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DOE/SC-ARM-15-036

Coordinated Airborne Studies in the Tropics (CAST) Field Campaign Report

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May 2016

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Work supported by the U.S. Department of Energy,
Office of Science, Office of Biological and Environmental Research

Executive Summary

The last field campaign held at the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility site on Manus Island, Papua New Guinea (PNG), was conducted in February 2014 as part of the Coordinated Airborne Studies in the Tropics (CAST) campaign. This campaign was a collaboration between the National Aeronautics and Space Administration (NASA), National Center for Atmospheric Research (NCAR), and the United Kingdom's (UK) Natural Environment Research Council (NERC) to study the composition of the Tropical Tropopause Layer (TTL) and the impact of deep convection on this composition. There are three main areas of interest: i) transport of trace gases in the tropical atmosphere (especially short-lived halogenated compounds that can be lifted rapidly into the TTL, where they augment the stratospheric loading of these species); ii) formation of cirrus and its impact on the TTL; and iii) the upper-atmosphere water vapor budget. Overall, the aim was to improve understanding of the dynamical, radiative, and chemical role of the TTL.

The Manus operation was a joint experiment between the Universities of Manchester and Cambridge and the UK National Centre for Atmospheric Science (NCAS). It consisted of two elements: an ozonesonde campaign to measure ozone vertical profiles through the TTL, and ground-based monitoring of ozone, halogenated hydrocarbons, and greenhouse gases to determine the composition of lower-boundary-layer air in the Warm Pool region. Thanks to the support from the ARM Climate Research Facility and the exemplary collaboration of ARM staff in the region, the campaign was very successful.

Thirty-nine ozonesondes were launched, at least one a day between February 2 and 25. Thirty-four of these gave good ozone profiles and the rest only meteorological (radiosonde) profiles. A controversial aspect of ozonesonde measurements in the tropics is how to handle the background current of the sonde; near-zero concentrations of ozone have been reported in the literature, which are not compatible with model predictions. Twice during the campaign, comparisons were conducted between the ozonesondes and aircraft measurements by the NCAR Gulfstream V. These allow us to confirm that subtracting a constant background current for the ozonesondes is the correct approach, provided this is around 50 nA or less. Where larger background currents were measured, laboratory and inter-comparison measurements confirm the need to reduce the background to 50 nA in the TTL.

The lowest ozone concentrations thus measured in the TTL were 10-15 ppbv, in a highly convective period when outflow from deep convection to the east passed over Manus in the TTL. Such concentrations are compatible with the ground-level concentrations measured by a TECO analyser during this time. Much lower concentrations—< 5 ppbv at night—were measured at the ground early in the campaign, when the Manus area was free of deep convection. During this period the CH_4 and CO_2 measurements showed the impact of local sources, due to the development of a stable nocturnal boundary layer in very light winds. The median dry air mole fractions for $CHBr_3$, $CHBr_2Cl$, CH_3I, and CH_2ClI were 0.81, 0.52, 0.3 and 0.17 ppt respectively, and maximum dry air mole fractions were 8.6, 1.5, 2.5, and 0.5 ppt respectively.

Acronyms and Abbreviations

ACTIVE	aerosol and chemical transport in tropical convection
ARM	Atmospheric Radiation Measurement Climate Research Facility
ATTREX	Airborne Tropical Tropopause Experiment
CAST	Coordinated Airborne Studies in the Tropics
CONTRAST	CONvective TRansport of Active Species in the Tropics
DOE	U.S. Department of Energy
ECC	electrochemical cell
GC-ECD	gas chromatograph- electron capture detector
HIAPER	High-performance Instrumented Airborne Platform for Environmental Research
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NCAS	National Centre for Atmospheric Science
NERC	Natural Environment Research Council
PNG	Papua New Guinea
RH	relative humidity
TTL	tropical tropopause layer
TWP-ICE	Tropical Warm Pool – International Cloud Experiment
UK	United Kingdom
UV	ultraviolet

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1.0 Background

CAST (Coordinated Airborne Studies in the Tropics) is a research project funded by the United Kingdom's (UK) Natural Environment Research Council (NERC) with the aim of collaborating with the National Aeronautics and Space Administration (NASA) in exploiting the Global Hawk unmanned aircraft to study the tropical tropopause layer (TTL). The project has two parts: participation in a field campaign conducted in the West Pacific from January to March 2014, and development of instruments to fly on the Global Hawk in the East Pacific during March 2015. There are three areas of interest: 1) transport of trace gases in the tropical atmosphere; 2) formation of cirrus and its impact on the TTL; and 3) the upper-atmosphere water vapor budget. Overall, the aim was to improve understanding of the dynamical, radiative, and chemical role of the TTL.

