Observation of pollution plume capping by a tropopause fold

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Abstract. Airborne lidar measurements reveal a case in which a layer of high-ozone air extruding from a tropopause fold appears to cap a pollution plume and force it to spread out in the lower troposphere. The morphology of the high-ozone layer resembles a three-dimensional model of tropopause fold evolution that produces a low-altitude potential vorticity tube. This is a mechanism that can complete the irreversible transfer of air from the stratosphere, and can also affect pollution levels at the surface if the capping layer reaches the top of the boundary layer.

Introduction

Many previous studies have shown that trace constituents in the atmosphere are transported in layers and filaments. Clearly, differential advection is the underlying mechanism that stretches and shears passive scalars into the observed intricate morphologies. In the stratosphere, large-scale dynamics [*Reid and Vaughan*, 1991] and inertio-gravity waves [*Danielsen et al.*, 1991] appear to dominate the advective process, with the balance between the two being modulated by height, season, and location [*Pierce and Grant*, 1998; *Grant et al.*, 1998]. The hypothesized cascade of scalar variance from large to small scales has now been observed and measured in the lower stratosphere [*Lindborg and Cho*, 2000]. In the troposphere the situation is complicated by the presence of highly localized pollution sources, strong cross-isentropic forcing by convection, and episodic input from the stratosphere through tropopause folds and other routes. The huge range in water vapor content might also play a role in layer maintenance through vertically variable radiative cooling, while fully developed three-dimensional (3D) turbulence intermittently homogenizes trace constituent structures.

Among the tropospheric trace gases, ozone, in particular, has drawn much attention. In the troposphere, ozone can add to the greenhouse effect on one hand and contribute to the regulation of the air's oxidizing (cleansing) capacity on the other hand. At the surface, an overabundance of ozone is dangerous to many organisms. Past studies using ozone and humidity measured by several thousand commercial aircraft flights have established the predominance of high-ozone, low-humidity layers in the troposphere; detailed statistics were given in *Newell et al.* [1999]. (The algorithm that defined what constituted a layer was outlined in *Stoller et al.* [1999].) Furthermore, most of the layers of this type were found to have higher static stability than the surrounding air, and the seasonal cycle in the observed tropospheric-ozone-layer-occurrence rate corresponded well to the seasonal variation in tropopause folding rate on the 350-K isentrope [*Thouret et al.*, 2000]. These findings point to stratospheric intrusions as the dominant source of tropospheric ozone layer formation in the free troposphere. However, a seemingly contradictory piece of evidence comes from more limited research aircraft studies incorporating carbon monoxide and methane in the layer type classification: A majority of the high-ozone, low-humidity type had high carbon monoxide and/or high methane, which suggested pollution as the main source of this type of layer, while a minority had low carbon monoxide and low methane expected for layers of purely stratospheric origin [*Stoller et al.*, 1999].

To solve this paradox, it was proposed [*Newell et al.*, 1999; *Stoller et al.*, 1999] that a combination of pollution plume plus stratospheric intrusion might be responsible for this type of layer, i.e., the former rising from the ground would be capped and flattened out like an anvil cloud by the strong stability of the latter. Mixing would then ensue to create a "dirty" stratospheric layer. In this letter we present the first observed evidence for such a capping process.

Observations

On October 13, 1997, the NASA DC-8 research aircraft flew across the country in the first leg of the Subsonic Assessment (SASS) Ozone and Nitrogen Experiment (SONEX) [*Singh et al.*, 1999; *Thompson et al.*, 2000]. A prominent stratospheric intrusion was present over the northcentral plains, and the DC-8 traversed its south end while over Kansas (Figure 1). The section of the tropopause fold penetrated by the aircraft was downstream of a trough and on the west side of a jet at the aircraft level. Satellite images showed a clear column of air along the eastern flank of the intrusion bordered farther east by a line of storm clouds associated with an accompanying lower-level cold front. (A detailed investigation of this tropopause fold was reported

