Atmospheric chemical transport based on high-resolution model-derived winds: A case study

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Abstract. Flight 10 of NASA’s Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (SONEX) extended southwest of Lajes, Azores. A variety of chemical signatures was encountered. These signatures are examined in detail, relating them to meteorological data from a high-resolution numerical model having a horizontal grid spacing of 30 and 90 km with 26 vertical levels. The meteorological output at hourly intervals is used to create backward trajectories from the locations of the chemical signatures. Four major categories of chemical signatures are discussed: stratospheric, lightning, continental pollution, and a mixed chemical layer. The strong stratospheric signal is encountered just south of the Azores in a region of depressed tropopause height. Three chemical signatures at different altitudes in the upper troposphere are attributed to lightning, Backward trajectories from these signatures extend to locations of cloud-to-ground lightning. Specifically, results show that the trajectories pass over regions of lightning 1–2 days earlier over the eastern Gulf of Mexico and off the southeast coast of the United States. The lowest leg of the flight exhibits a chemical signature consistent with continental pollution. Trajectories from this signature are found to pass over the highly populated Northeast Corridor of the United States. Surface-based pollution apparently is lofted to the altitudes of the trajectories by convective clouds along the East Coast that did not contain lightning. Finally, a mixed layer is described. Its chemical signature is intermediate to those of lightning and continental pollution. Backward trajectories from this layer pass between the trajectories of the lightning and pollution signatures. Thus they likely are impacted by both sources.

1. Introduction

NASA’s Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (SONEX) was conducted over the North Atlantic Flight Corridor (NAFC) during fall 1997 to study the effects of subsonic aircraft emissions on the atmospheric chemistry of the upper troposphere and lower stratosphere. During the 16 flights comprising the SONEX field phase, NASA’s instrumented DC-8 aircraft collected high resolution in situ chemical data throughout the NAFC and surrounding areas. Singh et al. [1999] give a comprehensive overview of the SONEX program.

SONEX flight 10 on October 29, 1997, was a tropical survey originating at Lajes, Terceira Island, Azores (38.7°N, 27.1°W). The flight track was oriented northeast to southwest, reaching a southwestern terminus near 19.9°N, 37.1°W (Figure 1). A stack maneuver of four legs was performed between the southernmost latitude and ~25°N to sample chemical characteristics at multiple levels. Although the goal of flight 10 was to sample clean, subtropical air exhibiting minimal continental, convective, and aircraft influence, complex chemical signatures from a variety of sources actually were encountered.

Insight into the transport history and origins of air parcels can be gained by relating meteorological data to chemical data. Species-species correlations are useful since they can indicate the types of atmospheric processes that air samples have experienced, for example, convection, pollution, mixing, and photochemical reactions [Thompson et al., 1999; D. D. Davis et al., unpublished manuscript, 1999]. Trajectory calculations can indicate possible source regions, transport paths, and age of the air [e.g., Board et al., 1999]. Origins and transport paths of atmospheric chemicals over the North Atlantic Ocean are not easily investigated. This is due to the sparsity of meteorological data over oceanic regions, as well as the vast distances that air parcels can travel over short periods of time in the midlatitudes. Transport paths and, consequently, the chemical composition of air in this region can vary greatly, particularly during the cool season due to strong winds and high-amplitude flow patterns.

This research relates observed chemical signatures during SONEX flight 10 to high-resolution numerically derived meteorological data. The signatures sometimes are inconclusive. Thus our objective is to better understand the chemical signatures by determining source regions and transport paths of the air parcels that were sampled. We use the National Center for Atmospheric Research (NCAR)/Penn State Fifth Generation Mesoscale Model (MMS) [Grell et al., 1994] to create a high-resolution three-dimensional set of meteorological data. The model-derived winds then are used to calculate backward tra-
jectories from the observed chemical signatures. Finally, the trajectory paths are related to meteorological and/or anthropogenic chemical sources.

We believe that our high-resolution MM5-derived meteorological data (30 or 90 km, hourly) provide an accurate representation of the actual wind regimes, and at a higher temporal resolution than is possible using global gridded analysis (typically 6-hourly). This improved resolution is important since trajectories are highly sensitive to the temporal frequency of the wind data [Doy and Perkey, 1993] as well as small errors in wind direction and speed. Other studies that have used high-resolution numerically derived meteorological data to investigate aircraft-derived chemical data include Chatfield et al. [1996, also unpublished manuscript, 1999], Pickering et al. [1996, also unpublished manuscript, 1999], Liu et al. [1996], and Wang et al. [1996].

