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Large-scale enhancements in NO/NOy from subsonic aircraft emissions: Comparisons with observations

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Abstract. One of the DC-8 flights from the 1992 AASE 2 campaign flew south from Maine over the Atlantic Ocean, sampling air downstream of areas in the eastern United States associated with heavy air traffic. We use a photochemical trajectory model to help interpret observed NO/NO_y ratios from the stratospheric portions of this flight. The model is run with and without an additional in situ NO_x source from the 1992 Boeing-McDonnell Douglas (BMD) emissions climatology. During the northern section of this flight, the inclusion of this additional NO_x source resulted in a significant improvement with observed large-scale NO/NO_y ratios. This comparison suggests that air traffic over the eastern United States is sufficiently dense to enhance NO/NO_y ratios on a regional scale, even when the characteristic NO spikes from exhaust plumes are absent. During the southern portion of the flight, in which the DC-8 flew at a higher altitude, observed NO/NO_y ratios agreed much better with the no-emissions scenario. This may be a reflection of the difficulty of using a climatological NO_x emissions database to infer instantaneous NO/NO_y ratios. It would be desirable to have a larger database of lower stratospheric NO and NO_y measurements downstream of the eastern United States. This would enable more stringent statistical comparisons of observed NO/NO_y ratios with the model-predicted enhancements of this ratio arising from aircraft emissions.

1. Introduction

The inclusion of aircraft emissions in multidimensional models gives rise to significant enhancements in the large scale distribution of NO_x (= NO + NO_2) in the upper troposphere and lower stratosphere [*Derwent*, 1982; *Beck et al.*, 1992; *Ehhalt et al.*, 1992; *Kasibhatla*, 1993; *Flatoy and Hov*, 1996; *Kraus et al.*, 1996; *Brasseur et al.*, 1996; *Lamarque et al.*, 1996]. Unfortunately, it is difficult to compare the climatological NO_x distributions generated by these models directly with observations. The NO/NO_y ratio of an air parcel is affected by many factors, including the concentrations of ozone (O_3) and total reactive nitrogen (NO_y), aerosol surface area, solar zenith angle, temperature, and its back trajectory over the past several days. These factors must be accurately reproduced by a model in order to attribute enhancements in NO/NO_y to aircraft emissions. Multidimensional models are not in general designed to do this. In this paper we use a photochemical trajectory model, run with and without an additional NO_x source from aircraft, to help interpret NO and NO_y measurements from the 1991-1992 second Airborne Arctic Stratospheric Expedition (AASE 2).

The NO_x/NO_y ratio of an air parcel recently exposed to aircraft exhaust will be much higher than the background value of 0.01 to 0.03 in the midlatitude winter lower stratosphere [*Folkins et al.*, 1994]. This ratio will decrease as the air parcel is mixed with ambient air and as the emitted NO_x is converted to other forms of NO_y such as nitric acid (HNO_3). Simulations have shown that most of the initial decrease in NO_x/NO_y associated with the entrainment of ambient air into the plume is nearly complete after 16 hours (0.75 days) [*Danilin et al.*, 1994]. Subsequent decreases in this ratio will be largely driven by photochemical relaxation to steady state. In regions where air traffic is sufficiently dense, plumes may start to coalesce before the photochemical relaxation of NO_x/NO_y to steady state is complete. If this happens, aircraft emissions will give rise to a large-scale enhancement in NO_x/NO_y. It is most likely to occur in regions sufficiently downstream of regions with heavy air traffic that most of the mixing has occurred, but not so far downstream that photochemical

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processing has reduced the NO_x/NO_y ratio to a near-background value.

In the lower stratosphere, most of the conversion of NO_x to HNO_3 occurs via the following three reactions:

