Co-ordinated Airborne Studies in the Tropics (CAST)

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Abstract
The main field activities of the CAST (Co-ordinated Airborne Studies in the Tropics) campaign took place in the West Pacific in January/February 2014. The field campaign was based in Guam (13.5°N, 144.8°E) using the UK FAAM BAe-146 atmospheric research aircraft and was coordinated with the ATTREX project with the unmanned Global Hawk and the CONTRAST campaign with the Gulfstream V aircraft. Together, the three aircraft were able to make detailed measurements of atmospheric structure and composition from the ocean surface to 20 km. These measurements are providing new information about the processes influencing halogen and ozone levels in the tropical West Pacific as well as the importance of trace gas transport in convection for the upper troposphere and stratosphere. The FAAM aircraft made a total of 25 flights between 1°S-14°N and 130°-155°E. It was used to sample at altitudes below 8 km with much of the time spent in the marine boundary layer. It measured a range of chemical species, and sampled extensively within the region of main inflow into the strong West Pacific convection. The CAST team also made ground-based measurements of a number of species (including daily ozonesondes) at the Atmospheric Radiation Measurement program site on Manus Island, Papua New Guinea (2.1°S, 147.4°E). This article presents an overview of the CAST project focussing on the design and operation of the West Pacific experiment. It additionally discusses some new developments in CAST, including flights of new instruments on the Global Hawk in February/March 2015.

Capsule: The Co-ordinated Airborne Studies in the Tropics (CAST) project is studying the chemical composition of the atmosphere in the Tropical Warm Pool region to improve understanding of trace gas transport in convection.

Introduction
The Tropical Tropopause Layer (TTL) is the region of the tropical atmosphere between the main convective outflow at ~12-13 km and the base of the stratosphere at 17-18 km and is a very important region for composition-aerosol-climate interactions (Randel and Jensen, 2013). Its overall structure is intermediate between the troposphere and stratosphere, with a lapse rate smaller than the saturated adiabatic up to the cold point (Fueglistaler et al., 2009). This is caused by the combined effect of slow radiative processes and the infrequent penetration of convective turrets to high
altitude. There is a marked longitudinal asymmetry in TTL temperatures, with a minimum in the region 130-180°E at all times of the year. This minimum corresponds to the warm waters of the Tropical Warm Pool (TWP) beneath, and there is an associated maximum in convection (Gettelman et al., 2002). The TTL is the predominant route for troposphere to stratosphere transport, so that conditions in the TTL set the entry concentrations at the base of the stratosphere for, e.g., stratospheric water vapour and very short-lived halogen species. Knowledge of the input into the TTL is a pre-requisite for correct modelling of TTL (and hence stratospheric) composition and yet many aspects are poorly constrained (Levine et al., 2007; Heyes et al., 2009). The coupling between the various processes are important. For example, improving the treatment of TTL water vapour and cirrus in global climate models requires a better understanding of convective transport and radiative transfer in the TTL, as well as improved model descriptions of the key processes.

We are still unclear about the entry and exit routes for the TTL, including how much material is transported quasi-horizontally into the extratropical lowermost stratosphere (Levine et al., 2008). What is the average residence time in the TTL? What is the nature, and importance for composition, of longitudinal variability within the TTL? How much of the very short-lived halogen species can pass through the TTL and so affect stratospheric ozone concentrations? Large discrepancies exist between models and measurements even for long-lived tracers. Some of these are due to transport – sharp horizontal gradients are observed in atmospheric tracers at boundaries between mid-latitude, subtropical and tropical airmasses which are not well represented by models (Wofsy et al., 2011) – and some to limited information on emissions, e.g. N₂O and CH₄ in this region (Ishijima et al., 2010). These issues are more important for very short-lived substances (VSLS - lifetimes < 6 months), including halogen-containing VSLS with their poorly understood sources, atmospheric transformations and geographic distribution (Carpenter, Reimann et al., 2014). Other effects such as the degree to which the locations of the emissions coincide with strong convection can also have a strong influence on the overall flux (Russo et al., 2015).

To address these issues, the Facility for Airborne Atmospheric Measurements (FAAM) BAe-146 atmospheric research aircraft was deployed in Guam in January and February 2014 as part of Coordinated Airborne Studies in the Tropics (CAST), a large multi-institutional project funded by the UK Natural Environment Research Council (NERC) and Science and Technology Facilities Council (STFC). In Guam, it flew alongside the NASA Global Hawk, a high altitude autonomous aircraft used in the NASA Airborne Tropical Tropopause Experiment (ATTREX) project, and the NSF/NCAR Gulfstream V (GV) in the NSF Convective Transport of Active Species in the Tropics (CONTRAST) project, as described in the companion papers, Jensen et al. (2016) and Pan et al.
The measurements from all three campaigns are being jointly used to diagnose how air is carried high into the atmosphere.

