Ozone and other trace gases in the Tropical Tropopause Layer (TTL) over the Pacific Ocean

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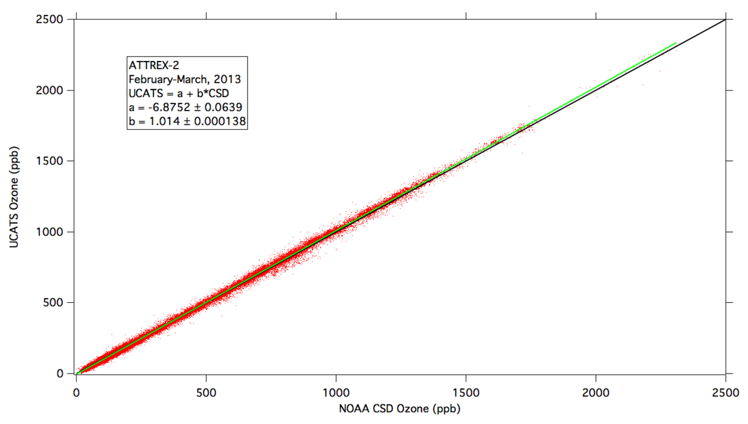
Introduction

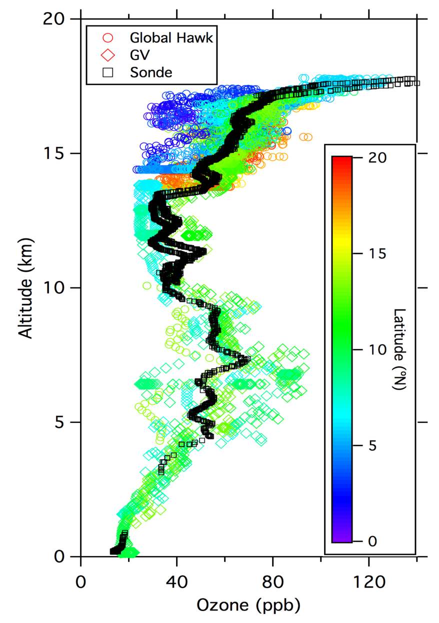
The distribution of ozone in the tropics is influenced by both chemistry and transport, and in turn affects the oxidation rate of a large number of organic species, including shorter-lived ozone depleting substances (ODS) such as organic bromine compounds. The tropical tropopause layer (TTL; ~14-18.5 km) over the western Pacific is one of the primary entry points of air from the troposphere into the stratosphere, yet *in situ* observations in this region have been relatively sparse. The NASA Airborne Tropical Tropopause Experiment (ATTREX) mission was designed to study trace gases, clouds, dehydration, and transport in the TTL over the Pacific Ocean, in order to better understand how water vapor and ozone-depleting gases reach the lower stratosphere or are removed in the TTL. Field campaigns were carried out on the NASA Global Hawk aircraft, with about 200 vertical profiles in the TTL over the central and eastern tropical Pacific (ATTREX-1 and 2; 2011 and 2013) and the western Pacific in January-March 2014 (ATTREX-3). During ATTREX-3, the Global Hawk was joined by the NSF/NCAR GV and British BAe-146 aircraft for the CONTRAST and CAST missions, providing coverage of the atmosphere from the boundary layer to 19 km. Coincident balloon profiles of ozone and water vapor were also obtained from Guam during ATTREX-3.