$$NO_2 + O_3 \rightarrow NO_3 \quad (1)$$

$$NO_2 + NO_3 + M \rightarrow N_2O_5 + M \quad (2)$$

$$N_2O_5 + H_2O(aq) \rightarrow 2\,HNO_3 \quad (3)$$

During the day, NO_3 photolyzes rather than combining with NO_2 via reaction (2), and N_2O_5 can photolyze to reform NO_x rather than react with water on the surface of a sulfate aerosol as in reaction (3). At night however, concentrations of NO_2 are usually sufficiently high that almost every NO_3 formed via reaction (1) will react with NO_2 to form N_2O_5. Aerosol surface areas are usually sufficiently large that reaction (3) is the dominant sink of N_2O_5 [*Fahey et al.*, 1993]. If in this case, one assumes that every reaction between NO_2 and O_3 at night represents the conversion of two NO_x molecules to HNO_3, the lifetime of NO_x at night is approximately given by $1/2k_1[O_3]$. Over a 24 hour period this timescale would have to be multiplied by the fraction of nighttime hours. For a 12-hour night, an ozone mixing ratio of 300 ppbv, and a pressure of 200 mbar, this lifetime is approximately 3 days. Ambient zonal winds in the lower stratosphere are about 20 m/s. One would therefore expect enhancements in NO_x/NO_y arising from aircraft emissions to persist for approximately 5000 km downstream of the major source regions over North America and Europe.

2. The February 20 AASE 2 Flight

Although the primary objective of AASE 2 was to investigate the extent of chlorine and bromine catalyzed ozone depletion within the polar vortex, DC-8 measurements of NO and NO_y during this campaign [*Weinheimer et al.*, 1994] can also be used to investigate the effects of aircraft NO_x emissions. The DC-8 usually flies in the upper troposphere and lower stratosphere between 9 and 12 km, the range of altitudes in which most aircraft emissions of NO_x occur. Many of the small-scale spikes in NO and NO/NO_y observed from the DC-8 during AASE 2 have been attributed to recent interceptions of aircraft exhaust [*Zheng et al.*, 1994; 1996]. The observed large-scale variation in NO/NO_y did not appear to require an in situ NO_x source from

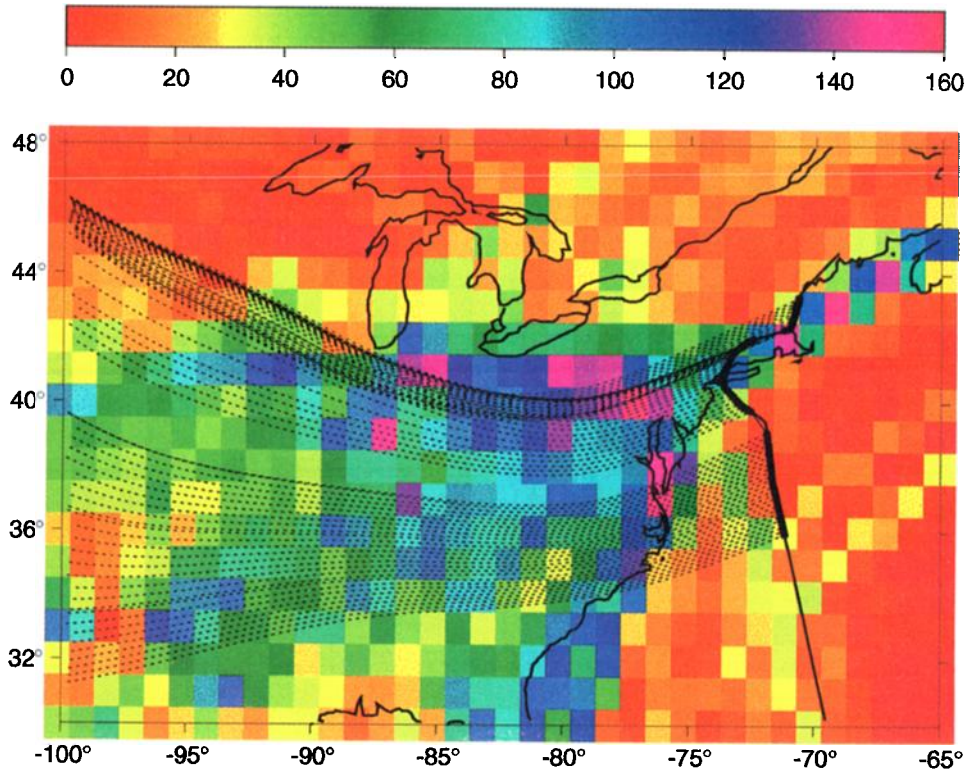

Plate 1. The annually averaged rate of NO_x emission (pptv/d) at 10 km from the February 1992 Boeing-McDonnell Douglas (BMD) inventory. Also shown is a portion of the February 20, 1992, DC-8 flight south from Maine toward Puerto Rico. The back trajectories from this flight were evaluated at the points along the flight where 1-min averaged measurements of NO were made. The photochemical trajectory model uses the inventory to introduce an additional source of NO_x into the air parcel as it travels along the trajectory. The back trajectories shown here extend back approximately 1 day from the DC-8 flight track.