The value inherent in having the three aircraft flying together was to be able to measure from the surface up into the stratosphere (see Figure 1 in Pan et al., 2016). The instrument payloads on the three aircraft made many common measurements which together have combined to provide a comprehensive data set for interpretative studies. However within this larger picture, each aircraft had its own scientific aims and objectives which were appropriate to the specific aircraft capabilities. The Global Hawk made measurements in upper tropical TTL (14-20 km), including in the outflow of convection. The GV aircraft principally sampled at the same altitudes as the main convective outflow (9-15 km), and additionally made measurements on profiles down into the boundary layer. In the case of the FAAM aircraft, the aims were to (i) investigate halocarbon production in the marine boundary layer, and (ii) characterise the composition of air in the main convective inflow. Knowledge of the distributions of trace gases in the boundary layer and lower troposphere is needed to estimate the flux of these gases into the TTL. The role of the FAAM research aircraft was to fly over the tropical West Pacific and to measure the composition in the lower troposphere (0-8 km). These measurements characterise the air masses in the region of the main convective inflow and so are valuable in interpreting the higher altitude measurements of the Global Hawk and the GV made in the same period. They can also be used to improve understanding of marine halocarbon production and to investigate the influence of polluted outflow from Asia.

Additional measurements were made on Manus, Papua New Guinea. The majority of this paper describes the CAST measurements in January/February 2014, and the flight planning tools used for the FAAM aircraft and for linking its measurements with those made by the other aircraft. Some early results are also discussed. The second CAST goal is to develop the UK capability to use autonomous aircraft for atmospheric research. Here, in addition to learning about deploying the Global Hawk and using the data collected, CAST scientists have produced two new instruments for use on the Global Hawk which flew over the East Pacific in February/March 2015. These are described in the final section.

**CAST measurements**

Measurements were made on two main platforms in the West Pacific. The FAAM BAe-146 research aircraft was based at the A.B. Won Pat International Airport, Guam (13.5°N, 144.8°E). The FAAM aircraft was co-located with the NCAR Gulfstream while the NASA Global Hawk was based at Andersen Air Force Base approximately 30km to the north east. A suite of ground-based instrument systems was based at the Atmospheric Radiation Measurement (ARM) facility at...
Manus, Papua New Guinea (2.1°S, 147.4°E), in order to characterise the tropospheric composition beyond the range of the FAAM aircraft.

**Flight planning**

The goal of the CAST FAAM flights was to characterise the inflow to convection in the lower troposphere in the West Pacific. In order to extend the range of the aircraft so that it could reach into the upwelling area near the equator, overnight stops were planned at the islands of Palau (Roman Tmetuchl International Airport, Babeldaob island, Republic of Palau; 7.4°N 134.5°E) and Chuuk (Chuuk International Airport, Weno Island, Federated States of Micronesia; 7.5°N, 151.8°E). When conditions allowed, transects were made at 100 feet (with occasional dips down to 50 feet) over the open ocean to give the opportunity to sample air influenced by fresh ocean emissions. Stacked runs with horizontal legs at different altitudes were planned where possible to provide information about the vertical profile of the short-lived species in the lower troposphere. A large part of the flight planning for the FAAM research aircraft was to ensure a good coverage of the lower troposphere within range from Guam.

Chemical forecast products were provided by the Monitoring Atmospheric Composition & Climate (MACC) project in support of all three field campaigns. MACC assimilates comprehensive global observations of chemical composition into the ECMWF meteorological forecasting system (Flemming et al., 2015). The operational MACC system runs at 80 km horizontal resolution (T255) with 60 vertical levels. During the campaign, forecast plots for the operation domain were provided for a number of chemical species, including the FAAM measurements: O₃, CO, CH₄, black carbon, NO, and NO₂. In addition, a number of hypothetical tracers were included to track air originating from different locations, e.g. regional emissions from China and India. A coastal emission tracer was used to track oceanic emissions of CHBr₃ and other short-lived halocarbons since these are preferentially released in coastal regions (Carpenter et al., 2009; Ashfold et al., 2014).