2. Data and Methodology

2.1. Meteorological Data

We employed a two-way interactive nested configuration for the nonhydrostatic MM5 simulations (Figure 2). The coarse outer grid extended far beyond the SONEX region, covering most of North America and Europe, to reduce the effects of boundary errors [Warner et al., 1997]. The horizontal spacing of the coarse grid was 90 km, with 147 grid points in the east to west direction and 87 grid points south to north. The higher-resolution inner grid, centered over the path of flight 10, had a horizontal spacing of 30 km. This grid contained 136 points east to west and 166 grid points south to north. Both domains utilized 26 sigma levels (up to 10 hPa) in the vertical that were relatively closely spaced near the tropopause (~20 hPa separation). The Grell cumulus parameterization scheme [Grell, 1993] and the Blackadar planetary boundary layer scheme [Zhang and Anthes, 1982] were chosen as the model physics parameterizations.

The MM5 was initialized with a gridded data set prepared by the European Centre for Medium-Range Weather Forecasting (ECMWF) [Bengtsson, 1985; Hollingsworth et al., 1986]. Boundary conditions at subsequent times also were obtained from the ECMWF data. Sea surface temperatures were taken from a data set provided by the National Center for Environmental Prediction (NCEP). The 67-hour simulation on the 90 km domain began at 0000 UTC October 27, 1997, and ended at 1900 UTC October 29. The simulation for the 30 km domain was started at 0000 UTC October 28 and also ended at 1900 UTC October 29. Time steps for integrations on the 90 and 30 km grids were 60 and 20 s, respectively, with information passing between the two grids at 60-s intervals. Four-dimensional data assimilation was not employed. Model data from the first 12 hours of each simulation were not utilized to allow for a sufficient "spin-up" period.

Trajectories were calculated from the MM5 data using the kinematic method. That is, air parcels were advected using only the three-dimensional wind field, without employing the isentropic assumption. Fuelberg et al. [1996a] give further details about the trajectory model as well as a comparison between the kinematic and isentropic methods.

Limitations of trajectories include incorrect placement of meteorological features by the input analyses. This is of particular concern over oceanic regions, such as the SONEX domain, where meteorological data are relatively sparse. Trajectories also are subject to the resolution of the wind data, since features of sufficiently small timescales or space scales will not be resolved [Doy and Perkey, 1993]. Since the resolution in our two domains was 1 hour and either 30 or 90 km, individual convective elements (e.g., their updrafts) are not resolved. However, the effects of the convection are depicted at the scale of the grid. Finally, numerical limitations of the kinematic model itself affect overall trajectory accuracy [Stohl et al., 1995]. The kinematic method has been widely used in many meteorological/chemical applications [e.g., Draxler, 1991; Doy and Perkey, 1993; Krishnamurti et al., 1993; Chatfield et al., 1996; Garstang et al., 1996; Bieberbach et al., this issue].

Backward trajectories were calculated along the flight path at intervals of 1-min flight time (~120–150 km intervals, depending on aircraft speed). The trajectories, derived from a combination of the 30 and 90 km MM5 wind data, arrived at the hours closest to the actual times of the in situ DC-8 chemical observations. Specifically, the first 19–29 hours from the flight track were calculated from the 30 km (innermost) data grid, depending on time of arrival at the boundary of the 30 km domain. The earlier 24–34 hours were calculated using the 90 km data. The high frequency at which trajectories were released (1-min intervals) effectively eliminated the need for employing a clustering or ensemble method [e.g., Merrill et al., 1985; Kahl, 1993; Fuelberg et al., 1996b].
Locations of convection were determined using cloud-to-ground lightning data from the National Lightning Detection Network (NLDN) and Long Range Field (LRF) [Cummins et al., 1998]. GOES-8 infrared imagery also was employed.

2.2. Chemical Data

The chemical data used in this study were taken from a 1-min merged data set prepared at Harvard University. This data set contains 1-min averages of all chemical, meteorological, and position data recorded during the flight, as well as model-derived values of certain chemical species. Singh et al. [1999] describe the chemical measurements in detail.