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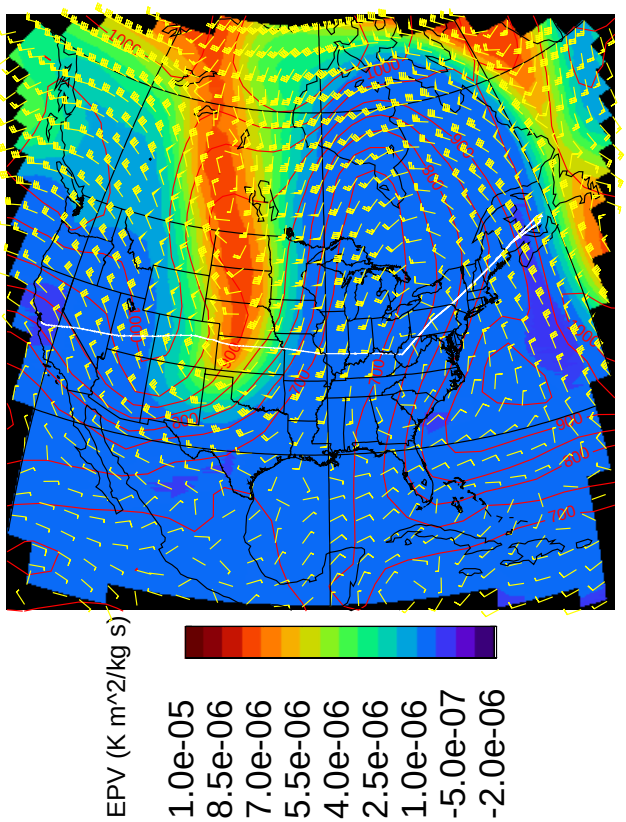

Figure 1. Map of geopotential height in decameters (red contour lines), Ertel's potential vorticity (color scale), and horizontal winds (yellow vectors) on the 330-K surface for 1200 UT, October 13, 1997. The scaling on the wind vectors is standard: one flag is 50 m s^{-1}, one bar is 10 m s^{-1}, and one half bar is 5 m s^{-1}. The plotted field values were output from a model run by the Data Assimilation Office of the NASA Goddard Space Flight Center. The longitudinal and latitudinal grid spacing used by the model was 2.5° by 2°. The white line shows the flight path of the DC-8, which departed Moffett Field, California, at 1519 UT and landed in Bangor, Maine, at 2241 UT. This figure is adapted from Plate 1 created by T. L. Kucsera and A. M. Thompson in *Cho et al.* [1999].

elsewhere [*Cho et al.*, 1999]. Air mass analyses for this flight were presented in *Grant et al.* [2000].)

The DC-8 carried on board differential absorption lidar (DIAL) systems capable of measuring vertical profiles of ozone and aerosol concentrations along the flight path [*Browell et al.*, 1998]. The ozone data were averaged over 300 m in the vertical and 300 s in time (or ∼70 km in distance), but archived in increments of 90 m and 60 s, respectively. The aerosol data (total atmospheric scattering ratio at 1064 nm) were averaged over 60 m in the vertical and 60 s in time (or ∼14 km in distance), but archived in increments of 90 m and 60 s, respectively. Because of the incomplete overlap of the transmitted beam and the receiver field of view at close range, there were no data from the DIAL near the aircraft height (∼±1 km). Thus, in this interval the profiles were interpolated, with the in situ chemiluminescence measurement [*Gregory et al.*, 1988] providing an additional point at aircraft level for the ozone data.

