (flight 8), with the first vertical profile near Shannon at the beginning of the flight, and the second near Tenerife at the end of the flight. The Falcon left Shannon at 0700 UTC, or 0600 local time, before the peak in eastbound traffic arrived in the Shannon ATC airspace. The profile shows a very low population density of particulates over Shannon throughout the profile. Figure 4 illustrates the nonvolatile altitude profiles recorded during the afternoon flight of October 14 (flight 9) when the Falcon left Tenerife at 1300 UTC arriving over Shannon at 1730 UTC, or 1830 local time. The two Tenerife profiles, separated by a 2-hour interval between measurements, remain relatively constant; but the Shannon profiles, separated by approximately 12 hours, with the second taken after the day's air traffic passed, showed a six-fold increase in the population density of nonvolatile particles at flight altitudes. These data provide evidence that the particle emissions from the day's air traffic, both westbound and eastbound, had accumulated at flight altitudes in the NAFC.

Again, by an analysis of the meteorological data provided by KNMI, the Shannon profile in Figure 3 is likely to have been an air mass that had originated in the Atlantic lower troposphere below 800 hPa within the past 36 hours. Trajectory calculations indicate that the Tenerife air mass likely originated from over the North African continental plains without profound vertical displacement. On the return trip, detailed in Figure 4, the Tenerife profile passed through several air parcels with widely divergent geographic origins which is quite common; most of the air parcels showed likely origins over the African continent, or over the Atlantic Ocean, without much vertical displacement. The final descent into