3. Meteorological Conditions

High-amplitude flow patterns dominated the SONEX period, including the day of flight 10 [Fuehrberg et al., this issue]. Figure 3 depicts winds and potential vorticity at 250 hPa (~10.5 km) on 1000 UTC October 29 derived from 1° × 1° ECMWF data (Figures 3a and 3b) and the 90 km MM5 simulation (Figures 3c and 3d).

Circulation patterns from the MM5 (Figure 3c) agree closely with those from ECMWF (Figure 3a), particularly the trough/ridge system near the NAFC. The cutoff low near the Azores also is similarly placed, although wind speeds (not shown) from the MM5 are slightly greater (~10 m s⁻¹) than those in the ECMWF analyses. The departure site of flight 10 (the Azores) was nearly coincident with this cyclone that had been drifting slowly eastward after becoming detached from the main flow. The long wave trough and ridge axes (solid lines) located south and west of the low, respectively, also are depicted similarly. The relatively deep trough south of Newfoundland had been oriented over the southeastern United States 2 days prior to the flight. This system and its associated frontal boundaries produced extensive lightning from the central Gulf of Mexico to the coastal waters off the eastern United States.

Potential vorticity (PV) (Figure 3b and 3d) provides a rigorous comparison between the ECMWF and MM5 data since PV requires derivatives of temperature and the horizontal wind components. The two versions of PV at 250 hPa exhibit close agreement; that is, major features are similarly placed with only minor differences in magnitude. The MM5 generates an 11 PVU (1 PVU = 1 × 10⁻⁵ Kmbar s⁻¹) maximum close to the Azores, compared to the 12 PVU maximum indicated by the ECMWF analysis. The two PV maxima located northeast and northwest of the main feature also are comparably represented in terms of magnitude, with variations of only 1 PVU.

Although only two parameters at 250 hPa are described here, we have compared the MM5 and ECMWF versions of many meteorological variables at various altitudes. Close agreement is found in all cases. Therefore we believe that the MM5 simulation can be used with confidence to understand the chemical signatures observed during flight 10. Unless otherwise noted, future meteorological analyses will be derived from the MM5.

4. Results

Results show that the complex chemical signatures observed during flight 10 can be directly linked to the meteorological features described in section 3. The sampling of stratospheric air during flight 10 was one of the most extensive of the entire SONEX mission [Thompson et al., 1999]. The convective influence also was pronounced (G. Chen et al., unpublished manuscript, 1999; D. D. Davis et al., unpublished manuscript, 1999), and a moderate amount of continental pollution was encountered.

Three types of chemical signatures were observed during flight 10, and correlations between NOx and CO and between NOx and ultrafine condensation nuclei (CN) are useful in differentiating between them [e.g., Thompson et al., 1999; Jaeglé et al., 1998]. Figure 4 shows scatter diagrams of these three species during the flight. Stratospheric air is easily distinguished from lightning- and continental air since NOx is anticorrelated with both CO and CN in the stratosphere. Large NOx, associated with moderate CO and large CN indicates lightning. Conversely, moderate NOx with moderate CO and CN denotes a continental influence. Stratospheric and tropospheric regimes can be further distinguished by the probability distribution of ozone mixing ratio (Figure 5). Although most ozone measurements during flight 10 are less than 100 ppbv, numerous samples exceed the often accepted 100 ppbv stratospheric threshold [e.g., Thompson et al., 1999], with some approaching 450 ppbv.

Figures 6 and 7 contain time series of selected chemical species measured along the path of flight 10. Chemical signatures are labeled along the tops of the figures. The bottom plot of each figure depicts flight altitude. Plate 1 shows the altitude and latitude of the flight as well as the chemical signatures.

Four distinct chemical signatures are suggested during the flight: two encounters of stratospheric air (STR), three lightning signatures (LT1, LT2, and LT3), one signature representing continental pollution (POL), and a broad mixed region along two flight legs (MIX). The following sections present the rationale for identifying each of these signatures based on its chemical characteristics and backward trajectories from it. The two stratospheric portions of the flight are examined first. Next, we explore the three signatures associated with lightning and then the pollution signal. Finally, the mixed region is examined.