Measurements and data quality

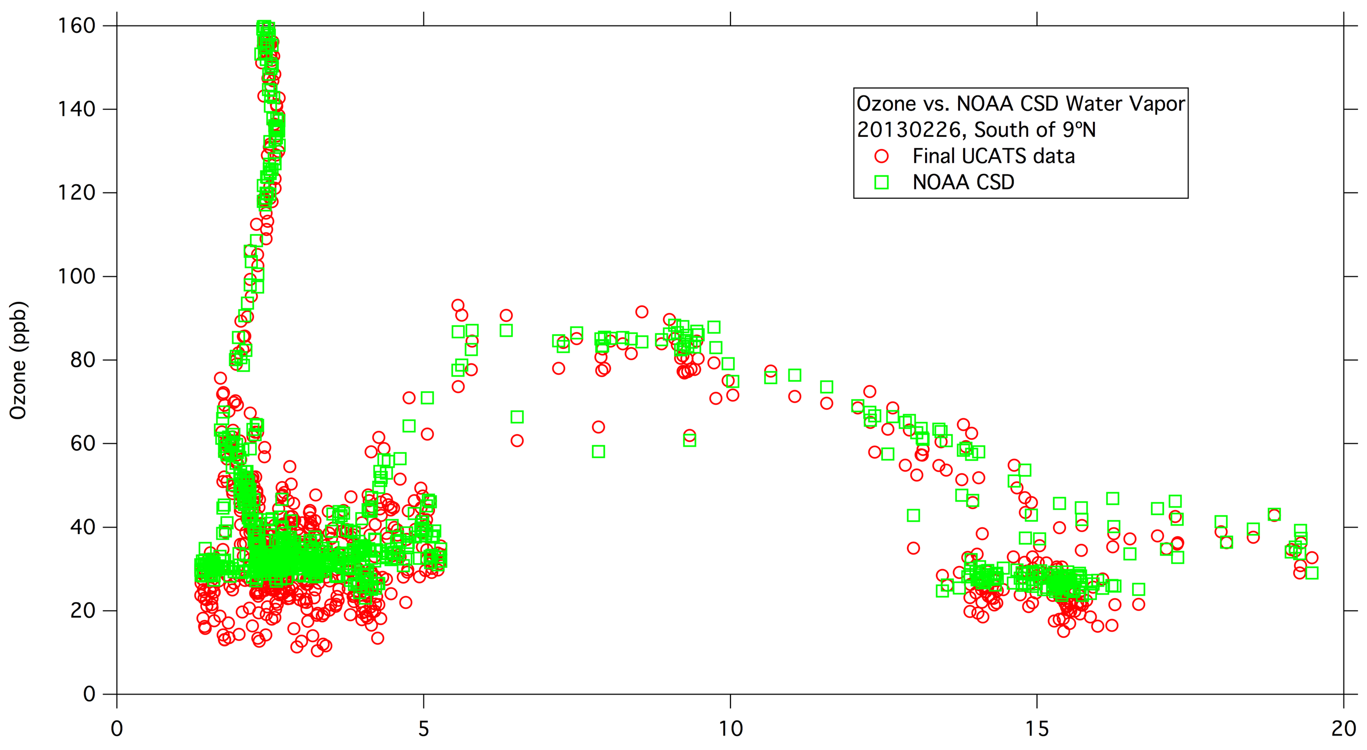
Measurements on the Global Hawk included water vapor and condensed water measured by the NOAA Chemical Sciences Division (CSD); long-lived trace species including N_2O, SF_6, and methane measured by the UAS Chromatograph for Atmospheric Trace Species (UCATS); ozone measured by both CSD and UCATS; CO_2, methane, and CO measured by a Picarro instrument from Harvard University; a large suite of trace gases including bromine-containing compounds measured by a Whole Air Sampler from the University of Miami; remote sensing instruments for cloud particles, temperature profiles, and BrO; and meteorological parameters. In ATTREX-1 and 2, both ozone instruments were on board; in ATTREX-3 the NOAA CSD instrument was not able to fly because of weight and balance considerations. Considerable effort was expended to ensure that the UCATS ozone data were of adequate quality for the science goals. This included adding a second ozone sensor from 2B Inc., extensive calibrations and checks for offsets, tests of pressure sensors and other components, merging the data from the two 2B sensors, and intercomparisons with the CSD instrument in flight. With all effects accounted for, the two instruments agreed to within about 1% with an offset of about 5 ppb. Illustrative examples of data are shown below.