The field campaign in 2014 was based mainly in Guam and had three components: the Airborne Tropical Tropopause Experiment (ATTREX) project organized by NASA and based around the Global Hawk, the NCAR-led (National Center for Atmospheric Research) CONvective Transport of Active Species in the Tropics (CONTRAST) campaign based around the Gulfstream V (HIAPER) aircraft, and CAST with the UK's BAe146 research aircraft. Together, the three aircraft made detailed measurements of atmospheric structure and composition from the ocean surface to 20 km.

Our primary scientific interest in CAST is the transport of short-lived halogenated compounds from the ocean surface to the TTL. Once there, these compounds can enter the stratosphere in the Brewer-Dobson circulation, where their breakdown products affect the ozone layer. The very low ozone concentrations in the troposphere in the West Pacific mean that the lifetime of the halogenated compounds is much longer than elsewhere and more of them may be getting into the stratosphere than previously thought. Our measurements in Manus were to provide a good characterization of their concentrations at a remote, oceanic location.

There has been some recent controversy about ozone in the TTL, especially in the West Pacific. Ozonesonde profiles in this region have often reported ozone concentrations below the detection limit of the sonde (Kley et al 1996, 1997; Rex et al. 2014). Such low values have not been measured by aircraft or satellite sensors and have been challenged by other authors (e.g., Vömel and Diaz 2010) as arising from problems with the sonde's background current. Nevertheless, ozone concentrations in the West Pacific TTL do seem to be lower than anywhere else—indeed, by applying back-trajectory analysis to ozonesonde profiles measured over Darwin, Australia during the period from November 2005 to February 2006, Heyes et al (2009) proposed that ozone concentrations ~ 10 ppbv around 16 km originated from the region around the Solomon Islands.

The CAST ground-based component on Manus Island was designed to monitor ozonesonde profiles and ground-level chemical species for comparison with the aircraft, to resolve the controversy surrounding previous ozonesonde measurements, and to provide a data set to compare with global chemical models. It would also investigate further the hypothesis of Heyes et al (2009) about a source region for very low TTL ozone concentrations just to the east of Manus.

2.0 Notable Events or Highlights

Thirty nine ozonesonde packages were flown in all between February 2 and 25, 2014, with at least one ascent every day. Thirty four of these gave good ozone profiles and all but one gave good radiosonde profiles. We therefore collected a data set of unprecedented detail in a region of the atmosphere where such measurements have not previously been done. In addition, these ozonesonde measurements nicely complement the aircraft measurements made principally in the northern hemisphere. At the same time, ground-level concentrations of ozone were measured with a Thermo-49 analyzer; CO_2, CH_4, and CO were measured with a Picarro G2401 analyzer, and halogenated hydrocarbons were measured with a custom-built gas chromatograph.

The most notable events for the scientific interpretation of the Manus data set came from inter-comparison flights of the NCAR Gulfstream V aircraft, which allowed us to confirm that the method we used to correct the ozonesonde profiles for background current gave results that agreed with the aircraft. In turn, this means that we can recommend to the ozonesonde community that for well-prepared ozonesondes with background currents ~50 nA or less, the correct procedure is to subtract a constant value for the background current, measured in the laboratory just before launch.

With this result, we can say that ozone concentrations in the TTL were always > 10 ppbv, with the lowest values encountered when widespread deep convection occurred to the east of Manus. This concurs with the results of Heyes et al (2009) based on the Darwin ozonesonde data set acquired during the TWP-ICE/ACTIVE project (also supported by the Atmospheric Radiation Measurement [ARM] Climate Research Facility).