aircraft, but did support previous studies [*Fahey et al.*, 1993, *Kawa et al.*, 1993, *Dessler et al.*, 1993] implicating a role for the N_2O_5 aerosol reaction in the suppression of NO/NO_y ratios in the lower stratosphere [*Folkins et al.*, 1994]. The lack of a large-scale signature in NO/NO_y from aircraft emissions can probably be accounted for by the fact that most of the DC-8 flights during AASE 2 occurred in high-latitude regions between bases in Alaska, Norway, Maine, and California. The only DC-8 flight that sampled air immediately downstream of a major continental emission region occurred on February 20, 1992. On this flight, the DC-8 left Bangor Maine flying almost directly south over the Atlantic Ocean. After reaching 15°N, it reversed direction and flew back to Bangor. The southbound portion of the flight track is shown in Plate 1.

Plate 1 also shows back trajectories from points along the February 20 flight track where measurements of NO were made. They were obtained using the Goddard Space Flight Center (GSFC) isentropic trajectory model [*Schoeberl and Sparling*, 1994]. The input winds for the model were derived from National Meteorological Center (NMC, now National Centers for Environmental Prediction (NCEP)) temperatures and pressures using the approximation of geostrophic balance. Back trajectories for most of the measurements of NO originate 10 days earlier over Europe. Plate 1 shows the portions of the trajectories approximately one day prior to intersecting the DC-8 flight track. The back trajectories are superimposed on a color plot of the diurnally averaged rate of NO_x emission from aircraft in parts per trillion per day at 10 km for February 1992. These emissions rates were derived from an inventory compiled by Boeing and McDonnell Douglas (BMD) [*Baughcum et al.*, 1996; *Metwally*, 1995]. The inventory has a horizontal resolution of 1°latitude by 1°longitude and a vertical resolution of 1 km.

Plate 1 illustrates the high degree of spatial variability of NO_x emission rates. Background rates of 10 pptv/d or less should have only a modest effect on observed NO_x/NO_y ratios because ambient NO_x mixing ratios during winter in the midlatitude lower stratosphere are about 60 pptv, and the lifetime of NO_x emissions is only several days. Emission rates as high as 160 pptv/d in parts of the eastern United States should, however, considerably enhance NO_x/NO_y ratios in downstream regions over the western Atlantic Ocean.

3. Photochemical Trajectory Model Description

It has been shown that photochemical models integrated along back trajectories can accurately predict observed NO_x/NO_y ratios in the lower stratosphere [*Kawa et al.*, 1993]. Here we use this approach to determine if better agreement with observed NO/NO_y ratios can be obtained by including aircraft emissions as an additional source of NO_x and NO_y. Each of the back trajectories shown in Plate 1 corresponds to a particular observed NO/NO_y ratio. At the starting point of each 10-day back trajectory, a 30-day initialization run drives the NO_x/NO_y ratio to local steady state. The mixing ratios of the long-lived species such as O_3, CO [*Sachse et al.*, 1991], C_2H_6 [*Anderson et al.*, 1993], aerosol surface area [*Pueschel et al.*, 1994], and H_2O are held fixed at the values observed at each NO measurement. At the end of the initialization run, the NO/NO_y ratio evolves forward in time along the back trajectory in a manner consistent with the prescribed variation in solar zenith angle, temperature, and pressure. Comparisons with observed NO/NO_y ratios using this type of approach are expected to be quite stringent because many of the factors that affect NO concentrations are realistically constrained in the model.

To test sensitivity to initial conditions, we multiplied the initial steady state NO_x/NO_y ratios at the starting point of each trajectory by factors of 0.5 and 2. In each case the final NO_x/NO_y ratios calculated by the photochemical trajectory model differed by less than 5%.