All coincident data from ATTREX-2 science flights, with merged data from the two 2B sensors in UCATS plotted against NOAA CSD ozone. Improvements to the slope came from calibrations of the pressure sensor and accounting for the pressure offset across the cell at low pressures (pressure is measured downstream of the photometer cells on the 2B sensors). Precision of the UCATS data was improved by merging the data from the two 2B sensors.

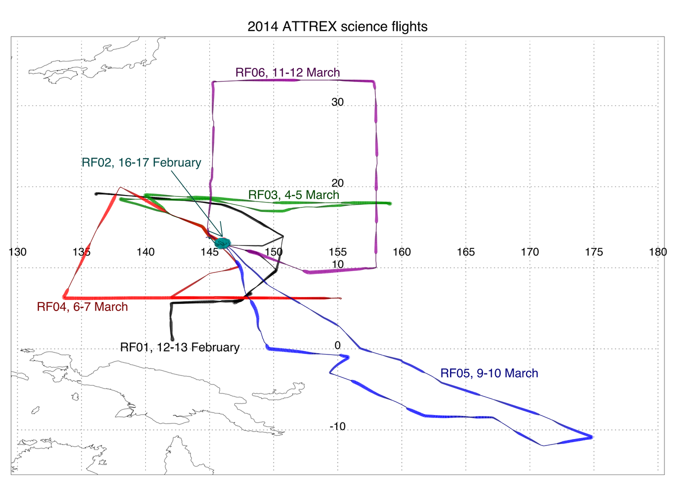


Ozone data from the Global Hawk, GV, and sonde launch from February 12-13, 2014, color-coded by latitude. The GV flight occurred on February 12, and the balloon launch was timed to coincide with the return of the Global Hawk on February 13. Despite a great deal of variability (caused by latitudinal gradients and tropospheric filaments), the data all show a consistent picture. Lowest ozone in the TTL was observed on the southernmost extent of the flight (dark colors).