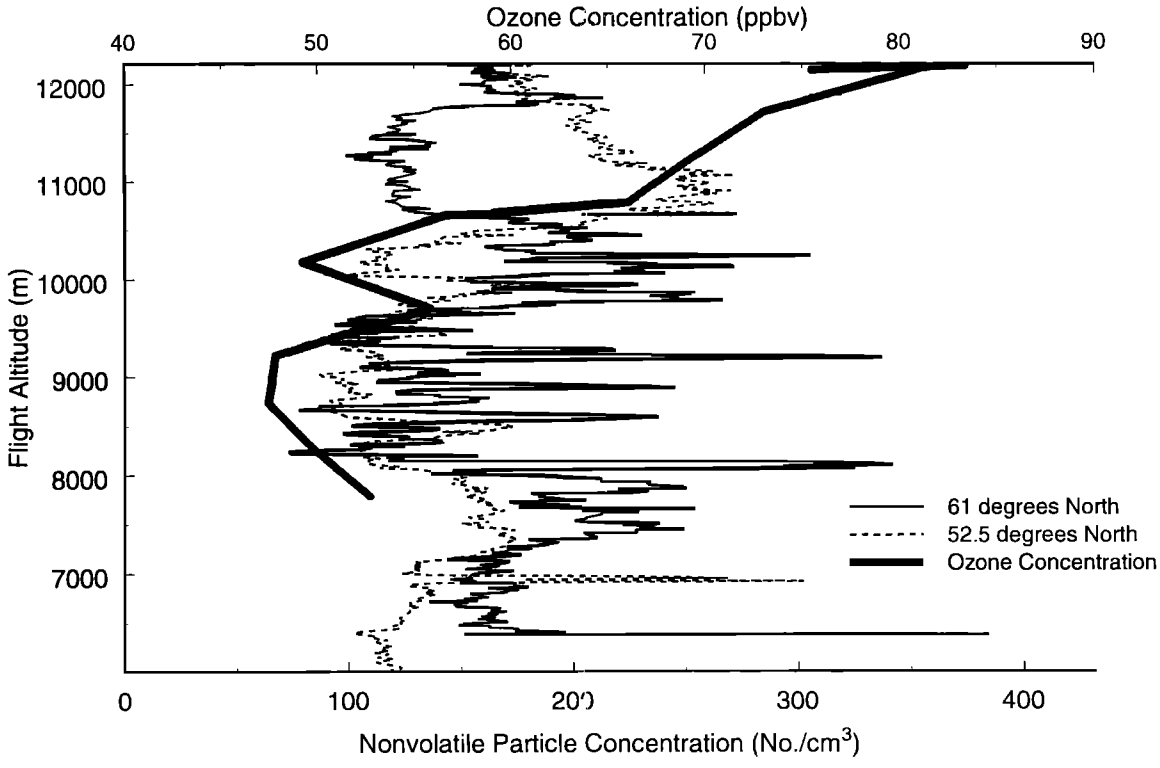

Figure 2. Altitude profiles of nonvolatile CN concentration at 61°N and 52.5°N during flight 7.

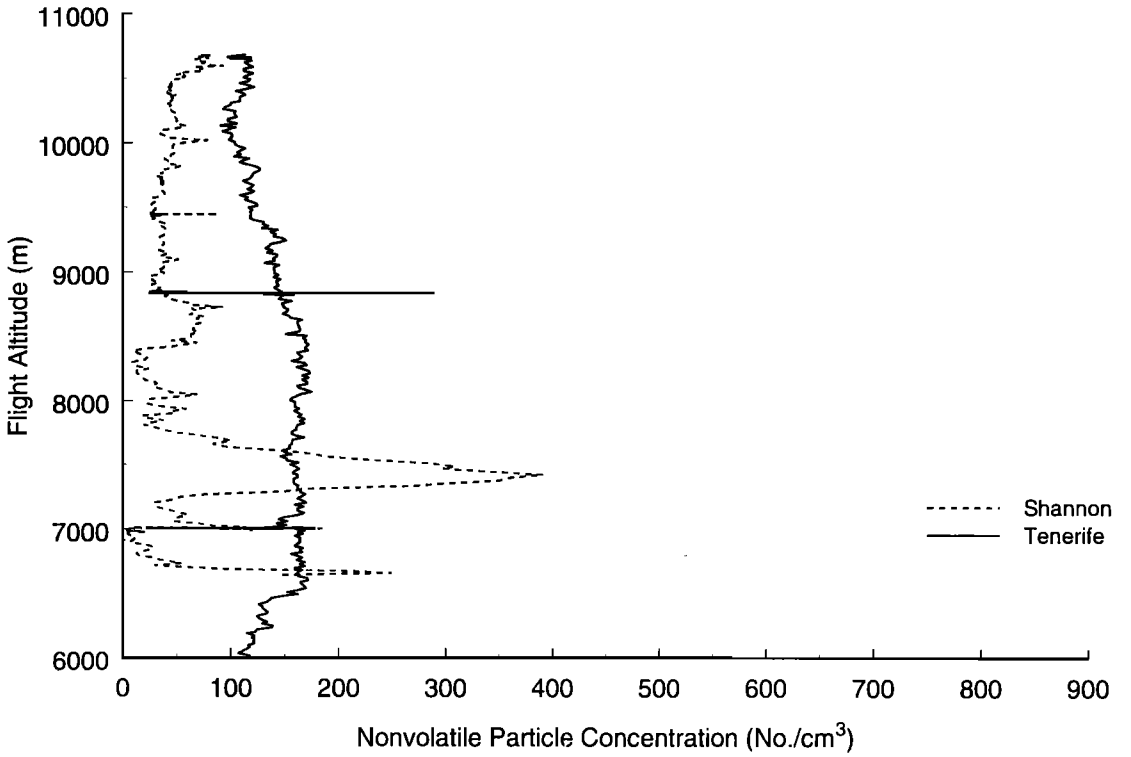

Figure 3. Altitude profiles of total CN concentration at 28°N and 52.5N during flight 8.