The model described above is run in two scenarios. In the emissions scenario we include an additional aircraft source of NO_x at each point along the back trajectory. This is calculated by linearly interpolating in height, latitude, and longitude from the NO_x emission rates given in the BMD inventory. The NO_y mixing ratio at the start of each back trajectory is set equal to the value observed from the DC-8, minus the sum of all the NO_x emissions into that air parcel during the 10-day back trajectory (typically 100 pptv). In the no-emissions scenario the initial NO_y mixing ratio for each trajectory is set equal to the NO_y mixing ratio measured by the DC-8, and the aircraft source of NO_x is not included.

There is no guarantee that the procedure for adding NO_x in the emissions scenario will reproduce the actual rate of NO_x emission into an air parcel as it passes through a particular grid cell in the BMD inventory. If, however, the density of aircraft plumes within a grid cell is sufficiently high, the NO_x enhancements from the individual plumes will start to merge together as the air is carried downstream. In general, one would expect that with a higher density of air traffic, the time needed for the plumes to coalesce will be shorter. If this timescale is smaller than the timescale needed for photochemical relaxation of the NO_x/NO_y ratio to steady state (about 3 days), then the enhancement in NO_x/NO_y from aircraft will be perceived not as a disjoint collection of NO spikes, but as a larger scale enhancement in the NO_x/NO_y ratio. There should then be an intermediate range of downstream sampling distances in which most of the mixing has occurred (ie., not many spikes remain) but background NO_x/NO_y ratios remain elevated. This paper assumes that air traffic over the eastern United States is sufficiently dense that it does give rise to a

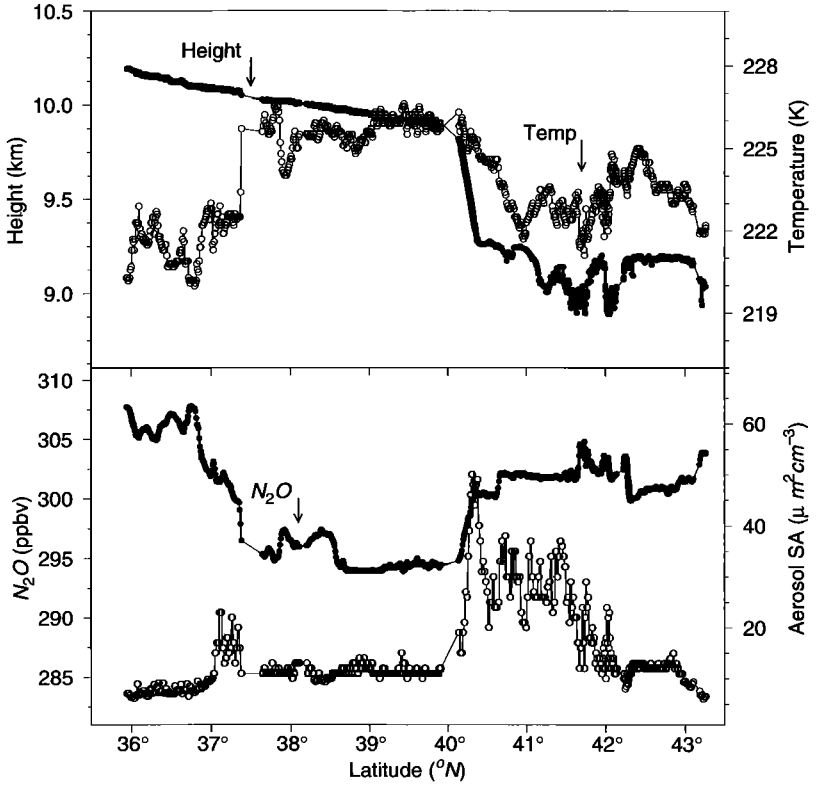

Figure 1. Temperature (open circles) and radar height (solid circles) during a portion of the February 20 flight. Most of the small scale variability in radar altitude north of 40°N is associated with orography. (bottom) Aerosol surface area (open circles) and N_2O mixing ratio (solid circles). Aerosol surface areas were enhanced as a result of the eruption of Mount Pinatubo the previous summer.

regional-scale enhancement in the NO_x/NO_y ratio over the western Atlantic Ocean.