4.1. Stratospheric Signature

Well-defined stratospheric air is encountered during flight 10. Stratospheric penetration occurs on both the outbound and inbound legs when the DC-8 passes through the cutoff low near the Azores (Figure 3). During the outbound penetration the stratospheric signal occurs between 1128 and 1214 UTC (Plate 1 and Figures 6 and 7), that is, as the aircraft ascends to 10.1 km (267 hPa). Values of O3 and NOx increase sharply to ~250 ppbv and 1000 parts per trillion by volume (pptv), respectively. Conversely, values of CO (<40 ppbv), ultrafine CN (<1000 cm⁻³), and water vapor (<100 ppmv) show dramatic decreases, while NO (<100 pptv) and peroxyacetyl nitrate (PAN) (~40 pptv) decrease to a small fraction of NOx. Stratospheric penetration during the return leg reaches a higher altitude than before (11.9 km (202 hPa)) between 1720 and 1825 UTC, producing an even greater value of ozone (~400 ppbv).

The tropopause is relatively depressed near the cutoff cyclone. This depression is depicted in the time series (Figure 7) and cross section of PV (Figure 8). If we assume that the stratospheric threshold is 3.0 PVU [Fuehrberg et al., this issue], the lowest altitude of the stratosphere is ~7 km. The differential absorption lidar (DIAL) [Dowrell, 1989, 1991] ozone image (Plate 2) shows the downward protrusion of ozone-rich stratospheric air. Specifically, O3 > 100 ppbv extends to ~6 km during the return portion of the flight (right side of plate), with
values >80 ppbv reaching nearly 3 km. Backward trajectories calculated from the stratospheric portion of the flight (not shown) remain at stratospheric altitudes throughout their 2-day histories.

4.2. Lightning Signatures

Three regions exhibiting elevated convection-related species of varying intensity and duration are attributed to lightning. Values of NO, NO₂, H₂O, and ultrafine CN all are enhanced in these three regions. However, other tracers (e.g., peroxides and hydrocarbons) are inconclusive in terms of indicating convection. For this reason, this section utilizes the formerly stated species as the main convective tracers. It should be noted that our use of the terms “enhanced” and “elevated” refers to comparisons with other segments of flight 10, not all SONEX flights. A major objective of this section is to use backward
Figure 4. (a) Correlation plot of NOx (pptv) versus CO (ppbv). The three main signatures of flight 10 are labeled. (b) Correlation plot of NOx (pptv) versus the logarithm of ultrafine CN (cm⁻³). The three main signatures of flight 10 are labeled.

trajectories to support our contention that these chemical signatures represent lightning.

The longest chemical signature attributed to lightning (LT1) occurs from 1404 to 1452 UTC (Figures 6 and 7). One should note that the bounds of LT1 encompass the entire flight leg at 9.5 km (292 hPa, Plate 1). The chemical signature of LT1 includes elevated mixing ratios of NO (300-500 pptv), NOy (500-1000 pptv), and water vapor (200-300 ppmv), as well as elevated ultrafine CN (10,000-17,000 cm⁻³). However, CO is slightly reduced (~80 ppbv), and other chemical signals expected to accompany convective transport are not elevated [Thompson et al., 1999; D. D. Davis et al., unpublished manuscript, 1999].

Figure 9 shows a horizontal plot of backward trajectories from flight locations comprising LT1. The trajectories pass over the Florida peninsula approximately 2 days prior to arriving at the DC-8, travel offshore of the Carolinas as they head northeast toward Newfoundland, and then turn southeast toward the flight track. To determine the extent to which parcels comprising LT1 were exposed to lightning, we superimpose trajectory and NLDN cloud-to-ground lightning data over 3-hour intervals. Although intracloud lightning also produces nitrogen oxides, no data about its occurrence were available at the needed resolution. The trajectories from LT1 experience two major encounters with cloud-to-ground lightning. The first occurs between 1200 and 1800 UTC October 27 (Figures 10a and 10b) as they pass through the eastern Gulf of Mexico into Florida. The greatest exposure occurs between 1200 and 1500 UTC when the trajectories are nearly coincident with an area of concentrated cloud-to-ground flashes. Heights of individual trajectories during this encounter (50-51 hours back, Figure 11) vary considerably, with the highest trajectories near 9.8 km and the lowest near 6.0 km. Satellite imagery (not shown) indicates cloud top temperatures in this area that correspond to cloud heights of ~9-12 km (~300-200 hPa).