Shannon, as shown in Figure 4, passes through an air parcel that originates in the North Atlantic, near the coast of New Foundland, and had arrived into the Shannon region on an anticyclonic wind pattern from the northwest. Thus the two Tenerife profiles and the initial Shannon profile of Figure 3 would be expected to be free of air traffic contamination. Only the fourth, the profile on the return to Shannon, showed a likelihood of being exposed to previous air traffic emissions, both from aged and fresh emissions at corridor altitudes. During the POLINAT 1 campaign, nonvolatile particulate concentration measurements were made during a period

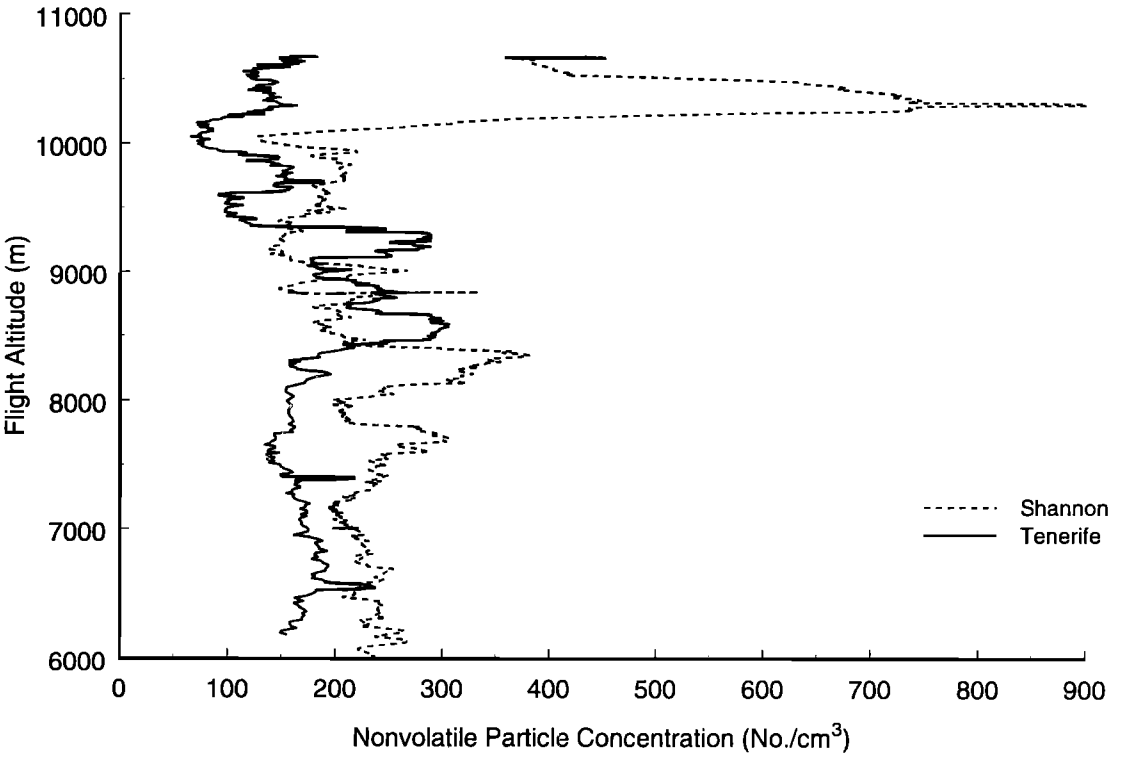

Figure 4. Altitude profiles of total CN concentration at 28°N and 52.5°N during flight 9.

of intense anticyclonic weather activity, and the buildup of nonvolatile particulates in the corridor background was observed [*Schumann*, 1997]. The concentration level, however, reached a maximum of 500 particles/cc after 7 days of accumulation. The observations of Flight 6 show measured concentrations as high as 900 particles/cm^3 which is indicative of fresh accumulation of emissions rather than aged background corridor emissions.