**Linking measurements**

In order to have near-real-time information about the air reaching the TTL from the lower troposphere, the trajectory-based approach of Ashfold et al (2012) was adapted to meet the needs of a multi-aircraft campaign. In this, the Numerical Atmospheric-dispersion Modelling Environment (NAME) was run as an adjunct to the Met Office operational forecasting model so that it could access meteorological forecasts on a timescale quick enough to provide useful flight planning information. The starting grid for the trajectories covered a large area of the West Pacific (Figure 7), with trajectories being released at altitudes between 8 and 18 km. Twelve day backward
trajectories were then calculated using a mixture of Met Office analyses and forecasts, so that information was available about the possible influence of lower tropospheric air in the regions which could be sampled by the Global Hawk and the GV. Each day, trajectories were produced for 1, 2, 3 and 5 days in the future. In each 2 km altitude layer, 5,000 particles were released in each $10^\circ \times 10^\circ$ box. During the campaign, these calculations were made for a larger area at higher altitudes to reflect the larger range of the Global Hawk. The horizontal resolution of the Met Office operational model was 25 km in early 2014.

An example is shown in Figure 1 for three altitude ranges (12-14 km, 14-16 km, and 16-18 km). Each point is the end-point of each parcel of air that had crossed below 1 km in the preceding 12 days. For graphical clarity, only a fraction of the trajectories are shown at each level. Thus strong, predicted low level influence is indicated by a high percentage in each box (shown by the number), and at a given level by the denser clouds. These maps were routinely checked against flight plans for the Global Hawk and the GV to ensure that a wide range of low level influence was sampled. In general, most flight plans met this criteria due to the proximity of the aircraft to the main convective region.

**FAAM BAe-146 aircraft**

The FAAM BAe-146 has a science payload of up to 4 tonnes devised according to the objectives of a particular campaign. The chemical composition of the tropical atmosphere is the focus of CAST and this dictated the scientific payload. The chemical species and physical parameters measured on the FAAM aircraft, along with the instruments used, are summarised in Table 1. Trace gases with a wide range of atmospheric lifetimes, sources and sinks were measured in order to provide information about the origin and fate of the air masses encountered as well as about the atmospheric timescales involved. In many cases these species were also measured by the Global Hawk and/or the GV aircraft giving good synergy between the three datasets. Understanding the distribution and chemistry of halogen species is a special focus for all three campaigns and this is reflected in the FAAM payload.

Whole air samples (WAS) were collected as described in Andrews et al. (2013). Analysis of WAS canisters was carried out in the aircraft hangar, usually within 72 hours of collection. Two litres of sample air were pre-concentrated using a thermal desorption unit (Markes Unity2 CIA-T) and analysed with gas chromatography, mass spectrometry (GC-MS, Agilent 7890 GC, 5977 Xtr MSD). Halocarbons were quantified using a NOAA calibration gas standard. Dimethylsulfide was quantified using a secondary standard prepared and referenced to a primary KRISS standard. The full method is detailed in Andrews et al. (2013, 2016).
Measurements of a subset of halocarbons and other volatile organic compounds (VOCs) were made in-flight using a new thermal desorption (TD) GC-MS system. 1 L of sample air, drawn from a window blank inlet, pressurised to 2.5 atm and dried using a multi-core counter-current Nafion drier was alternately pre-concentrated or analysed from two parallel adsorption traps (Tenax TA) of a two channel TD system (Markes International, model TT 24/7). Analytes were refocussed at the head of the column using liquid CO₂ prior to separation (10 m, 180 micron I.D., 1 micron film, Restek RTX502.2 column; 40 to 150 °C at 40 °C min⁻¹) by GC (Agilent 6850) and detection by electron impact MS single ion monitoring (Agilent 5975C), calibrated pre-flight against the WAS gas standard (NOAA, SX-3581). Instrument temporal resolution, and associated sample integration period, was 5 min.

The chemical ionisation mass spectrometer (CIMS) from the Georgia Institute of Technology was configured similarly to previous deployments (Le Breton et al., 2012; 2013). The I⁻ ionization scheme was used to detect inorganic halogens, carboxylic acids, HCN and other trace species. For CAST, the CIMS made simultaneous measurements of BrO, BrCl, Br₂ and HOBr. The 1 Hz data were averaged to 30 s for analysis. Pre-campaign and post-flight laboratory calibrations were used relative to in-flight formic acid calibrations to quantify the sensitivities and limits of detection for the inorganic halogens, similar to that used for dinitrogen pentoxide (Le Breton et al., 2014). The sensitivities ranged from 1 to 50 ion counts ppt⁻¹ s⁻¹ determined by in-flight and post-campaign calibrations. The limits of detection for species varied from 0.36 ppt to 37 ppt for 30 s averaged data. (All mixing ratios given in this paper are by volume.) An acid scrubber was used to quantify background signal in the instrument and inlet line.