The trajectories from LT1 experience a second major encounter with lightning approximately 12 hours after the first.
Figure 7. Time series of data along flight 10. Concentrations of ultrafine CN (cm$^{-3}$), mixing ratios of PAN (pptv) and H$_2$O (ppmv), and ECMWF-derived potential vorticity (PVU) along the flight track are given in the first four panels. The bottom panel indicates the altitude of the DC-8. Chemical signatures are indicated on the top panel (STR, stratospheric; MIX, mixed; LT, lightning; POL, pollution). Arrows indicate the location of brief lightning events (spikes) at the end of the stratospheric leg and within the mixed layer as well as LT2.

This occurs between 0600 and 0900 UTC October 28 as the trajectories exit the northern edge of the flash region. Maximum cloud heights in this area (38–42 hours back, Figure 11) are ∼12 km (200 hPa), while trajectory heights have risen to ∼7.0–11.5 km (213–473 hPa).

Figure 8. Cross section of MM5-derived potential vorticity (PVU) through the cutoff low-pressure system at 1200 UTC October 29, 1997. Altitude (km) and horizontal distance are indicated. The cross section axis is shown in Figure 3d.

Figure 9. Backward trajectories from LT1 (50–51 hours back). Arrival time at flight level is 1400–1500 UTC October 29, 1997. Arrows along trajectory paths indicate locations at 24-hour intervals. The flight track is indicated by the thin line.

It is informative to examine signature LT2 in the context of the LT1 lightning signature just described. LT2 is brief, extending only from 1322 to 1331 UTC (Figures 6 and 7). It occurs as the DC-8 ascends (Plate 1) from a leg at 8.8 km (319 km)
Plate 1. Altitude profile of SONEX flight 10 as a function of latitude. Chemical signature types are indicated by colored segments as follows: red, stratospheric; blue, lightning; orange, pollution; green, mixed. Individual signatures are labeled in black. Brief lightning signatures (spikes) within the mixed layer and at the end of the stratospheric leg are denoted by ellipses.

Plate 2. Cross section of DIAL-derived O₃ mixing ratio. The O₃ scale (ppbv) and time (UTC) are given at the top, while latitude and longitude are given at the bottom. Altitude in kilometers is given along the sides.
hPa) to a leg at 10.7 km (243 hPa). Specifically, LT2 begins when the DC-8 ascends through the altitude of LT1 (9.5 km, 292 hPa) in a location directly above LT1. Values of NO, NOy, CN, CO, and water vapor for LT2 are very similar to those observed during LT1. In addition, backward trajectories from LT2 (Figure 12) have lightning encounters that are very similar in both time and space to the LT1 trajectories (Figure 10). Thus LT2 appears to be a quick sampling of the same lightning event that produces LT1.

The LT2 signature ends part way through the 10.7 km leg at ~1331 UTC (Figures 6 and 7 and Plate 1). This ending corresponds to an increase in O₃ to ~90 ppbv, and a decrease in H₂O to values similar to those encountered in the stratosphere. The DIAL image (Plate 2) indicates a downward protrusion of relatively large O₃ in the region, and the horizontal analysis of PV (Figures 3b and 3d) shows that this location (~23°N, 33°W) lies along a narrow band of elevated PV. The time series of flight level PV (Figure 7) indicates that values in this area increase to ~3.0 PVU. These factors suggest that the region exhibits a weak stratospheric influence. The only counter indication is that CO does not show the decrease that is expected of stratospheric air.

The third signature attributed to lightning, LT3, occurs between 1646 and 1716 UTC as the DC-8 heads back to the Azores after the stack maneuver. The signature begins at an altitude of 10.3 km (245 hPa) as the aircraft ascends from the 5.8 km (486 hPa) leg to the 11.9 km (202 hPa) leg (Plate 1). The signal continues along the 11.9 leg until the beginning of the second stratospheric penetration (section 4.1). The chemical signature of LT3 is much stronger than those of LT1 and LT2 (Figures 6 and 7). LT3 exhibits the greatest NO (1500 pptv) and NOy (2000 pptv) of the entire flight. Values of ultrafine CN (30,000 cm⁻³) also are greatest in this segment, while CO is relatively small (~80 ppbv).