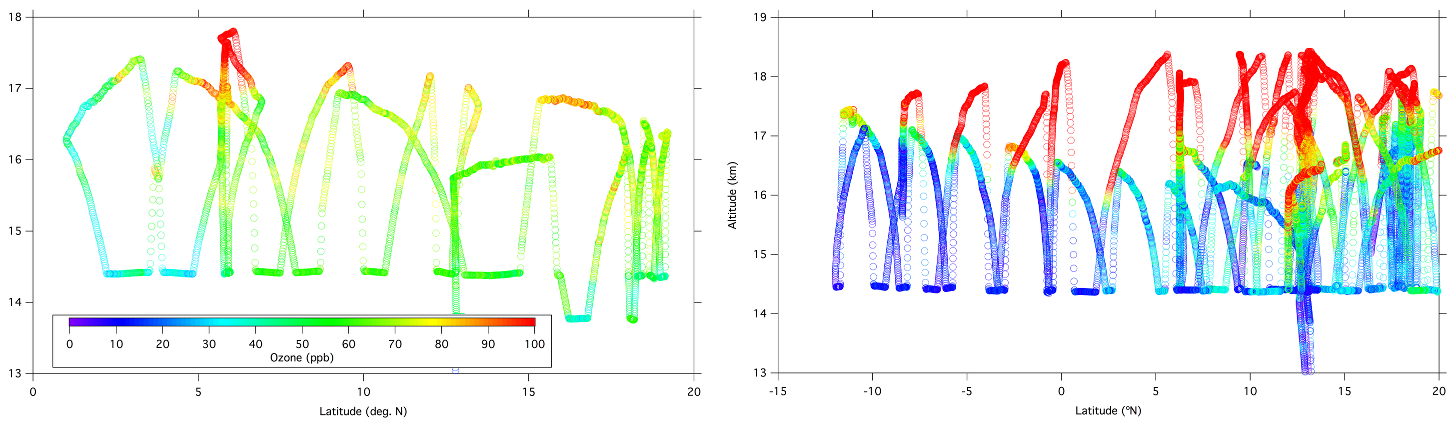


Ozone and water vapor are typically anti-correlated, with high water vapor and low ozone in the troposphere, and high ozone and lower water in the stratosphere. On February 26, 2013 over the central Pacific near 9°N, a more complicated pattern was observed, with ozone and water redistributed by recent convective activity. The CSD instrument shows a somewhat tighter correlation and better precision, but the UCATS data are qualitatively and quantitatively similar, and lead to the same conclusions. This particular event is being analyzed now; the UCATS data will enable similar studies for ATTREX-3.

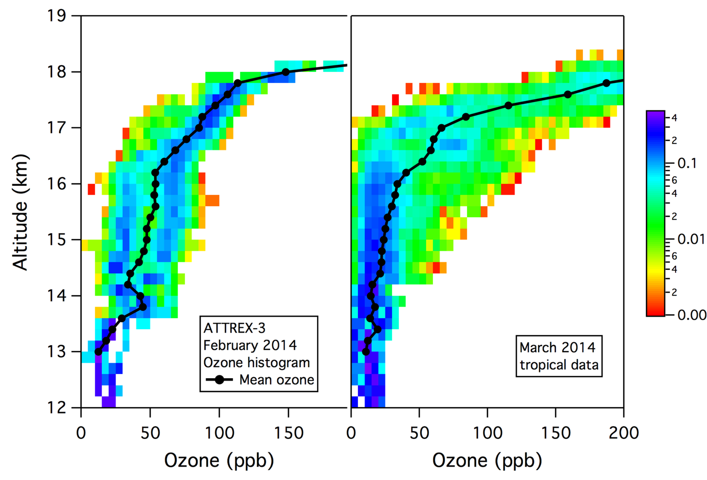
ATTREX results



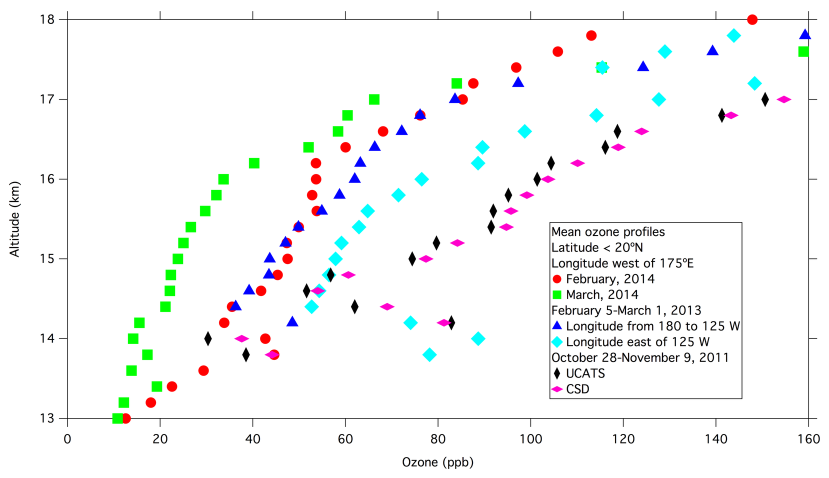
Map of Global Hawk flight tracks for ATTREX-3 science flights. Thicker lines indicate higher altitudes (~18 km); thinner lines indicate lower altitudes during profiles (~14 km). The transit flight on March 13 produced additional profiles in the tropics east of Guam (not shown); the transit flight to Guam on January 16 was entirely at the maximum altitude of the Global Hawk. The flight of February 16-17 was confined to the "UAV zone" just east of Guam and appears as a blob in this figure. Only one flight (March 9-10) reached into the Southern Hemisphere.