The photochemical trajectory model was run for those measurements of NO for which simultaneously observed N_2O mixing ratios were less than 310 ppbv and O_3 mixing ratios were greater than 100 ppbv. Restricting attention to the lower stratosphere helps avoid difficulties associated with the presence of other NO_x sources and sinks in the troposphere such as lightning [*Tuck*, 1976; *Ridley et al.*, 1996], washout, and the convective injection of surface emissions into the upper troposphere. Solar zenith angles during this restricted portion of the flight (between 43.2°N and 35.9°N) were between 51°and 54°. NO mixing ratios were therefore always considerably higher than the detection limit of 10 pptv [*Folkins et al.*, 1994].

The photochemical part of the model is essentially the same as that used in a previous study [*Folkins et al.*, 1994]. In addition to the fixed species listed above, the model also includes total reactive chlorine Cl_y and total reactive bromine Br_y. Mixing ratios of these species were inferred from correlations with N_2O [*Daniel et al.*, 1996]. Figure 1 shows various meteorological and chemical tracers along the DC-8 flight track. N_2O mixing ratios varied from 294 ppbv to 310 ppbv for the section of the flight analyzed here. The *Daniel et al.* [1996] correlations give $Br_y = 0$ for N_2O greater than 302.7 ppbv and $Cl_y = 0$ for N_2O greater than 305.5 ppbv. The mixing ratios of both Br_y and Cl_y were therefore extremely small or zero for much of the flight, and their inclusion had little effect on modeled NO/NO_y ratios. These correlations are, however, largely based on measurements taken at much higher altitudes. The relative error associated with extrapolating these measurements to air parcels just above the midlatitude tropopause may be quite large.

The model also includes 33 intermediate species and 12 short-lived species. The time evolution of the intermediate species was calculated using an implicit expression, while the instantaneous concentrations of the short-lived species were calculated using steady state assumptions. The model was run with a time step of 7.2 min. The photolysis rates of 24 species were calculated using a delta-Eddington method with a climatological midlatitude winter ozone profile (A. Kylling, Phodis, A program for calculation of photodissociation rates in the Earth's atmosphere, 1995, available by anonymous ftp to kaja.gi.alaska.edu, cd pub/arve). The surface albedo was fixed at 0.5. Reactions rates were obtained from the 1994 Jet Propulsion Laboratory compilation [*DeMore et al.*, 1994]. The two heterogeneous reactions included in the model were reaction (3) and $BrONO_2$

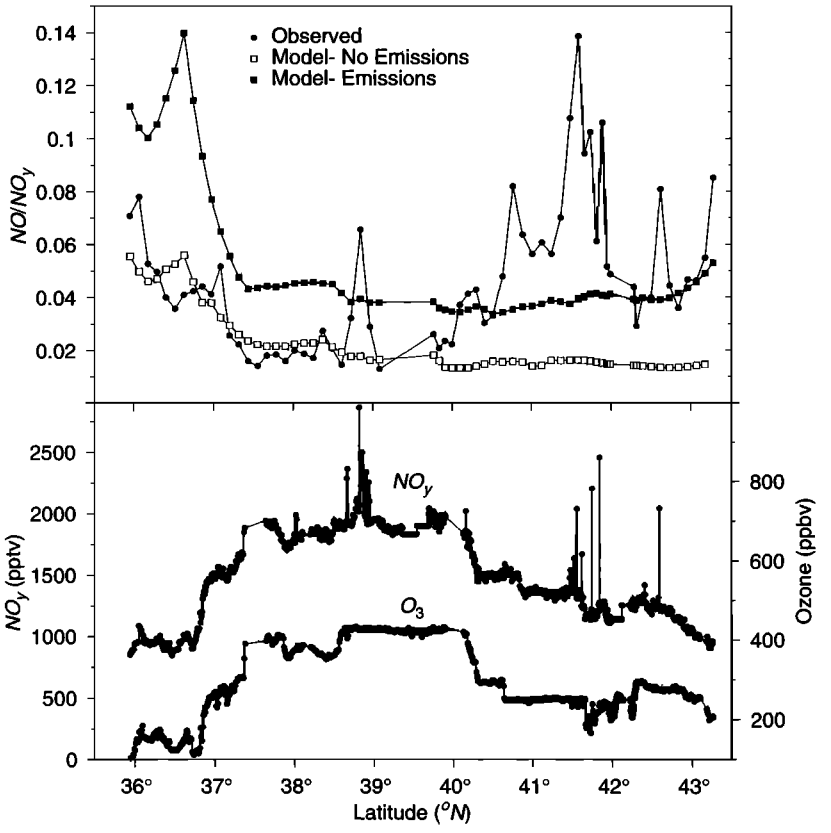

Figure 2. (top) Observed NO/NO_y (solid circles), modeled NO/NO_y without aircraft emissions (open squares), and modeled NO/NO_y with aircraft emissions (solid squares). (bottom) NO_y and O_3 along the flight track, measured at 1-sec intervals.