Backward trajectories from the level leg of LT3 at 11.9 km are shown in Figure 13. Their paths are not greatly different from those comprising LT1 (Figure 9). However, even the slight differences cause the LT3 trajectories to coincide more closely with the cloud-to-ground lightning locations and for a longer period of time than those of the other lightning signatures. These lightning encounters are shown in Figure 14. The event begins near 1500 UTC October 27 (Figure 14a) as the trajectories first reach the southwestern edge of an extensive line of intense convection over the Gulf of Mexico. This encounter region, immediately north of the Yucatan Peninsula, contains the highest cloud tops of the entire line (≥12 km, ≤200 hPa). At this initial time, only the southernmost trajectories coincide with the region of cloud-to-ground lightning. However, as the trajectories move eastward, their horizontal separation decreases, leading to a greater number of trajectory/lightning encounters. Thus, between 1800 and 2100 UTC (Figure 14c), a significantly greater number of trajectories is colocated with lightning. This first encounter ends at ~2100 UTC October 27, and over the next 12 hours (not shown), the number of lightning flashes decreases greatly. The trajectories pass over South Florida and emerge over the coastal waters of the Atlantic during this period.

Cloud-to-ground lightning flashes increase dramatically over the western Atlantic Ocean after 0300 UTC October 28 (not shown), and the eastward moving trajectories pass through the southern part of this region between 0900 and 1500 UTC October 28 (Figures 14d and 14e). One should note that almost all of the trajectories pass through the area of intense lightning at some time during this period. Over this 6-hour span, trajectory heights remain virtually constant near 12 km.
Figure 13. Lightning event 3. Backward trajectories from LT3 (53 hours back). Arrival time at flight level is 1700 UTC October 29, 1997. Arrows along trajectory paths indicate locations at 24-hour intervals. The flight track is indicated by the thin line.

(200 hPa, not shown), corresponding approximately to cloud top level.

It should be noted that the high-resolution MM5 wind data were valuable for computing the trajectories arriving at LT3. For example, trajectories calculated from ECMWF data, whose resolution (6-hourly, 1°) was coarser than the MM5, were not coincident with the observed lightning flashes at any time during their 2-day histories (not shown). Instead, these trajectories remained several hundred kilometers to the west-northwest of the line of convection during their passage near the East Coast.

Finally, a brief chemical spike occurs at ~1230 UTC at the southern end of the 10.1 km flight leg (Figures 6 and 7 and Plate 1). It is located just south of the stratospheric region, near 32°N at an altitude between LT1 and LT3. The spike is characterized by slightly elevated values of NO (~250 pptv), NO₂ (~600 pptv), and ultrafine CN (~8000 cm⁻³) in relation to other portions of the flight. This signature is consistent with previous lightning encounters and is likely a brief sampling of the lowest altitude of the lightning layer.

To summarize, results indicate that the layer between ~9 and 12 km located south of the stratospheric region is characterized by lightning signatures. The LT3 chemical signature (Figures 6 and 7) is the strongest lightning signal of flight 10, apparently due to the long period over which its trajectories are exposed to extensive lightning. This exposure for LT3 is greater than observed with the two other lightning signatures (LT1 and LT2).

4.3. Pollution Signature

The chemical signature attributed to pollution (POL) begins as the DC-8 descends to its lowest altitude of the flight (5.8 km, 486 hPa) (Figures 6 and 7 and Plate 1). It occurs during the entire length of this leg, from ~1528 to 1646 UTC (20°–28°N). Several species that exhibit moderately enhanced values relative to the rest of flight 10 suggest surface-based pollution. The most notable is CO, which increases from 80 to 85 ppbv during most of the flight to >90 ppbv in POL (Figure 6). Similarly, ozone is relatively high compared to nonstratospheric portions of the flight. Its mixing ratio for the POL leg exceeds 75 ppbv, corresponding to the shoulder in the probability distribution function (PDF) (Figure 5). Similar increases are observed in PAN (70–120 pptv) and H₂O (500–1500 ppmv) in Figures 6 and 7, and C₂H₂ (100–140 pptv), H₂O₂ (250–450 pptv), and

Figure 14. Lightning event 3. Hourly trajectory positions (circles) and trajectory paths (lines) over 3-hour intervals are indicated. NLDN cloud-to-ground lightning flashes are indicated in gray. The specific time interval is given at the top of each panel.
Figure 15. Backward trajectories from POL (51–53 hours back). Arrival time at flight level is 1500–1700 UTC October 29, 1997. Arrows along trajectory paths indicate trajectory locations at 24-hour intervals. The flight track is indicated by the thin line.