(Left) Altitude-latitude curtain plot for the flight of February 12-13, 2014, color-coded by ozone. All ozone values greater than 100 ppb appear as red; for this flight, where altitude never exceeded 18 km, the highest ozone value recorded was only about 125 ppb. The lowest ozone values (close to 30 ppb) were only observed near the southernmost extent of the flight. The actual flight path appears as the black trace in the map for ATTREX-3 flights. (Right) Curtain plot showing all the March flights in the western tropical Pacific. Ozone was clearly much lower in March compared to February in the TTL, with many values around 20 ppb or lower. On average, the lowest ozone was in the southern hemisphere (difficult to see with this color scheme), but the southern hemisphere data are from only one flight that crossed the equator (March 9-10).



Histogram of ozone values for each 200 m vertical bin for western tropical Pacific data from all flights in February (left) and the four science flights and the first part of the transit flight from Guam in March, 2014 (right). Magenta and blue colors correspond to higher probabilities. The black line and circles show the mean ozone value at each altitude starting at 13 km; data are much more sparse below this altitude.



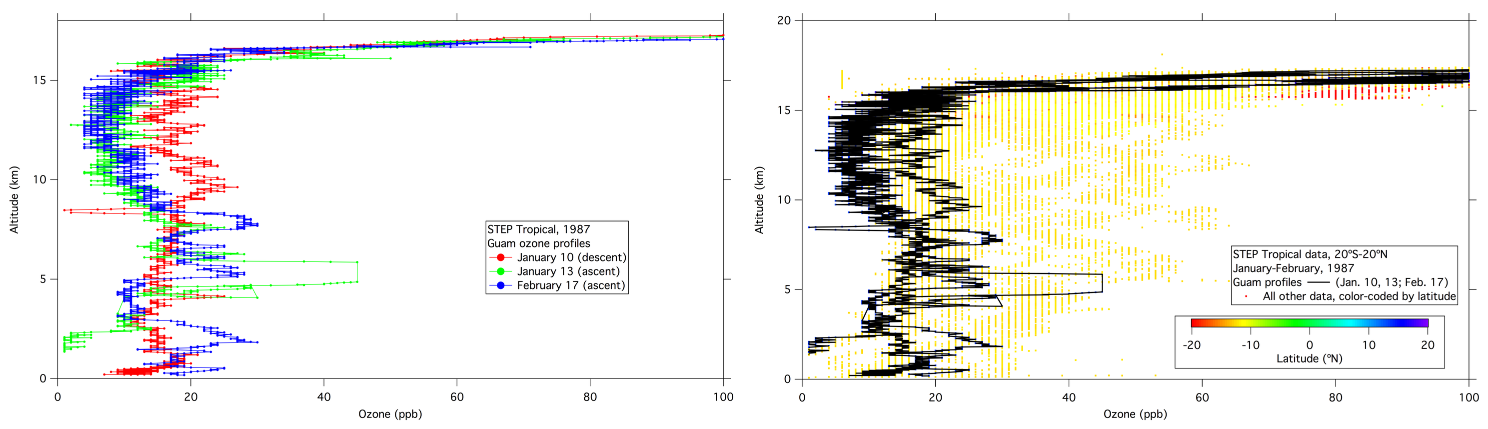
Mean ozone profiles for February (red circles) and March (green squares) 2014 from the western tropical Pacific, February-March 2013 from the central (blue triangles) and eastern (cyan diamonds) tropical Pacific, and October-November ATTREX-1 data from the eastern and central Pacific. The lowest altitudes of some profiles are dominated by only a few points and are likely not representative of average conditions. Ozone in the lower TTL was clearly lowest in March 2014. 10-day back trajectories (not shown) from March largely originated in the central and western tropical Pacific; trajectories from the February flights originated far to the west and passed over Africa, the Middle East, and south and southeast Asia.