3.0 Lessons Learned

As always, there were plenty of challenges in getting to and operating in such a remote area, but because of the extensive help given by ARM Climate Research Facility staff, both locally and in the United States, we were able to overcome all our difficulties. Were it not for ARM's excellent infrastructure on Manus and the enthusiastic cooperation of all the staff, we would not have been able to achieve our objectives. We were sorry to learn on arriving on Manus that we would be the last people coming there for a campaign, as the site was decommissioned soon after we left.

4.0 Results

4.1 Introduction

Electrochemical Ozonesondes (ECCs) measure the ozone concentration by bubbling air through the cathode half of an electrochemical cell containing potassium iodide solution (Komhyr and Harris 1971). The current produced from this reaction is proportional to the amount of ozone passing through the cell, with each ozone molecule producing two electrons. In principle, this is an absolute measurement of the ozone flux through the cell; however, in practice there are other contributions to the ozone current that are not well understood. These produce a residual background current (Thornton and Niazy 1982), which

must be allowed for in the data processing. This background current is of particular importance in the TTL, where ozone concentrations are low and the background current can be a substantial fraction of the total current measured by the sonde. It is therefore important to determine which method of background current correction should be used for the TTL.

4.2 Ozonesonde Preparation

The ozonesondes used in this project were EnSci model Z sondes supplied by Droplet Measurement Technologies, coupled to Vaisala RS92-SGPD radiosondes that provided pressure, temperature, humidity, and wind profiles. Standard procedures for preparing ozonesondes follow a two-stage process aimed at measuring the sonde's pump flow rate and reducing the background current I_{bg} to < 50 nA at the time of launch. In this work, the background current was obtained by drawing air into the sonde through a charcoal filter in an air-conditioned cabin where relative humidity (RH) was < 50% at all times.

The ozonesonde preparation procedures involve, at different stages, purging the electrochemical cell and/or the pump with high concentrations of ozone, characterizing the cell response to expected atmospheric concentrations of ozone, and drawing ozone-free air through the cell. For this, a Science Pump TSC01 ozone calibration unit was used. Standard procedures normally follow manufacturer's guidelines. For this experiment, however, the procedures were adapted to reduce the background current as low as possible—in particular, introducing more changes of solution in the preparation than is normally recommended (the number of changes varied from sonde to sonde according to its requirements). The background current was taken to be the minimum value recorded by the Vaisala software after the sonde package was finally assembled, but before taking it out of the air-conditioned environment (in the humid tropical atmosphere outside the cabin, the charcoal destruction filter does not work correctly).

4.3 Contamination

A complication encountered during this experiment was the sudden appearance of a contamination source inside the TSC01 that produced a large signal from the ozonesonde. This badly affected the first two sondes, rendering their data unusable. Contamination also rendered the calibration cell on the TSC01 unusable. Sondes 3 and 4 were again clean on first preparation but were only very briefly exposed to the TSC01 on second preparation. The remaining sondes were not exposed to the TSC01 at all during the second preparation. Sondes 5-14 were, however, exposed briefly to ozone on first preparation and were subsequently found to have elevated background currents. Sondes 15 onwards were not exposed at all to the TSC01, and the background currents from these sondes were around 50 nA before launch.

Figure 1 shows how the background currents measured for each sonde varied during the campaign, compared to the minimum current measured by the cell in the TTL. During the latter part of the campaign the background current was around half the minimum measured in the TTL, but during the early part the minimum current is close to or even lower than the background, implying an impossible negative ozone concentration in the TTL. Given the consistency in the minimum measured values in the TTL, the most plausible hypothesis for the contaminated cells is that the effects of contamination disappeared during flight, leaving a background current of around 50 nA similar to that in the uncontaminated sondes. Note

also that the effects of contamination seem to have been reducing over time as the campaign proceeded: the measured background currents decreased from ~150 nA to < 100 nA between sonde numbers 5 and 14.