Horizontal plots of backward trajectories from POL are shown in Figure 15. Although their basic orientation is similar to those of the previously described signatures, the important difference is that these trajectories pass very near the heavily populated Northeast Corridor of the United States. Figure 16 focuses on the trajectories between 1500 and 2100 UTC October 27 as they pass along the East Coast. One should note that there are virtually no encounters between the trajectories and cloud-to-ground lightning; that is, the trajectories pass west of the lightning. This is consistent with the chemical signature. On the other hand, satellite imagery (Figure 16a) indicates that the trajectories do pass near relatively deep clouds (darker shades) that do not contain cloud-to-ground lightning. Cloud locations at 2045 UTC are especially interesting (Figure 16c); that is, a narrow band of rain showers parallels the coast from North Carolina to Cape Cod. Coldest cloud

Figure 16. (a) GOES-8 infrared satellite image at 1445 UTC October 27, 1997. (b) Three-hour trajectory paths (black) and NLDN cloud-to-ground lightning strikes (gray) for the period of 1500–1800 UTC October 27, 1997. (c) GOES-8 infrared satellite image at 2045 October 27, 1997. (d) Three-hour trajectory paths (black) and NLDN cloud-to-ground lightning strikes (gray) for the period of 1800–2100 UTC October 27, 1997.
top temperatures range from $-35^\circ$ to $-45^\circ$C, corresponding to heights of $\sim$7.5–9.5 km. The median trajectory altitude in this region is $\sim$8.6 km (between 36 and 42 hours back in Figure 17). Since this area is west of a cold front that is located offshore over the Atlantic, low-level winds are from the northwest (not shown). Thus it appears that surface-based pollution from the Northeast Corridor is transported at low levels toward the line of convection, transported aloft by the convective updrafts, and then carried horizontally to the location of the POL signature.

4.4. Mixed Layer

The layer from $\sim$7.5 to 9 km is between the lightning and pollution signatures and appears to be a mixed region (MIX) of the two signatures. This layer is sampled during two flight legs, that is, first at 8.8 km (320 hPa) between 1235 and 1319 UTC, and again at 7.6 km (376 hPa) between 1453 and 1526 UTC (Plate 1). The results show that the signatures of these two flight legs are not identical (Figures 6 and 7), with the lower leg exhibiting a weak pollution signal and the upper leg consistent with a slight lightning influence.

The dominant feature of the 8.8 km leg is a chemical spike that is evident at $\sim$1310 UTC (indicated by the ellipse in Plate 1 and arrows in Figures 6 and 7). This brief signature is characterized by the following values: NO (90 pptv), NO$_2$ (300 pptv), and ultrafine CN ($5000 \text{ cm}^{-3}$). Although these values are only slightly elevated compared to the background, they suggest a weak lightning signature.

Figure 18 shows that the backward trajectories from the spike do experience a slight encounter with cloud-to-ground lightning between 2100 UTC October 27 to 0300 UTC October 28.
28 as they exit the coastal waters of the United States to the northeast. There are no close encounters during the periods that are not shown. These trajectories originate over Florida 49 hours earlier and then pass just south of Newfoundland before arriving at the flight track from the northwest (not shown). At the times when trajectory paths coincide with lightning locations, trajectory altitudes (not shown) are −5.7−6.2 km (−450−490 hPa), while satellite-derived cloud top temperatures indicate heights near −7−9 km (−300−400 hPa).

The 8.8 km flight leg containing the lightning spike is below the legs containing the major lightning signatures (LT1−LT3, Plate 1). In addition, backward trajectories from the spike pass over the coastal waters of the East Coast at lower altitudes than trajectories from LT1 to LT3 (not shown). These lower levels appear to be below the main outflow of the extensive storms producing the intense cloud-to-ground lightning. Thus the DC-8 may be sampling outflow from intermittent lower-level lightning producing storms. This hypothesis is difficult to verify with the hourly satellite-derived cloud top data that are available.