Summary of ATTREX results

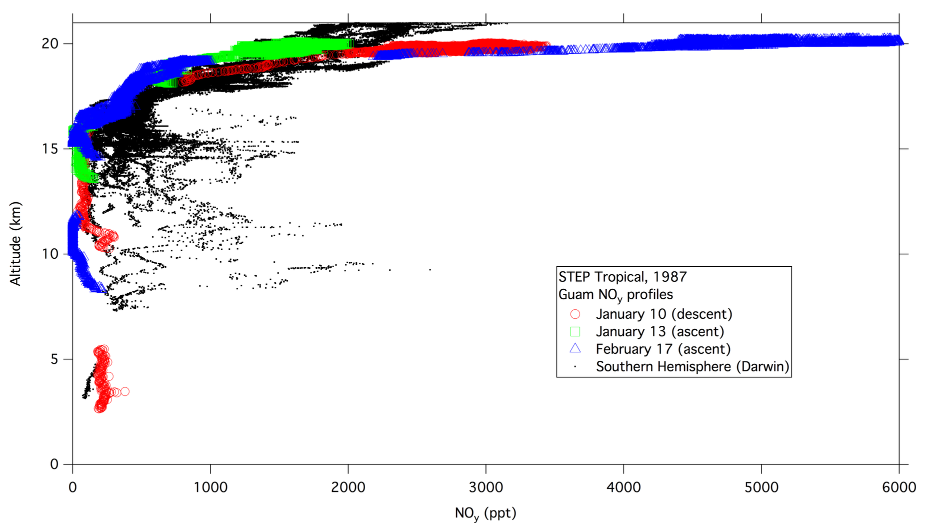
Ozone was consistently low (10-40 ppb) in the lower TTL over the western Pacific, with low values extending up to the cold point tropopause, particularly in March 2014. Ozone over the central and eastern Pacific in February-March 2013 often averaged 40-50 ppb, and typically increased slowly with height from about 14 km to the tropopause. The results are consistent with frequent but not uniform deep convection, bringing low-ozone air from the marine boundary layer directly to the upper troposphere over the western tropical Pacific.

The STEP Tropical Mission

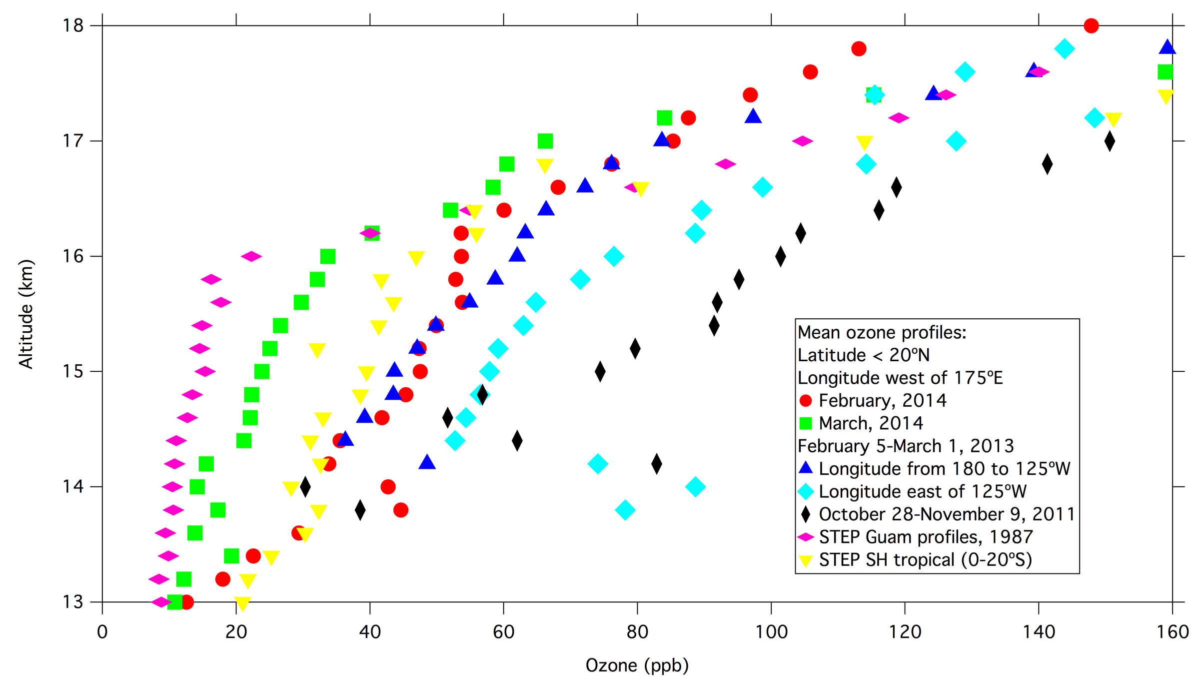
During the winter 1987 STEP Tropical mission, the NASA ER-2 aircraft made several profiles at Guam during transit flights and encountered very low values of ozone, lower than observed at their principal study site in Australia.