A broadband cavity-enhanced absorption spectrometer (BBCEAS) was adapted to measure IO in the 410-482 nm wavelength region. No clear absorption feature was observable from spectra by eye with up to 100 s averaging, pointing to very low mixing ratios (~0.5 ppt) of IO over the sampled area. When using averaged data, a small positive bias (~0.3 ppt) of IO was observed with respect to the zero. These observations appear to support the existence of IO in the remote marine boundary layer at sub-ppt levels, but the limited sensitivity precludes robust identification of spatial gradients.

NO was measured using chemiluminescence. NO₂ was quantified using a second channel, with NO₂ being converted to NO using a blue light LED converter centred at 395 nm. The NO₂ mixing ratio is derived from the difference between total NOₓ and NO mixing ratios. The instrument is calibrated via addition of 5 sccm of known NO concentration to the ambient sample. The conversion efficiency of the LED converter is measured in each calibration using gas phase titration of the NO to NO₂ on addition of O₃. In flight calibrations were conducted above the boundary layer to ensure
stable low levels of NO\textsubscript{x} with before and after flight calibrations made using an overflow at the inlet of zero grade air. A more detailed description of a similar system can be found in Lee et al. (2009). O\textsubscript{3} was measured by a UV absorption photometer (Thermo Fisher, model 49C), traceable to the UK National Physical Laboratory primary ozone standard with an uncertainty of 2%, and a precision of 1 ppb for 4 s measurements.

CO was measured by a vacuum UV fluorescence analyser (Aero Laser GmbH, model AL5002, Gerbig et al., 1999). The instrument was calibrated in-flight every ~45 minutes using a synthetic air working standard (Air Liquide, ~500 ppb), traceable to the NOAA-Earth System Research Laboratory (GMD-CGG) surveillance standard and the World Meteorological Organisation CO scale X2004. 1 Hz CO measurements have a 2% uncertainty and 3 ppb precision.

CO\textsubscript{2} and CH\textsubscript{4} were measured by a cavity-enhanced IR absorption spectrometer (Los Gatos Research Inc. Fast Greenhouse Gas Analyser, model RMT-200). The instrument was customised for airborne operation (O’Shea et al, 2013), so CO\textsubscript{2} and CH\textsubscript{4} dry mole fractions can be linearised in-flight using natural air working standards, traceable to the World Meteorological Organisation CO\textsubscript{2} scale X2007 and CH\textsubscript{4} scale X2004. The performance of the system is estimated from the 1σ standard deviation of all in-flight ‘target’ calibration data. The 1 Hz measurement precisions are estimated at 0.7 ppm and 2.5 ppb for CO\textsubscript{2} and CH\textsubscript{4}. Through the addition of all known uncertainties we estimate a total accuracy of ±1.3 ppb for CH\textsubscript{4} and ±0.2 ppm for CO\textsubscript{2}.

The Passive Cavity Aerosol Spectrometer Probe 100-X (PCASP), upgraded with the SPP-200 electronics package from Droplet Measurement Technologies (DMT), measures aerosol particles with nominal diameters 0.1 to 3 \(\mu\)m. Light from a 0.6328 \(\mu\)m laser is scattered by the particles and a photodetector sums the forward (over solid angles subtended by 35°-120°) and backward (60°-145°) scattered light. The probe is canister-mounted under the wing and was operated at 1 Hz. The instrument was calibrated for particle size before and after the campaign. Uncertainties exist in both the sizing and counting of particles and these are discussed, along with the calibration procedure, in Rosenberg et al. [2012].

The DMT Cloud Droplet Probe (CDP; Lance et al., 2010) was flown on the same under-wing pylon as the PCASP. The CDP is an open path instrument that measures the forward scattered light (over solid angles nominally subtended by 1.7°-14°) from the 0.658 \(\mu\)m incident laser beam. Particles are assigned to one of thirty size bins over the nominal size range 3-50 \(\mu\)m. Calibration with certified diameter glass beads was carried out before each flight (Rosenberg et al., 2012). The sample rate of the CDP was the same as for the PCASP, 1 Hz.
Observations started at the ARM Climate facility on Manus Island in October 1996 (Mather et al., 1998) and continued until August 2014. These observations provided the basis for many studies of the climate in the West Pacific (e.g., Long et al., 2013 and references therein). In February 2014, a suite of ground-based instruments was deployed as part of CAST to make measurements of ozone (ground and profile), short-lived halocarbons, carbon dioxide, carbon monoxide and methane. The instruments used are now described and are summarised in Table 2.