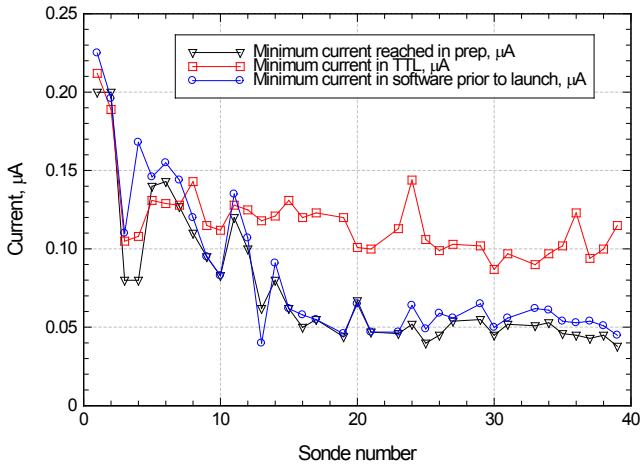

Figure 1. Background currents calculated with two different methods (blue, black) compared with minimum current measured by the sonde in the TTL (red) for the Manus sondes. Note how the effect of the contamination effect becomes less prominent between sondes 5 and 14.

On return from Manus, a series of laboratory experiments were conducted to ascertain the properties of the contamination that remained in the TSC01. The salient result is that for lightly contaminated sondes (such as 3-15) the effects of the contamination tended to disappear over a similar timescale, ~1 hour, to that taken by a sonde to reach the TTL. This is consistent with the hypothesis above about the background current. A hybrid background current correction was thus devised, $I_{bg} = I_{bg0} + (I_{bgm} - I_{bg0})\ p/p_0$ where I_{bgm} was the measured background current before launch, I_{bg0} = 50 nA, p the pressure, and p0 the pressure at the ground. A constant I_{bg} equal to the measured value in the laboratory was applied to the uncontaminated sondes from no. 15 onwards.

4.4 Ozonesonde Validation

Flights of the NCAR Gulfstream V were conducted in the vicinity of Manus Island on February 6 and 22 (flights RF09 and RF14 respectively). On February 6 the aircraft was unable to execute its original flight plan but managed to collect a profile just to the west of Manus Island, which is shown in Figure 2 below. This contaminated sonde had a background current of 143 nA, so the hybrid correction was used in the data analysis. In the left panel of Figure 2 the aircraft profile is compared with three ozonesonde profiles: one using a constant background current of 143 nA, (left dashed profile), one using the hybrid method (solid line) and one using a pressure-dependent background current (rightmost dashed curve). The profile with the hybrid correction agrees with the aircraft to around 3 ppbv. By contrast, applying a constant background current correction underestimates the amount of ozone—indeed, the ozone concentration goes negative at 150 hPa—while the pressure-dependent background current correction overestimates the ozone concentrations compared to the Gulfstream V. This comparison validates our choice of the hybrid correction.