A resulting composite figure is shown in Figure 2. The ozone concentration in parts per billion by volume (ppbv) is displayed as a contour map, while the atmospheric scattering ratio is rendered as a color map. The thick black line cutting across the middle is the aircraft height, while the black line along the bottom of the aerosol data is the surface height. The Rocky Mountains rise toward the left edge, and the flat region to the immediate right starting at 105°W contains the Denver metropolitan area. Note the plume of dense aerosol concentration above this region that is apparently capped and flattened by the layer of high ozone streaming out from the tropopause fold. We believe this plume was urban/industrial haze, sometimes called the "Denver brown cloud," which was visually confirmed from the videotape record of the forward-pointing camera on the DC-8. There appears to be mixing of the two air masses in the region of the source plume.

For further clarity, Figure 3 shows the vertical profiles of ozone and aerosol at three longitudes along the flight track. Note the clear separation of the two layers at 100°W and 97°W consistent with the capping of the pollution layer by the ozone intrusion layer, while the graph at 103°W shows some signs of vertical mixing with the layer peaks closer together and a slight high-ozone, low-aerosol region below at ∼3.8 km. As can be seen from Figure 2, this latter region was closer to the convective source.

To examine the profile of meteorological variables, we obtained data from the nearest rawinsonde site, North Platte, Nebraska (41°07'N, 100°40'W). Figure 4 shows the profiles of relative humidity, the square of the Brunt-Väisälä fre-

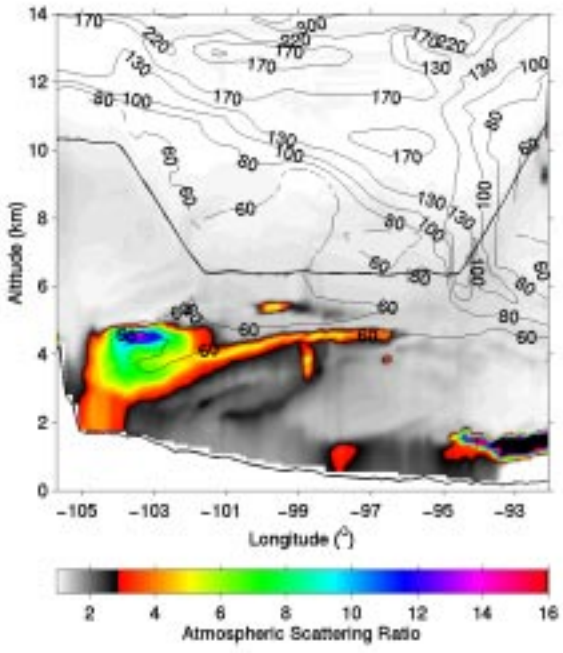

Figure 2. Ozone (contour lines, values in ppbv) and aerosol (color map of scattering ratio) measurements made by the onboard DIAL system in the vicinity of the tropopause fold over the central United States on October 13, 1997. The thick black line cutting across the center shows the altitude of the aircraft, and the thick black line along the bottom of the data display marks the surface height. The time range corresponding to this flight segment was 1700 UT to 1818 UT. The latitude of the aircraft changed slowly, going from 39.6°N at the left edge to 39.0°N at the right edge.

quency (N^2), and the square of the vertical shear in horizontal winds at 0 UT, October 14, 1997. The sharp transition from wet to dry air at ∼4.5 km matches Figure 2 very well. Note the very high static stability just above the transition height (capping), and the strong shear at the transition level (differential advection). The strong shear could lead to Kelvin-Helmholtz instability and mixing. The ozone data resolution of ∼70 km in the horizontal precludes direct observation of such an instability, however. Note also that N^2 is negative just below the capping height, implying convective instability, which can also facilitate mixing across the interface.