Ozone profiles were measured using ozonesondes. Air is pumped through a KI solution in a cathode half-cell, with two electrons produced for each ozone molecule; the cell current is directly proportional to the flow of ozone through the cell. Ozonesondes have a typical response time of ~ 1 minute at the tropopause level, with a precision of a few ppb. In the TTL the accuracy of the measurement is dominated by the background current (Newton et al., 2016 and references therein). Simultaneously, vertical profiles of temperature, humidity, wind and pressure were measured with Vaisala RS92 radiosondes.

Ground-level ozone was measured by a Thermo-Electric Corporation TE49C which is a dual-channel ultraviolet photometer measuring ozone through absorption of radiation at 254 nm. The incoming air stream is split between two identical cells, with a scrubber removing ozone from one of the streams. The TE49C provides a measurement every 10 s and has a 20 s response time.

Ground-level trace gas concentrations were measured by a Picarro Cavity Ring-Down Spectrometer G2401 (CRDS) (Crosson, 2008). The sample air inlet was at ~8 m above ground level with a rain cover and a 2 µm particulate filter. Water vapour in the instrument was kept below 1.5 ppm and was controlled by passing the sample flow (~250 mL min⁻¹) through a chiller at ~5 °C and then through a dessicant-based nafion drier. CO₂ and CH₄ concentrations were recorded every 5 s, with precisions of ~1 ppb and ~200 ppb respectively. Calibrations were achieved using a target gas (CH₄, 2024 ppb; CO₂, 390 ppm) measured every 2 days for 10 minutes with low / high calibration runs on intermediate days (low/high: CH₄, 1919/2736 ppb; CO₂, 360/495 ppm). The calibration gases are linked to the NOAA/WMO calibration scale.

Surface concentrations of short-lived halocarbons were measured using a µDirac instrument, a gas chromatograph with electron capture detector (GC-ECD) based on that described in Gostlow et al (2010) but with a 10 m separation column. The instrument sampled ambient air from the ~ 8 m high mast, with a 10-20 ml min⁻¹ flow dried using a counter flow nafion drier. Calibration runs, using a NOAA-ESRL air cylinder spiked with the target compounds, were conducted regularly (every 3 samples). The calibration volumes ranged from 3 to 50 ml to allow correction for drifts in
instrument sensitivity and linearity. Measurement precision is species dependent, typically 2-10 % (±1sd), with accuracy in the range 5-10 % (±1sd).

Overview of measurements

The FAAM BAe-146 made a total of 25 science flights with 90 flight hours during the CAST deployment in the West Pacific (Figure 2). Brief summaries of the flights are given in Table 3. The flight tracks are shown in Figure 2, with the altitude represented by the colour of the line. The large majority of the flights were below 5 km altitude, with a significant fraction in the marine boundary layer (below ~1 km), with good coverage between 130°E-160°E and 2°S-14°N.

The vertical distribution of the science flights can also be seen in Figure 3 which shows O3 and CO concentrations as a function of altitude and latitude. In general lower O3 values are found in the marine boundary layer and at lower latitudes, while high values are found at higher altitudes and at higher latitudes. There is no obvious correlation with CO. However when the O3 and CO data are plotted against each other (Figure 4), a bimodal relationship emerges. Further, the lower ozone values (10-40 ppb) occur when the relative humidity is high (Figure 4, top panel). This finding reinforces that of Pan et al. (2015) who report this bimodality throughout the altitude range covered by the NCAR GV, with a background mode of nearly constant (~20 ppb) values throughout the troposphere and a secondary mode of higher ozone (~35-95 ppb) in layers with lower relative humidity. The previously reported S-shaped mean profile (Folkins et al., 2002) results from averaging the two modes.

The CAST measurements (Figure 4) show that high ozone and lower relative humidity often occur with higher NO concentrations and do not occur with low CO concentrations. Preliminary analysis of the high NO measurements indicates that the air masses encountered had previously been in regions close to anthropogenic activities and/or biomass burning. For example, the MACC forecasts show transport of biomass burning and SE Asian tracers to the West Pacific. The possible role of biomass burning has been thoroughly investigated by Anderson et al. (2016) using CAST and CONTRAST measurements. The presence of HCN, CH3CN and other tracers in the high ozone levels is explained by biomass burning plumes which are convectively lofted into the free troposphere undergoing dehydration during the convection. As this air descends, its relative humidity drops and ozone is produced photochemically.