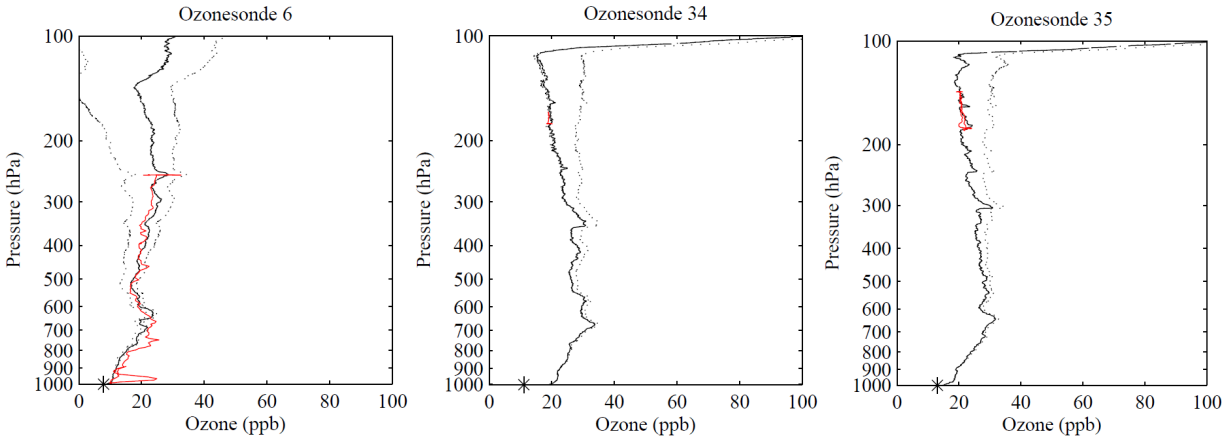

Figure 2. Comparison between aircraft measurements (red) and ozonesonde measurements (black) on February 6 (sonde 6) and 22 (sondes 34 and 35). Black star is the ground-level ozone measured by the TECO analyzer.

The other two sondes (34 and 35) are compared with southbound and northbound legs of the Gulfstream V passing just west of Manus on February 22. By now the contamination problem had been solved and a constant background current of around 50 nA (Figure 1) was applied in the analysis. The excellent agreement between aircraft and sonde supports the procedure that for a well-prepared sonde, with background current ~50 nA or less, a constant background current should be applied in the data analysis.

4.5 Summary of Ozonesonde Results

A summary of the results of the ozonesonde campaign is shown in Figure 3.They show the 'C' type tropospheric profiles typical of tropical stations, with low ozone concentrations in the boundary layer and at high latitude. The tropopause was generally around 16 km altitude (360 K potential temperature). The lowest TTL ozone concentrations of around 10 ppbv were measured towards the end of the campaign (with uncontaminated sondes) when deep convection was occurring to the east of Manus, near the Solomon Islands. The wind profiles show that these events corresponded to strong easterly flow, consistent with the outflow from deep convection.

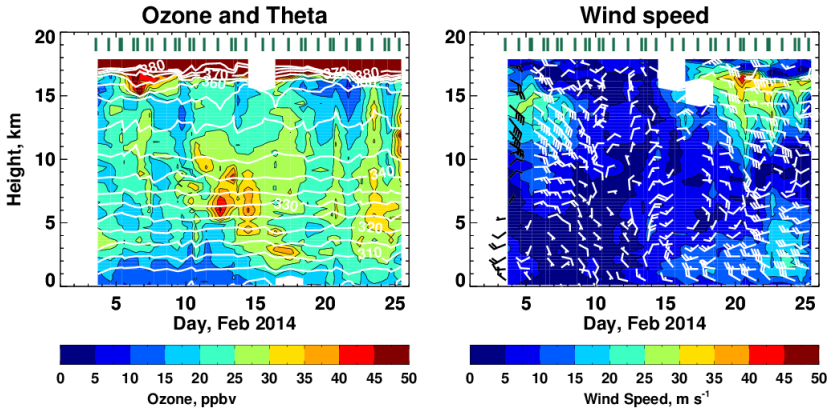

Figure 3. Time series of ozone profiles measured during the campaign between the ground and 17 km. Left panel: ozone (ppbv) and potential temperature (K, white); Right panel: Wind speed (coloured), wind barbs.

4.6 Summary of the Ground-Based Measurements

Daily meteorological measurements from the Momote station are summarized in Figure 4. They show that the first half of the campaign was characterised by dry weather, with increasing rainfall in the second half of the campaign. During the dry spell (February 1-10), ground-level ozone concentrations were very low (< 10 ppbv) with a very pronounced diurnal variation—night-time concentrations fell as low as 2 ppbv. Ozonesonde profiles during this period showed a rapid increase to ~ 20 ppbv in the first 300 m of the profile, showing that the layer of low ozone was shallow, and probably the result of local effects.

In the second half of the campaign, when convective conditions returned to Manus, ground-level ozone was higher (8-15 ppbv) without a pronounced diurnal variation. The wettest period, between February 20 and 22, was when the lowest TTL ozone was measured, with values similar to those at the surface, suggestive of widespread uplift of boundary-layer air in the large convective clusters to the east of Manus.