(Left) These data are from the original version of the NOAA CSD photometer (Proffitt and McLaughlin, 1983) used on the Antarctic and Arctic ozone depletions missions. The profiles at Guam from transit flights in STEP Tropical show among the lowest values of ozone recorded by an in situ photometric instrument. There is evidence of layers of slightly enhanced ozone (e.g., at 5 km on January 13, 1987) but much less than what was frequently observed during CONTRAST in 2014. (Right) Ozone values in the free troposphere and TTL over Guam were actually lower than near the STEP Tropical study site at Darwin, Australia, where frequent and intense deep convection was observed.



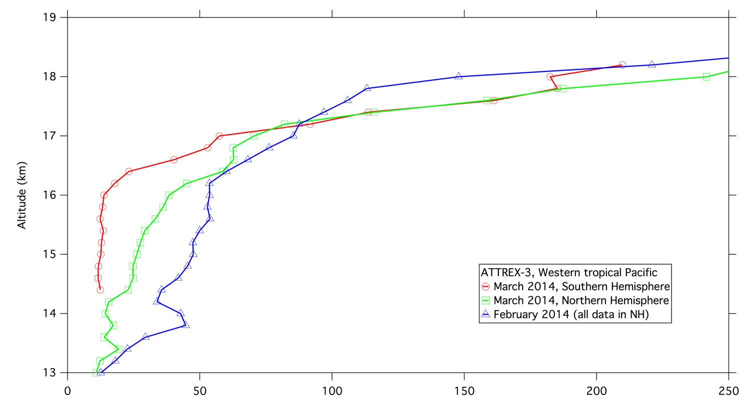
Altitude profiles of reactive nitrogen (NO_y) help to explain the difference between the Guam and Darwin data. Over Australia, NO_y is entrained in an anti-cyclonic system and brought upward by frequent and intense convection. Over Guam, NO_y was consistently low, reflecting the origin of the observed air masses in the clean marine boundary layer of the central and western tropical Pacific.