The CHBr3 concentrations measured with the Whole Air Sampler and the on-board GC-MS are shown in Figure 5. In general the values are low with even the higher values not far above the background values seen in this region (Brinckmann et al., 2012). The lower amounts of CHBr3 were encountered out of the boundary layer (Figure 4b). The background in Figure 2 shows that the
Chlorophyll-a concentrations in the surface waters of the West Pacific were low in this period. Higher Chl-a values are seen in the shallower waters approaching the islands of the Maritime Continent. The lagoon inside Chuuk atoll is relatively shallow (<60 m) and is embedded in much deeper ocean waters. It has a circumference of ~200 km and an area of ~3000 km². If halocarbons are emitted preferentially in shallow waters (Carpenter et al., 2009), then it should be discernible as an emission hotspot. The influence of short-lived halocarbon emissions from shallower waters was investigated in the FAAM flights by circling Chuuk atoll at low altitudes. The inset of Figure 5a shows the CHBr₃ observed on these flights as well as the instantaneous wind speed observed by the FAAM aircraft. Higher concentrations of CHBr₃ (red) are found when air has previously passed over the atoll, indicating that the atoll is a source of CHBr₃.

The NAME model driven by Met Office analysed fields has been used to interpret the CHBr₃ and other brominated VSLS measurements made near the tropopause on the Global Hawk in the East Pacific in 2013 and the West Pacific in 2014 (Navarro et al., 2015). The approach is similar to the forecast information produced during the campaign (see above). They find that the majority of air recently injected into the TTL had come from the West Pacific in both years with similar amounts, ~6 (4-9) ppt, of combined organic and inorganic bromine derived from brominated VSLS.

CHBr₃ was also observed at the ARM facility in Manus (Figure 5). The median value in this period was 0.81 ppt, about half that previously observed at a coastal site in Malaysian Borneo (Robinson et al., 2014) and similar to the values observed on the FAAM aircraft (Figure 4). A strong diurnal cycle is seen in early February in several trace gases measured at Manus with increased nocturnal amounts providing evidence for local night-time sources of CO₂, CH₄, CHBr₃ and CH₃I. This diurnal behaviour was seen from February 3rd to 12th when the winds were low and a stable boundary layer was able to form. Before and after this period winds were higher and the night-time build-up was much less.

Together, the CHBr₃ observations appear consistent with past work focussed on Southeast Asia. Elevated levels are frequently observed close to coasts (e.g. Pyle et al., 2011) or above shallow waters, but measurements collected a relatively small distance away (less than a typical global model grid cell) rarely contain above background levels of CHBr₃. This suggests that coasts are not a large source in a regional/global sense (as found by Ashfold et al., 2014), and for coastal CHBr₃ emissions to contribute significantly to the TTL and stratosphere would require co-location of convection (Russo et al., 2015).

Ground-based ozone at Manus showed decreases at night in the quiescent period from a peak daytime value of 10 ppb to sub 5 ppb levels which are consistent with oxidative uptake to the local vegetation (Figure 6). This is the only time such low values of ozone were seen in CAST. In the
absence of local sources, C2Cl4 is a good tracer of large scale transport, and its concentrations in this period were generally in the range 1-1.5 ppt which are typical of those seen in the clean West Pacific (Ashfold et al., 2015). Manus was mainly influenced by flow from the north in this period. A total of 39 ozone sondes were launched from Manus in February 2014, with 34 sondes providing good ozone profiles (Figure 7(a); Newton et al., 2016). These measurements are hardest in the tropics as the ozone concentrations are low, so that any error in estimating the background current is important. Particular attention was therefore paid to measurements of the background current, leading to recommendations for changes to the standard operation procedures used in the sonde preparation. Support for this approach is provided by good agreement in a coordinated ozonesonde / GV flight (see Figure 14 in Pan et al., 2016). The ozone measurements are shown in Figure 7 alongside the corresponding MACC 1 and 4 day forecasts. The forecasts predicted the main characteristics of the observations such as increased ozone at about 400 hPa from 14-16 Feb and the low concentrations near the TTL from 19-23 Feb. The minimum reproducible ozone concentration measured in the TTL was 12 ppb, consistent with the minimum of 13 ppb measured by the GV during CONTRAST (Pan et al., 2016).

New technology developments

As part of the collaboration with ATTREX, three new developments were included in CAST: two instruments for use on the Global Hawk, the Aerosol-Ice-Interface Transition Spectrometer (AIITS) and the GreenHouse gas Observations in the Stratosphere and Troposphere (GHOST); and a software tool, Real-time Atmospheric Science Cluster AnaLysis (RASCAL), designed to assist aircraft scientists by performing real-time data analysis during flights. The two new instruments were flown for a total of 40 hours in one test flight and two science flights in February-March 2015 from the NASA Armstrong Flight Research Center, California. They were part of a payload which also included Hawkeye, the NOAA H2O and O3 instruments, the Global Hawk Whole Air Sampler (GWAS), and Microwave Temperature Profiler (MTP) (see Jensen et al, 2016 for more details).