Summary Discussion

At midlatitudes, tropopause folds are believed to be the major source of stratosphere-to-troposphere transfer of air. However, the fold itself can be reversible, so for permanent stratosphere-troposphere exchange to take place, the stratospheric air in the fold must mix irreversibly with the tropospheric air in the outside environment. The high-ozone layer protruding from a tropopause fold observed here is reminiscent of the low-level potential vorticity (PV) tubes seen in a 3D model of tropopause fold evolution [*Cox et al.*, 1997; *Bithell et al.*, 1999]. These PV tube offsprings could detach themselves from the mother fold and remain in the troposphere, thereby completing the transfer of mass from the stratosphere. Such low-altitude extensions of the stratospheric influence may not only affect mid-tropospheric chemistry, but also pollution levels at the surface. For example, if, in Figure 2, the fold had been closer to the Rockies, then the high-ozone layer could have extended right into the boundary layer of the foothills, trapping the pollution along the ground and increasing the ozone level. Indeed, tropopause folds themselves have been known to reach to the top of the planetary boundary layer [*Browell et al.*, 1987].

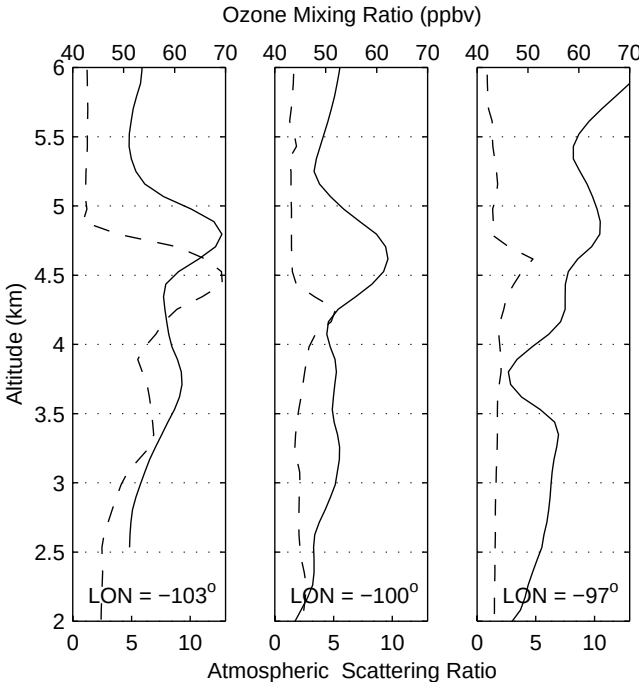

Figure 3. Vertical profiles of ozone (solid) and aerosol (dashed) extracted from the data presented in Figure 2 at three longitudes along the flight track.

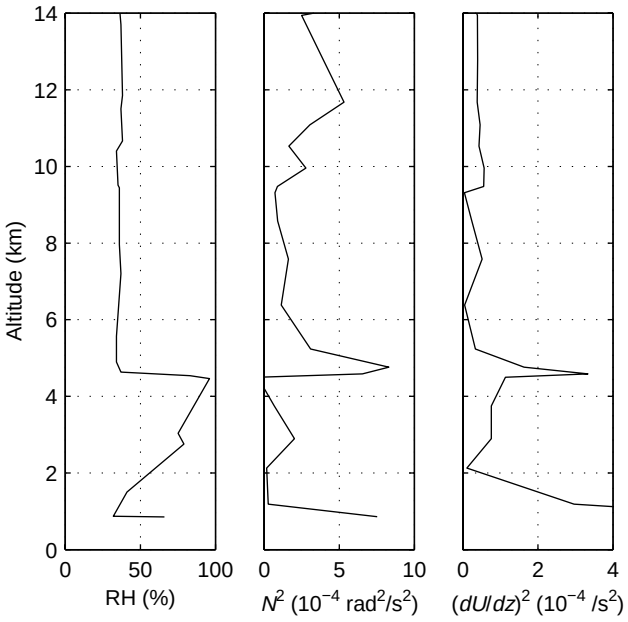

Figure 4. Data from a rawinsonde launched from North Platte, Nebraska (41°07'N, 100°40'W) at 0 UT, October 14, 1997. The left panel is relative humidity, the center panel is the square of the Brunt-Väisälä frequency, and the right panel is the square of the vertical shear in horizontal winds.