$+ H_2O \rightarrow HOBr + HNO_3$ [*Hanson et al.*, 1996], with reaction probabilities of 0.1 and 0.8 respectively. Although there are several other heterogeneous reactions of possible importance in the stratosphere, they tend to become significant below 210 K [*Lary et al.*, 1996]. Minimum temperatures along the 10-day back trajectories were rarely below 210 K.

4. Discussion

Figure 2 shows observed NO/NO_y ratios from the stratospheric portion of the February 20 flight track, as well as NO/NO_y ratios generated by the two model scenarios with and without aircraft emissions of NO_x. North of 39.5°N, the model scenario without an aircraft source of NO_x significantly underestimates observed NO/NO_y ratios. The inclusion of the aircraft source of NO_x from the BMD climatology results in much better agreement with the observed ratio. Most of the measurements from this portion of the flight were taken within, or very near, the North Atlantic flight corridor (NAFC) (see Plate 1). For the most part, however, the increase in NO/NO_y associated with the inclusion of aircraft NO_x emissions in the model arises from emissions further west over the continental United States, rather than from recent emissions within the NAFC. This appears to be consistent with observations. Most of the large-scale enhancement in observed NO/NO_y over the no-emissions model between 39.5°N and 43.5°N is not associated with the presence of NO_y spikes. The persistence of enhancements in NO/NO_y arising from aircraft emissions, despite the absence of spikes, presumably reflects the fact that the chemical timescale for the conversion of NO_x to HNO_3 is longer than the dynamical timescale required to mix emissions into the background atmosphere.

In contrast with the northern portion of the flight, Figure 2 shows that the observed variation in NO/NO_y south of 39.5°N is well reproduced by the scenario that does not include an aircraft source of NO_x. For the most part, inclusion of aircraft emissions results in a significant overestimate of observed NO/NO_y ratios. This overestimate is associated with a change in altitude of the DC-8. Figure 1 shows that the DC-8 ascended almost 1 km at 40°N, near the latitude at which the observed NO/NO_y ratio first approached the no-emissions scenario. The DC-8 may have ascended above the altitude at which most aircraft over the continental United States were flying on February 20, 1992.

The time needed for an air parcel to traverse the eastern United States where aircraft emissions are highest is approximately 1 day. Most air traffic occurs during

the day. It is therefore conceivable that some of the failures of the no-emissions scenario are attributable to the fact that the BMD inventory is a diurnally averaged climatology. We performed sensitivity studies in which we doubled the emissions between 0800 and 2000 LT, and set the emissions to zero at night. This enhanced modeled NO/NO_y ratios by up to 10 %, which did not improve agreement with observed NO/NO_y during the southern portion of the flight.

5. Conclusions

This paper is a first attempt to use the BMD NO_x aircraft emissions database in a photochemical trajectory model to quantitatively predict enhancements in NO/NO_y downstream of a major flight region. The inclusion of this additional NO_x source did not unambiguously improve agreement with observed NO/NO_y ratios. In the northern section of the February 20 flight, the no-emissions scenario substantially underestimated observed NO/NO_y ratios. Adding the aircraft emissions increased modeled NO/NO_y ratios and brought them much closer to those observed. However in the southern portion of the flight, observed NO/NO_y ratios agreed well with the no-emissions scenario, and the addition of the aircraft emissions degraded this prior agreement. It is difficult, on the basis of one flight, to determine whether or not these differences are attributable to incorrect model assumptions, or to the statistical difficulties inherent in using a climatological NO_x emissions database to infer observed NO/NO_y ratios. It would be desirable to have a larger number of lower stratospheric NO and NO_y measurements off the eastern coast of the continental United States. This would generate a climatology of NO and NO_y downstream of a major aircraft source region and help determine whether or not observed NO_x enhancements from subsonic aircraft are consistent with theoretical expectations.

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