The Aerosol-Ice-Interface Transition Spectrometer (AIITS) was designed to probe different cirrus regimes in the TTL in order to understand fundamental nucleation and sublimation processes influencing the stratospheric water budget and fluxes, as well as the potential impact of biomass burning on cirrus ice crystal activation and growth. It is the next instrument in the Small Ice Detector (SID) family (Hirst et al., 2001; Kaye et al., 2008). AIITS acquires 2-D forward scattering patterns from particles in the size range from about one to a few hundred micrometres and can measure the depolarisation in backward and forward scattering. The patterns allow quantification of the phase, habit and fine surface features of large aerosol and small ice crystals in the size range 2-
100 µm (Cotton et al., 2010; Ulanowski et al., 2014). Unique results were obtained by AIITS during cirrus penetrations at 16.5 km and at temperatures down to -80°C (Figure 8). These revealed a transition to smooth quasi-spherical ice particle regimes in specific regions of TTL layers in response to changing supersaturation regimes. The impact on the radiative scattering properties of cirrus in these regimes is being investigated.

GHOST is a novel grating spectrometer designed for remote sensing of greenhouse gases from aircraft (Humpage et al., 2014). It measures spectrally-resolved shortwave-infrared radiance across four spectral bands from 1.27 µm to 2.3 µm, with a spectral resolution between 0.1 and 0.3 nm. An optical gimbal underneath the aircraft is programmed to pass solar radiation reflected from the ocean surface through a fibre optic bundle into the spectrometer with a single grating and detector for all 4 bands. The bands are chosen to include absorption bands for CO₂ and CH₄ as well as CO, H₂O and O₂. O₂ is used to infer information on the scattering contributions towards the measured light. The third Global Hawk flight of the CAST/ATTREX campaign targeted the overpasses of two greenhouse gas observing satellites during clear sky conditions over the Eastern Pacific (Figure 9); the NASA Orbiting Carbon Observatory (OCO-2) and the JAXA Greenhouse gas Observing SATellite (GOSAT). This Global Hawk flight therefore provides a very useful validation dataset for these satellites, since they both make greenhouse gas measurements using a similar spectral range to GHOST.

As real-time data becomes increasingly available, mission scientists are faced with a potentially overwhelming data torrent from which they are required to find the information on which to base decisions. At present, mission scientists often focus on a subset of the data stream, limiting the depth of the analysis which can be carried out. As part of CAST, a new software framework, RASCAL, has been developed based on recent developments in arbitrarily-shaped cluster detection algorithms (Hyde and Angelov, 2015). It interfaces intuitively with mission scientist expert knowledge and provides real-time on-the-fly cluster and anomaly detection (i.e. for real-time diagnosis of structures such as those diagnosed in Figure 4, for example, but tested simultaneously across many chemical 'dimensions'). The data stream can be separated in real-time, without a priori assumptions about parameter relationships, to reveal different data groups and hence isolate specific regions of interest that can be revisited virtually post-flight. In combination with the expert knowledge of the mission scientists, support tools like RASCAL have the potential to be used on many research aircraft, potentially adding significant value to the results achieved in field measurement campaigns.

Summary
Based in Guam as part of a joint deployment with the NASA ATTREX Global Hawk and the NSF CONTRAST GV, the FAAM research aircraft deployment in CAST has provided an excellent characterisation of the lower tropospheric atmospheric composition in the Tropical Warm Pool region. The majority of the FAAM aircraft flights were below 5 km altitude, and a significant fraction was in the marine boundary layer with good coverage in 130°E-160°E and 2°S-14°N. A suite of organic and inorganic halogen compounds was measured, with the bromine-containing species particularly well covered.

Ground-based measurements were made at the ARM facility on Manus Island, Papua New Guinea during February 2014. These measurements characterise the tropospheric composition just south of the equator in a region inaccessible to the FAAM aircraft in this deployment. The Manus ozonesonde measurements are a valuable resource, providing a good picture of the vertical distribution of ozone in the Tropical Warm Pool region during February with a minimum ozone concentration in the TTL of 12 ppb.