Of course, the anomalously high static stability associated with the high-ozone layer will eventually be destroyed by turbulent mixing and radiative transfer. But we have observed it lasting down to 2 km in altitude (Plate 5 of *Cho et al.* [1999]), and a dry layer is self-stabilizing from a radiative cooling perspective (Figure 9 of *Stoller et al.* [1999]). But even after the enhanced static stability is gone, back trajectory studies suggest that dry high-ozone layers can exist for more than 10 days without diffusing into the ambient air [*Bithell et al.*, 2000]. Indeed, in some locations, mid-tropospheric ozone layers can be a quasi-permanent feature [*Baray et al.*, 2000].

This study confirms that a pollution plume can be capped and flattened by a dry, statically stable, high-ozone layer emanating from a stratospheric intrusion. It proves that a tropospheric trace constituent layer can have both stratospheric and pollution sources. However, one case study does not prove that it happens frequently. Another possible instance of pollution capping by a stratospheric layer published in the literature is the case of *Parrish et al.* [2000]. Their measurements were made only horizontally in situ, but they estimated from the altitude gradient of equivalent potential temperature that the stratospheric and polluted layers were vertically adjacent with a transition thickness of ∼250 m.

However, because the pollution plume was quite wet in our study (Figure 4), the explanation for the prevalence of high-ozone, low-humidity, high-CO, high-CH_4 layers is still problematic. Since this statistic comes from a limited set of campaign measurements over the Pacific, with the tropical campaign flights often much farther away from the pollution sources than the case examined here, it may be that the dryness resulted from gradual subsidence over long distances rather than from a local stratospheric source. This

is consistent with the fact that the results from the remote tropical Pacific mission contained a larger percentage of this type of layer than the two missions held over the western Pacific (closer to continental pollution sources) [*Fenn et al.*, 1999; *Stoller et al.*, 1999].

Another aspect of the possible superposition of a stratospheric and pollution layer is that high ozone brought into contact with a strong moisture source provides a potential source of hydroxyl, which can oxidize some of the pollutants, particularly when there is ample sunlight. This cleansing mechanism is akin to an atmospheric washing machine.

Current chemical transport models (CTMs) cannot reproduce high-resolution results as seen in Figure 2. However, nonlinear chemical reactions provide great leverage to such large trace gas concentration gradients in changing the overall results. For example, ozone production may be overpredicted by as much as 60% in a model with coarse resolution compared to a finer resolution model [*Liang and Jacobson*, 2000]. It is important to push for finer resolution in CTMs or to somehow find ways to parameterize subgrid processes effectively.

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References

Baray, J.-L., V. Daniel, G. Ancellet, and B. Legras, Planetary-scale tropopause folds in the southern subtropics, *Geophys. Res. Lett.*, *27*, 353–356, 2000.

Bithell, M., L. J. Gray, and B. D. Cox, A three-dimensional view of the evolution of midlatitude stratospheric intrusions, *J. Atmos. Sci.*, *56*, 673–688, 1999.

Bithell, M., G. Vaughan, and L. J. Gray, Persistence of stratospheric ozone layers in the troposphere, *Atmos. Environ.*, *34*, 2563–2570, 2000.

Browell, E. V., E. F. Danielsen, S. Ismail, G. L. Gregory, and S. M. Beck, Tropopause fold structure determined from airborne lidar and in situ measurements, *J. Geophys. Res.*, *92*, 2112–2120, 1987.

Browell, E. V., S. Ismail, and W. B. Grant, Differential absorption lidar (DIAL) measurements from air and space, *Appl. Phys.*, *B67*, 399–410, 1998.

Cho, J. Y. N., et al., Observations of convective and dynamical instabilities in tropopause folds and their contribution to stratosphere-troposphere exchange, *J. Geophys. Res.*, *104*, 21,549–21,568, 1999.