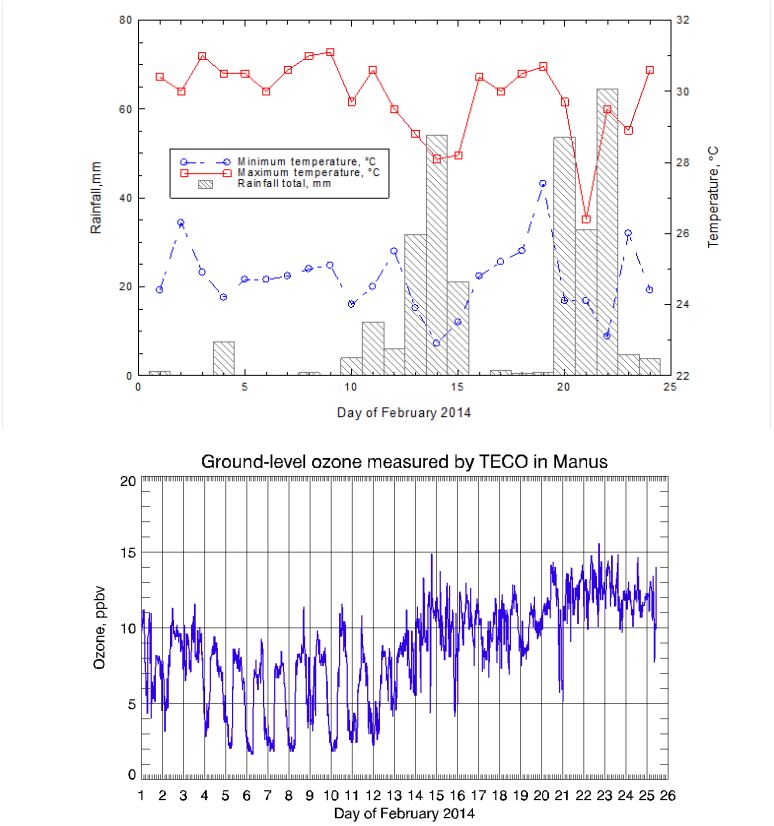

Figure 4. Top: Daily meteorological measurments from the Momote station (PNG weather service). Bottom: ground-level ozone measured by a TECO TE-49 ultraviolet (UV) absorption analyser.

In addition to the ground level ozone measurements, we also measured a range of halocarbons at the ARM site, from a sampling inlet ~10 m above the ground, using a custom-built gas chromatograph (μDirac) with electron capture detector (GC-ECD) (Gostlow et al 2010). Time series of four key short-lived compounds of natural marine origin are shown in **Figure 5**, together with wind speed data from the site. Bromoform ($CHBr_3$) and dibromochloromethane ($CHBr_2Cl$) have relatively long local atmospheric lifetimes of 24 and 59 days respectively, while methyliodide (CH_3I) and chloroiodomethane (CH_2ClI)

have much shorter lifetimes of 7 and 0.1 days respectively (Montzka and Reimann 2011). During the first half of the campaign, winds were often calm during the night, allowing compounds emitted at the surface to accumulate in the boundary layer. These events can be seen by night-time maxima in abundance, particularly in the first week of the campaign. The latter half of the campaign was characterized by windier surface conditions, even during the night, preventing significant build-up of locally emitted compounds in the boundary layer. The median dry air mole fractions for $CHBr_3$, $CHBr_2Cl$, CH_3I, and CH_2ClI were 0.81, 0.52, 0.3, and 0.17 ppt respectively and maximum dry air mole fractions were 8.6, 1.5, 2.5, and 0.5 ppt respectively.