Figure 5 shows the NO gas concentration recorded during the flight 9 altitude profiles. Large ambient enhancement of NO within the NAFC is observed. This enhancement correlates well with the nonvolatile particulate data shown in Figure 4, and further supports the use of the nonvolatile particle data as a means of defining the vertical extent of the NAFC at the time of the measurement. The NO enhancement occurs at slightly lower altitudes than the particle enhancement as seen when Figures 4 and 5 are compared. This is not surprising since the transport and evolutional properties of gas phase species and particulates are different. Additionally, enhancements in the population of nonvolatile particles at 7000-8000 m often seen in the Shannon profiles are potentially due to the same process. No time- dependant data from altitudes below the corridor that could explain these effects were taken.

3.2. Corridor Profiles

Flights 8, 9, 11, and 12 crossed the NAFC from Shannon to Tenerife in an effort to profile the southern extent corridor. Each flight was performed at a constant altitude to a geographical location from within the NAFC (Shannon) to a location clearly beyond the NAFC (Tenerife).

The nonvolatile aerosol concentrations for each degree of latitude were used to iteratively calculate the extent of the North Atlantic Flight Corridor, by using the ratio of nonvolatile particulates in/out of corridor to determine a maxima.

Figure 6 shows the calculated "in-corridor"/"out of corridor" ratio values as a function of latitude measured at a constant flight level for the flight data acquired during flights 8, 9, 11, and 12. Flight 9 at flight level 350 (10,600 m) shows the clearest enhancement, with a peak value of 2.8 at 42.5°N. It is of interest to note, that when the constant altitude NO data are averaged in a similar manner, the corresponding peak is shifted to 35.5°N with a ratio value of 2.0, again indicating a difference in the transport and aging processes of gas phase species as opposed to the particulate species.

From a meteorological standpoint, the air masses were quite ideal for this study. During flights 8 and 9, backward trajectory calculations indicate that the air parcels had possible origins between 18°-22°N and 25°-60°W with no strong vertical displacements and no boundary layer uplifts of note. The wind field for the day was anticyclonic around a high-pressure cell located near 40°N and 20°W with wind speeds near to 40 m/s as observed at 250 hPa. This pattern was stagnant for the 12-hour duration of the two flights. Flights 11 and 12 were made in slightly less ideal conditions. The air masses examined in flight 11 had most likely originated in the Atlantic lower troposphere at 18°N and 40°-60°W with no pronounced change in altitude over the past 24 hours. The air masses sampled on the return trip as flight 12 were likely uplifted within the past 48 hours from between 10°-20°N and 30°-70°W, those later in the flight had trajectories