The second sampling of the mixed layer begins at ~1453 UTC when the DC-8 descends to 7.6 km (376 hPa) (see Figures 6 and 7 and Plate 1). In addition, backward trajectories from the spike pass over the coastal waters of the East Coast at lower altitudes than trajectories from LT1 to LT3 (not shown). These lower levels appear to be below the main outflow of the extensive storms producing the intense cloud-to-ground lightning. Thus the DC-8 may be sampling outflow from intermittent lower-level lightning producing storms. This hypothesis is difficult to verify with the hourly satellite-derived cloud top data that are available.

5. Summary and Conclusions
Flight 10 of NASA’s Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (SONEX) originated in Lajes,
A stack maneuver along the southern half of the flight track provided samples at four different altitudes, that is, 10.7, 9.5, 7.6, and 5.8 km. The horizontal length of these flight legs was relatively short, only ~400–800 km. A variety of chemical signatures was encountered during the flight. This research has examined these chemical signatures of Flight 10 in detail, relating them to high-resolution meteorological data from a mesoscale numerical model.

The National Center for Atmospheric Research (NCAR)/Penn State Fifth Generation Mesoscale Model (MM5) was used to create a high-resolution three-dimensional meteorological data set. The model was configured with two-way interactive nesting, that is, with a coarse (90 km) outer grid and a finer (30 km) internal grid. Each domain contained 26 vertical levels. Analyses from the MM5 simulation agreed closely with global analyses. The MM5 data were available at hourly intervals and then used to create backward trajectories from the locations of the chemical signatures. We believe that the MM5-derived meteorological data provided an accurate depiction of actual wind regimes at a higher temporal resolution than traditional global analyses. This improved resolution is important since trajectories are very sensitive to slight errors in both wind direction and speed as well as the temporal frequency of the wind data.

Four major categories of chemical signatures were discussed: stratospheric, lightning, continental pollution, and a mixed layer. The strong stratospheric signal was encountered just south of the Azores. Values of O₃ were greatly elevated, while values of CO and water vapor were very small in comparison to other portions of the flight. The tropopause was relatively low in the region, extending down to ~6 km, due to a nearby closed low-pressure circulation.

Three chemical signatures were attributed to lightning. These signatures were located south of the lowered stratospheric region at relatively high altitudes (11.9, 10.7, and 9.5 km). Two of the signatures occurred during the stack maneuver. Their chemical characteristics included elevated values of NO, NOₓ, and water vapor, as well as elevated ultrafine CN. Backward trajectories from locations of these chemical signatures were related to locations of cloud-to-ground lightning. Results showed that the trajectories had passed over regions of lightning located over the eastern Gulf of Mexico and off the southeast coast of the United States 1–2 days earlier. Two additional lightning signatures (the spikes) were observed during legs at 8.8 and 10.9 km.

The lowest leg of the stack maneuver (5.8 km) exhibited a chemical signature consistent with continental pollution. Species that were enhanced included PAN, water vapor, O₃, C₃H₂, H₂O₂, and CH₃OOH. Backward trajectories from the pollution signature had passed over the highly populated Northeast Corridor of the United States. Since they passed to the west of the region of lightning that was farther offshore, no lightning signature was indicated. Surface-based pollution apparently was lofted to the altitudes of the trajectories by convective clouds along the East Coast that did not contain lightning.

Finally, a mixed layer was detected along two flight legs between ~7.5 and 9 km. The chemical signature at 7.6 km was intermediate to signatures located at higher and lower flight legs. Species whose values were intermediate to those of the lightning and pollution segments included NO, NOₓ, CN, PAN, water vapor, and hydrogen peroxide. The lightning signature was located directly over the 7.6 km leg at an altitude of 9.5 km, while the pollution signature was located directly below at 5.8 km. Trajectories arriving in this region had passed over locations off the East Coast that were between those of the lightning and pollution signatures. Thus they likely were impacted by both lightning and pollution. These trajectories exhibited a wide range of altitudes when passing over the region of influence. A second leg of mixed signatures was located at 8.8 km. The chemical signature along this leg exhibited a brief spike that is attributable to lightning.

In summary, the results show that high-resolution numerical modeling can provide meteorological data that are useful for understanding complex chemical signatures. This capability was especially evident during the aircraft’s stack maneuver, when four different chemical signatures were collocated at different altitudes, that is, pollution, mixed, lightning, and stratospherically influenced.

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