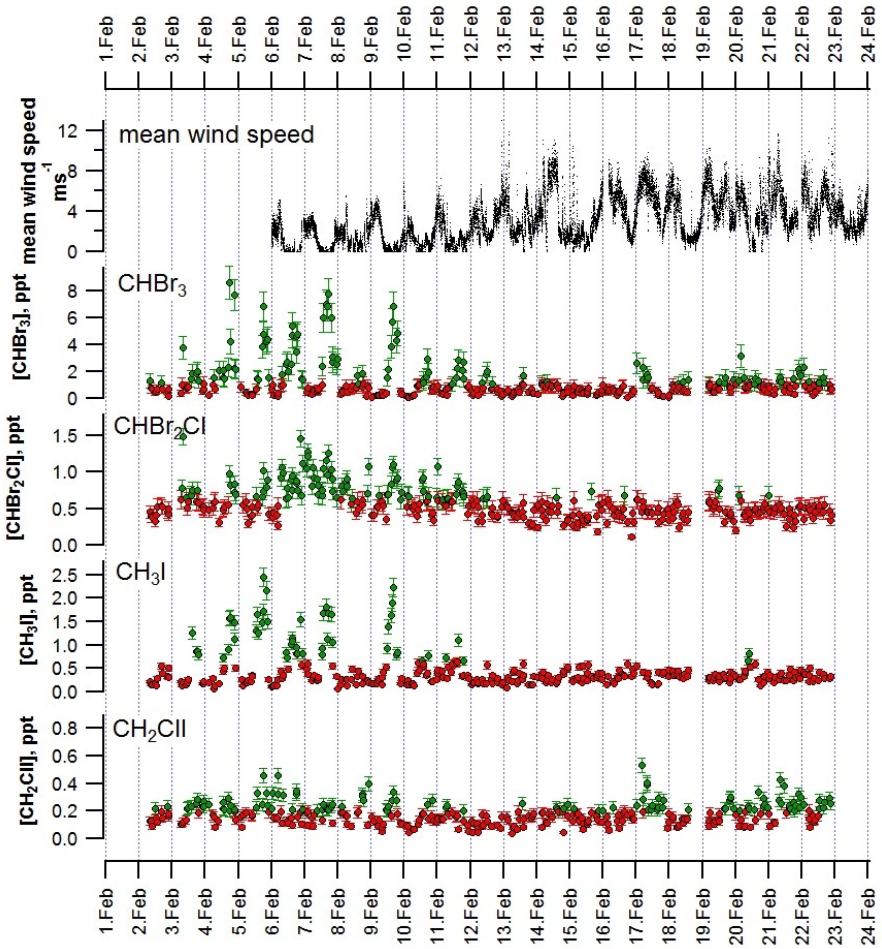

Figure 5. Time series of wind speed and short-lived halocarbons: $CHBr_3$, $CHBr_2Cl$, CH_3I, and CH_2ClI measured by the μDirac instrument at the Momote station.

We also ran a commercially available Picarro G-2401 cavity ring-down spectrometer on the ground using an inlet separate to the GC-ECD but at the same sampling height. This instrument produced calibrated time series of carbon dioxide (CO_2) and methane (CH_4) and an uncalibrated time series of carbon monoxide (CO) (**Figure 6**). Strong diurnal behaviors in CO_2 and CH_4 were evident, particularly during the first half of the campaign when winds were light during the night and local emission sources (likely from soil and vegetation) made a significant impact on boundary-layer abundance. CO, on the other hand, showed no noticeable diurnal behavior, but did show intermittent spikes that are attributed to aircraft movements and airport-related road traffic.