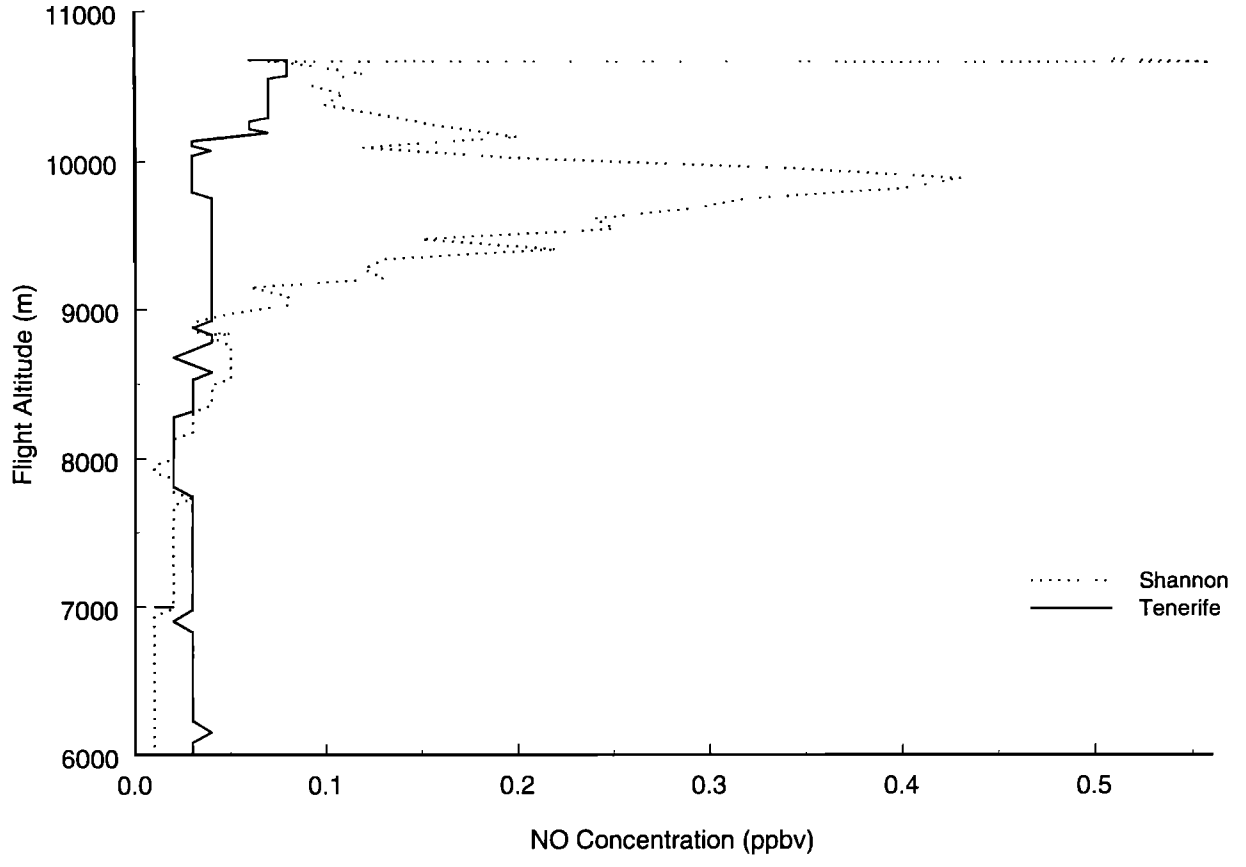

Figure 5. Altitude profiles of NO concentration at 28°N and 52.5°N during flight 9.