Cox, B. D., M. Bithell, and L. J. Gray, Modelling of stratospheric intrusions within a mid-latitude synoptic-scale disturbance, *Q. J. R. Meteorol. Soc.*, *123*, 1377–1403, 1997.

Danielsen, E. F., R. S. Hipskind, W. L. Starr, J. F. Vedder, S. E. Gaines, D. Kley, and K. K. Kelly, Irreversible transport in the stratosphere by internal waves of short vertical wavelength, *J. Geophys. Res.*, *96*, 17,433–17,452, 1991.

Fenn, M. A., et al., Ozone and aerosol distributions and air mass characteristics over the south Pacific during the burning season, *J. Geophys. Res.*, *104*, 16,197–16,212, 1999.

Grant, W. B., R. B. Pierce, S. J. Oltmans, and E. V. Browell, Seasonal evolution of total and gravity wave induced laminae in ozonesonde data in the tropics and subtropics, *Geophys. Res. Lett.*, *25*, 1863–1866, 1998.

Grant, W. B., et al., A case study of transport of tropical marine boundary layer and lower tropospheric air masses to the northern midlatitude upper troposphere, *J. Geophys. Res.*, *105*, 3757–3769, 2000.

Gregory, G. L., E. V. Browell, and L. S. Warren, Boundary layer ozone: An airborne survey above the Amazon Basin, *J. Geophys. Res.*, *93*, 1452–1468, 1988.

Liang, J., and M. Z. Jacobson, Effects of subgrid segregation on ozone production efficiency in a chemical model, *Atmos. Environ.*, *34*, 2975–2982, 2000.

Lindborg, E., and J. Y. N. Cho, Determining the cascade of passive scalar variance in the lower stratosphere, *Phys. Rev. Lett.*, *85*, 5663–5666, 2000.

Newell, R. E., V. Thouret, J. Y. N. Cho, P. Stoller, A. Marenco, and H. G. Smit, Ubiquity of quasi-horizontal layers in the troposphere, *Nature*, *398*, 316–319, 1999.

Parrish, D. D., J. S. Holloway, R. Jakoubek, M. Trainer, T. B. Ryerson, G. Hübler, F. C. Fehsenfeld, J. L. Moody, and O. R. Cooper, Mixing of anthropogenic pollution with stratospheric ozone: A case study from the North Atlantic wintertime troposphere, *J. Geophys. Res.*, *105*, 24,363–24,374, 2000.

Pierce, R. B., and W. B. Grant, Seasonal evolution of Rossby and gravity wave induced laminae in ozonesonde data obtained from Wallops Island, Virginia, *Geophys. Res. Lett.*, *25*, 1859–1862, 1998.

Reid, S. J., and G. Vaughan, Lamination in ozone profiles in the lower stratosphere, *Q. J. R. Meteorol. Soc.*, *117*, 825–844, 1991.

Singh, H. B., A. M. Thompson, and H. Schlager, SONEX airborne mission and coordinated POLINAT-2 activity: Overview and accomplishments, *Geophys. Res. Lett.*, *26*, 3053–3056, 1999.

Stoller, P., et al., Measurements of atmospheric layers from the NASA DC-8 and P-3B aircraft during PEM-Tropics A, *J. Geophys. Res.*, *104*, 5745–5764, 1999.

Thompson, A. M., H. B. Singh, H. Schlager, Introduction to special section: Subsonic Assessment Ozone and Nitrogen Oxide Experiment (SONEX) and Pollution From Aircraft Emissions in the North Atlantic Flight Corridor (POLINAT 2), *J. Geophys. Res.*, *105*, 3595–3603, 2000.

Thouret, V., J. Y. N. Cho, R. E. Newell, A. Marenco, and H. G. J. Smit, General characteristics of tropospheric trace constituent layers observed in the MOZAIC program, *J. Geophys. Res.*, *105*, 17,379–17,392, 2000.

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