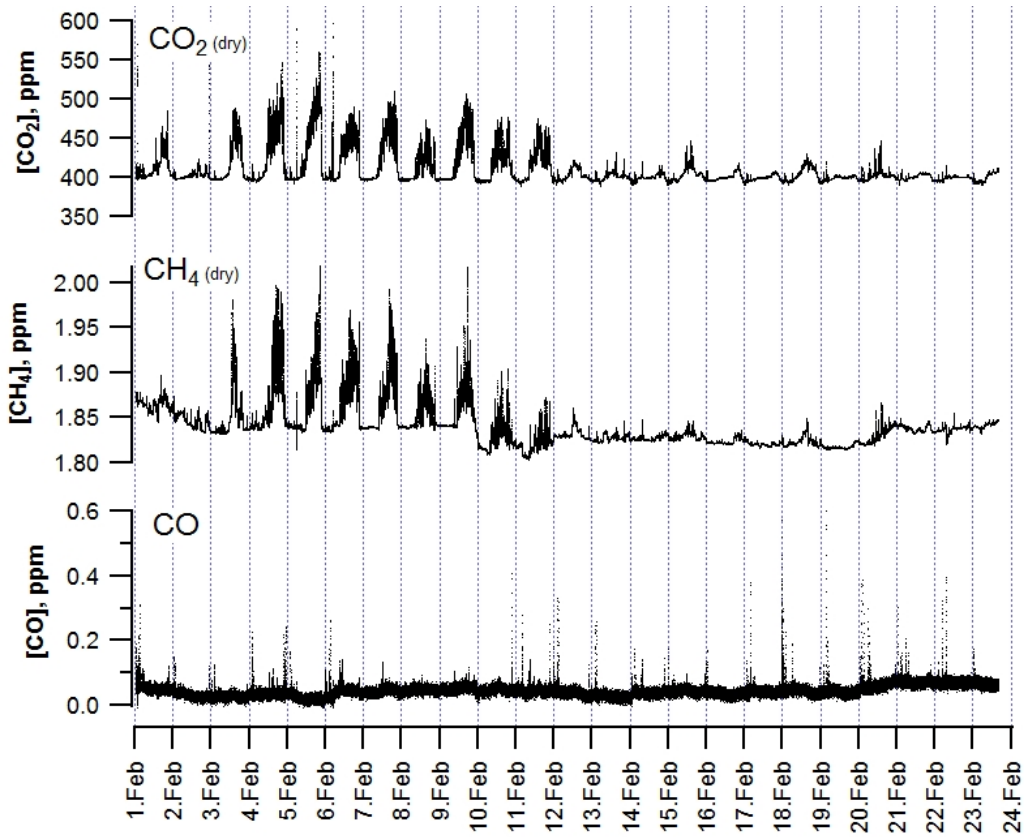

Figure 6. Time series of CO_2, CH_4, and CO measured by the Picarro instrument at the Momote station.

The methane time series shows a noticeable drop in baseline level around February 10/11. We believe that this may be the result of a change in air-mass origin arriving at the site. Our µDirac instrument also measured tetrachloroethene (C_2Cl_4), which is a good tracer of anthropogenic activity, having no significant natural sources (Ashfold et al 2014). Comparing the CH_4 time series with that of C_2Cl_4 shows a dip in both compounds around February 10/11 (**Figure 7**).

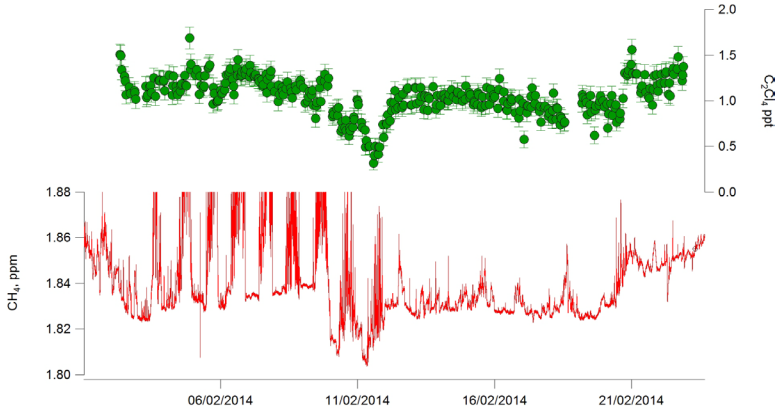

Figure 7. Time series of C_2Cl_4 from the µDirac instrument (top) and CH_4 from the Picarro (bottom) at the Momote station, clearly showing the dip in baseline levels around February 10/11.

5.0 CAST Publications

5.1 Meeting Abstracts/Presentations/Posters

Carpenter, LJ, and NRP Harris. 2014. "Co-ordinated airborne measurements in the tropics," 2014 AGU Fall meeting, poster https://agu.confex.com/agu/fm14/meetingapp.cgi#Paper/9373

Western Pacific Airborne Campaigns Science Team Meeting, Boulder, Co, 20-23 October, 2014:

- NRP Harris and the CAST team – "BAMS," in preparation.
- G Vaughan – "Ozonesonde measurements of low ozone during CAST"
- N. R. P. Harris – "Ground-based measurements of trace gases in Manus and the Western Pacific"
- R. Newton – "Tests on the CAST ozonesondes" (poster)

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