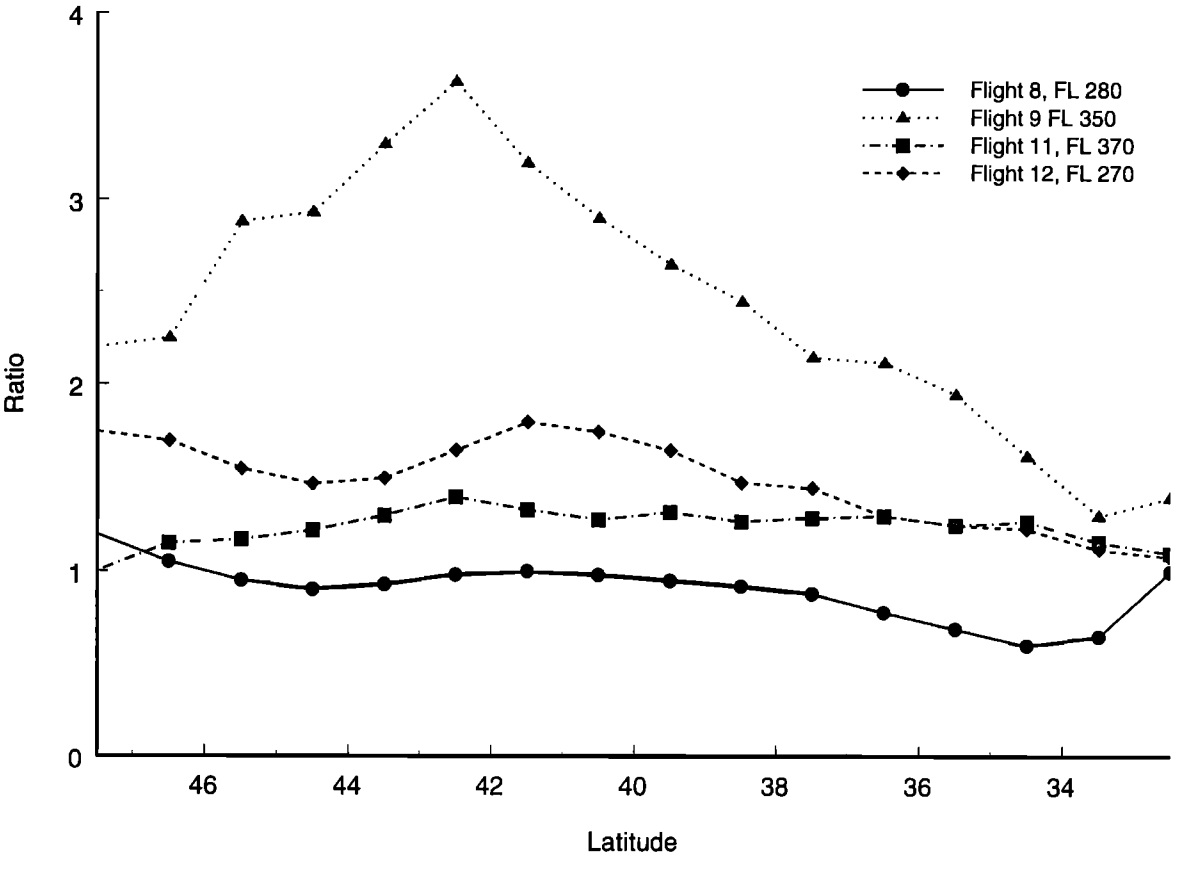

Figure 6. In-corridor/out of corridor nonvolatile CN ratios for various latitudes and flight levels.