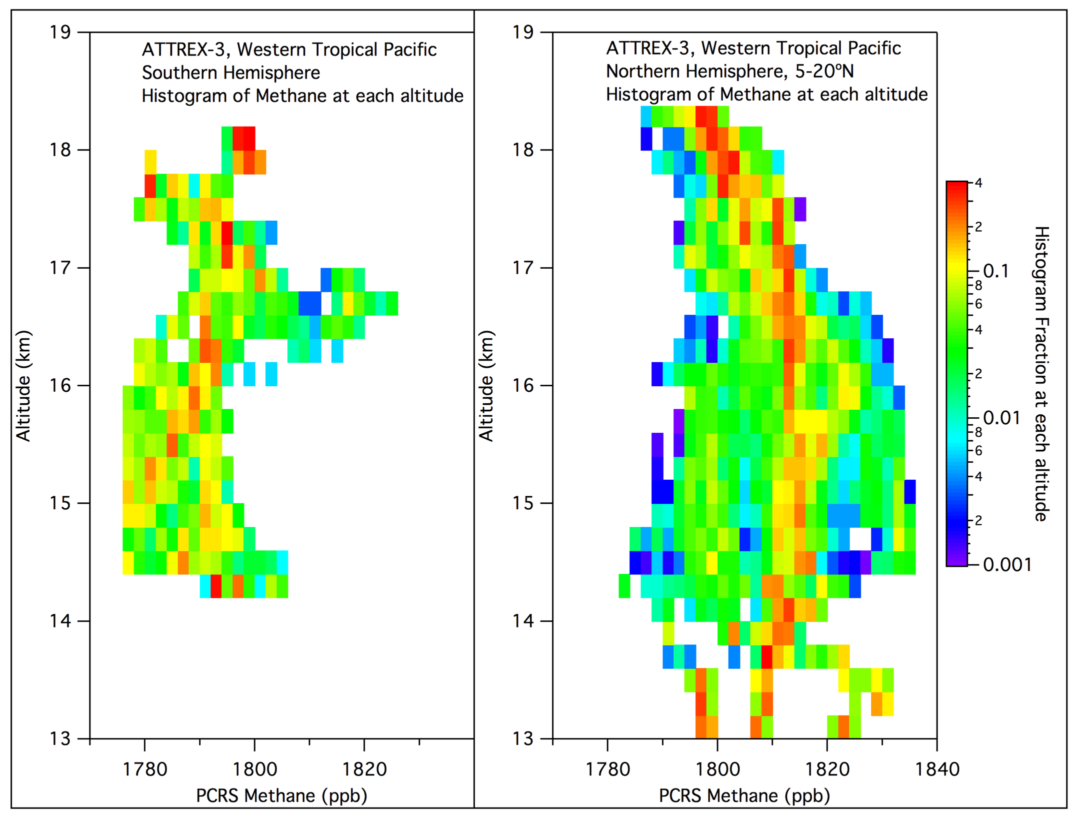
Average vertical profiles of ozone in the TTL, with STEP Tropical data from Guam and the southern hemisphere added in. As seen in the individual flight profiles, the mean ozone values at Guam from 1987 are extremely low, averaging between 10 and 20 ppb up to the approximate tropopause (near 16 km). The lowest ozone observed in the TTL was about 5 ppb, but it did not reach zero.

Latitude gradients

ATTREX-3 results also showed gradients in ozone and other trace gases between northern and southern hemispheres, but with lower values of ozone in the southern hemisphere. A few results are presented here illustrating the distributions of ozone and other trace gases, and northern hemisphere/southern hemisphere differences.



In ATTREX, the lowest values of ozone were observed in the southern hemisphere, in contrast to STEP Tropical. ATTREX southern hemisphere data came from only one flight, over the ocean, whereas the southern hemisphere STEP data were from continental air over Australia.



Measurements of methane and other tracers also showed differences between the northern and southern hemispheres in 2014 during ATTREX-3. Values of anthropogenic gases were higher in the northern hemisphere, reflecting their sources at higher latitudes. Latitudinal gradients of shorter-lived ozone depleting substances such as CH_2Br_2 and $CHBr_3$ (not shown) were not significant, reflecting their natural sources in the tropical ocean.

Conclusions

- Low ozone (<20 ppb) can occur in the western tropical Pacific TTL, but it does not do so uniformly.
- The troposphere over the tropical western Pacific near Guam has changed dramatically since 1987. Or, meteorological conditions were much different during STEP tropical (future work).
- Interhemispheric gradients may reflect the influence of the ITCZ/SPCZ (intertropical and South Pacific convergence zones), which form a barrier between the northern and southern hemispheres.

Acknowledgements

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