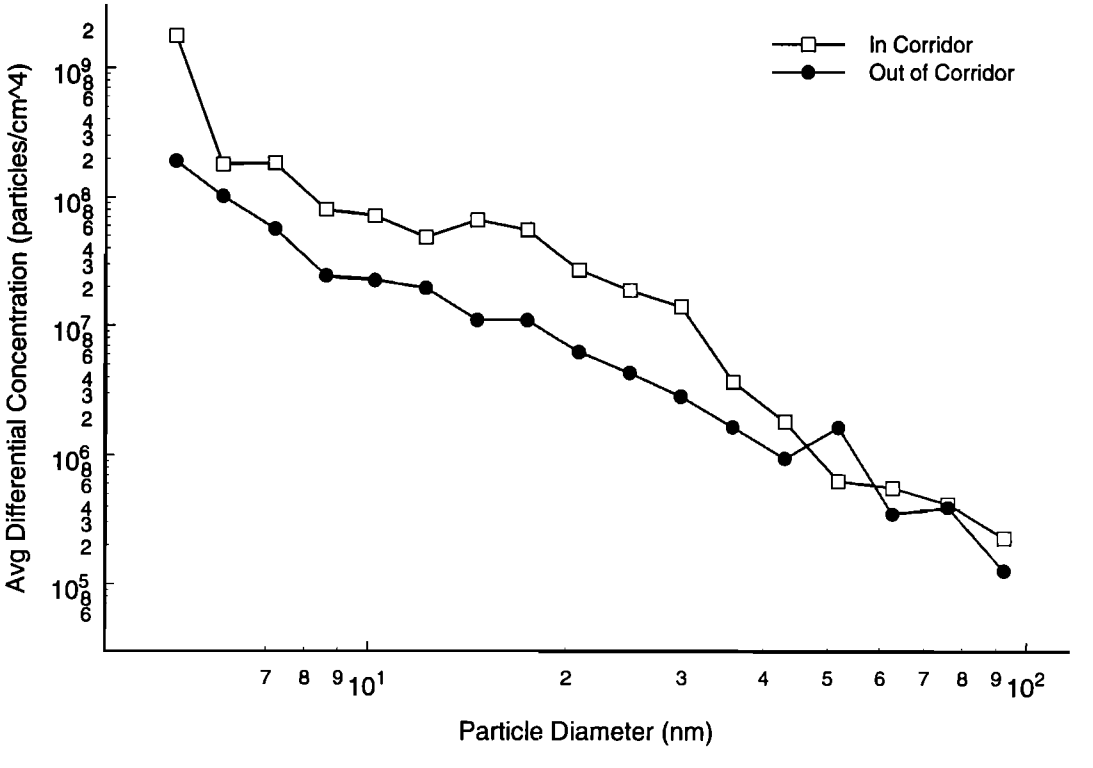

Figure 7. Mean non-volatile CN size distributions in/out of corridor for flight 9 at a constant flight altitude 10,600 m/35,000 feet.

from the Atlantic marine boundary layer. The wind field for the flight exhibited a strong jet stream pattern with the winds moving to east-northeast across most of the flight track at 60 m/s for the majority of the flight. The wind field near the Shannon region originated from an arctic low-pressure cell and were blowing to the southwest at 30 m/s.

For Figure 6 this lets us understand that the data represented for flights 8 and 9 show a relatively undisturbed air mass with little boundary layer contamination, that is no uplift. Since the data represented by flight 9 are of prime interest at a heavily traveled corridor altitude, this data represent an almost ideal set for interpreting corridor particulate levels and corridor extent. The data from flights 11 and 12 could be a little more difficult to interpret considering the nature of the air masses interrogated and the conflicting wind fields; however, the flight levels were outside those most frequently traveled, and the concentrations were relatively low in reference to the other data gathered in flights 8 and 9.

3.3. Size Distributions

A size distribution analysis was performed on the data acquired during the constant altitude portion of flight 9, and flight 11. Figure 7 shows the averaged size distributions for the two classifications of size distributions, that is, in-corridor and out of corridor as determined by the discussion above. There is a noticeable corridor-based particle concentration enhancement for particle diameters <40 nm, where the enhancement expressed as the ratio of in/out of corridor measurements can be as great as a factor of 6. This size-dependant enhancement observed within the corridor is consistent with the size-related soot emission characteristics of a typical jet engine, which emit the bulk of their particulates at sizes below 50 nm [*Schumann*, 1997; *Hagen et al.*, 1998; *Paladino et al.*, 1998; *Petzold et al.*, 1999; *Petzold and Shroder*, 1998a; *Rickey*, 1995; *Whitefield et al.*, 1996; *Whitefield* and *Hagen*, 1995].

Figure 8 shows the average size distributions acquired during the constant altitude portion of Flight 11 at 11,300 m. In this case, there is no significant corridor-based enhancement within the uncertainty of the measurements, and both distributions correlate well with the "out of corridor" data for flight 9. Analysis of the size distributions for flights 8 and 12 yields similar results. The constant altitude data in flights 8, 11, and 12 were all beyond the vertical extent of the NAFC. (See section 1.) The results from Figure 7 offer a new means of possibly identifying regions of airspace containing jet engine combustion particulates by using <40 nm nonvolatile particulates as a tracer species.

4. Conclusions

During the POLINAT 2/SONEX campaign, observations of the nonvolatile, particle population in the upper troposphere/lower stratosphere through altitude profiling yield evidence of particle loading in the commonly used flight altitudes. These profiles define the vertical extent of the corridor with respect to particle enhancement, and show the corridor to have a time dependence with respect to particle enhancement on the day of measurement which synchronizes well with air traffic in the flight corridor. Analysis of the southern extent of the NAFC yielded a southern boundary at latitude 42.5°N at the time the measurements were made. The altitude profile particle enhancement and daily peaks in eastbound and westbound traffic in the North Atlantic Flight Corridor correspond well. Long-range, constant altitude profiling

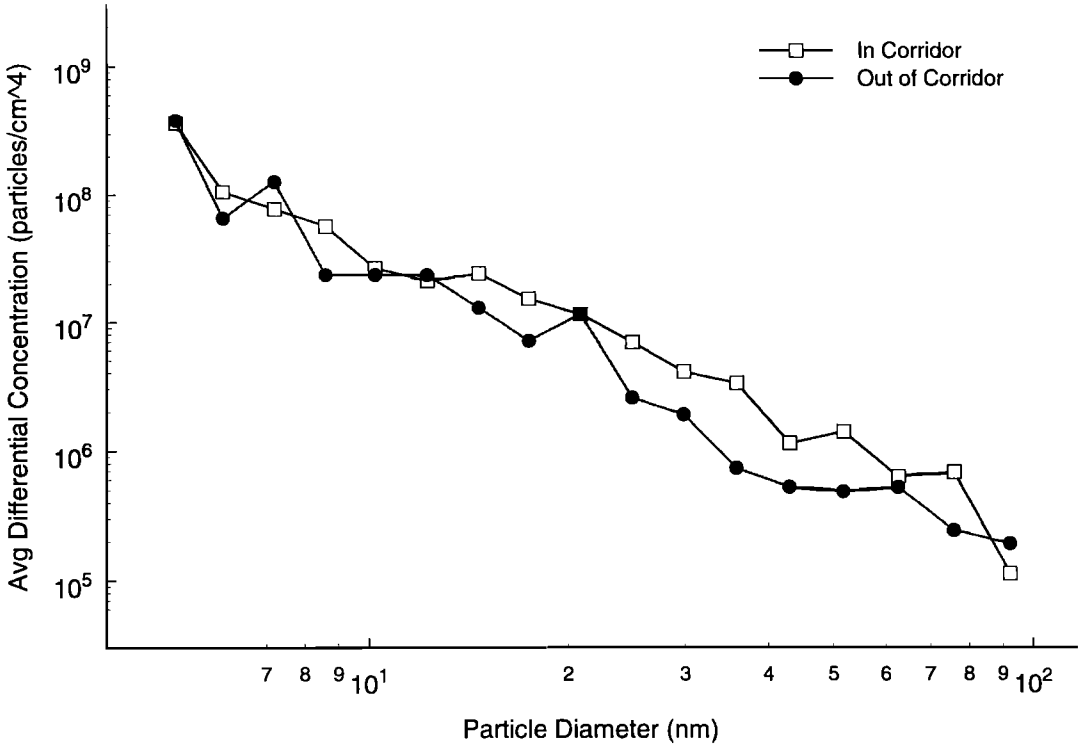

Figure 8. Mean nonvolatile CN size distributions in/out of corridor for flight 11 at a constant flight altitude 11,200 m/37,000 feet.

flights that extend from within the NAFC to latitudes well south of its southern flight tracks yield a nonvolatile particulate enhancement factor of 3.6 within the flight corridor at flight level 350. A comparative analysis of the size distributions of in-corridor and out of corridor air demonstrate that particle concentrations for sizes <40 nm were particularly enhanced in comparison to all other diameters examined. This size-dependant enhancement observed within the corridor is consistent with the size-related soot emission characteristics of a typical jet engine. An analysis of the meteorological conditions under which the observations were obtained was facilitated by the back trajectory calculations and satellite observations of the Dutch Meteorological Institute (KNMI). The vertical profiles were shown by meteorological backward trajectory analysis to have likely occurred primarily in recently uplifted air masses that were unlikely to contain prior aged air traffic emissions, and those air masses sampled during the long-range horizontal profiling used to determine the southern extent of the corridor also showed little prior contamination, though wind fields in the latter flights were less than ideal for comprehensive analysis. Thus, the meteorological conditions support the analysis of the corridor extent described in this paper.

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