These measurements are being interpreted by CAST scientists in conjunction with measurements from ATTREX and CONTRAST using a range of modelling and data analysis approaches. The CAST data are stored at the British Atmospheric Data Centre (http://badc.nerc.ac.uk/), and interested parties are encouraged to use them for their own studies. All users are strongly encouraged to involve the responsible instrument scientists in these studies in order to have insight into the strengths and weaknesses of these data.

Never before has the atmosphere over the West Pacific been observed in such detail, particularly the chemical composition, with three aircraft covering all altitudes from 0 to 20 km. New insights are starting to emerge with much improved understanding of the tropical ozone distribution (Pan et al., 2015; Anderson et al., 2016; Newton et al., 2016). These will be underpinned by advances in the understanding of halogen distribution (Navarro et al., 2015) and chemistry building on the new tropospheric halogen measurements (Le Breton et al., 2016) and modelling (Sherwen et al., 2016). Such research will lead to a much greater quantitative understanding of the role of (a) VSLS reaching the stratosphere and (b) how halogen chemistry affects tropospheric ozone over the tropical oceans. Similar advances can be expected with respect to transport and dynamics, the role of cirrus cloud in climate and dehydration of the stratosphere. The benefits of this unique, coordinated campaign are just starting to become clear.

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NE/J00619X/1, and NE/J006173/1. N. R. P. Harris was supported by a NERC Advanced Research Fellowship (NE/G014655/1). P. I. Palmer acknowledges his Royal Society Wolfson Research Merit Award. The BAe-146-301 Atmospheric Research Aircraft is flown by Directflight Ltd and managed by the Facility for Airborne Atmospheric Measurements, which is a joint entity of the Natural Environment Research Council and the Met Office. The authors thank the staff at FAAM, Directflight and Avalon Aero who worked so hard toward the success of the aircraft deployment in Guam, especially for their untiring efforts when spending an unforeseen 9 days in Chuuk. We thank the local staff at Chuuk and Palau, as well as the authorities in the Federated States of Micronesia for their help in facilitating our research flights. Special thanks go to the personnel associated with the ARM facility at Manus, Papua New Guinea without whose help the ground-based measurements would not have been possible. Thanks to the British Atmospheric Data Centre (BADC) for hosting our data and the NCAS Atmospheric Measurement Facility for providing the radiosonde and ground-based ozone equipment. Chlorophyll-a data used in Figure 1 were extracted using the Giovanni online data system, maintained by the NASA GES DISC. We acknowledge the MODIS mission scientists and associated NASA personnel for the production of this data set. Finally we thank many individuals associated with the ATTREX and CONTRAST campaigns for their help in the logistical planning, and we would like to single out Jim Bresch for his excellent and freely provided meteorological advice.

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Table 1: Instruments and measurements made on the BAe 146 (FAAM) aircraft during the CAST project. The table also indicates the synergy with other aircraft from the CONTRAST (Gulfstream-V (GV)) and ATTREX (Global Hawk (GH) projects.

<table>
<thead>
<tr>
<th>Species / parameter</th>
<th>Method / instrument details</th>
<th>Averaging time</th>
<th>Precision, accuracy</th>
<th>Synergy with other aircraft</th>
<th>Affiliation, reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Position, winds, u, v, w</td>
<td>INS, GPS, 5-port turbulence probe</td>
<td>0.1 s</td>
<td>0.01 Δ P/P</td>
<td>GV, GH</td>
<td>FAAM Peterson and Renfrew (2009)</td>
</tr>
<tr>
<td>Humidity (Dew point T)</td>
<td>Hygrometer, General Eastern 1011b</td>
<td>0.25 s</td>
<td>± 0.5 - ± 3 K dependent on dew point and ambient conditions</td>
<td>GV, GH</td>
<td>FAAM Ström et al. (1994)</td>
</tr>
<tr>
<td>Temperature</td>
<td>Rosemount Aerospace Ltd. sensor 102 AL</td>
<td>.05 s</td>
<td>± 0.3K</td>
<td>GV, GH</td>
<td>FAAM Lenschow (1986)</td>
</tr>
<tr>
<td>CO</td>
<td>VUV resonance / fluorescence, Aerolaser 5002</td>
<td>1 s</td>
<td>±1 ppb, 3%</td>
<td>GV, GH</td>
<td>FAAM Gerbig et al. (1999)</td>
</tr>
<tr>
<td>O3</td>
<td>UV absorption, TEI 49C</td>
<td>4 s</td>
<td>±1 ppb, ±5%</td>
<td>GV, GH</td>
<td>FAAM Wilson and Birks (2006)</td>
</tr>
<tr>
<td>CO2, CH4</td>
<td>Cavity enhanced absorption spectrometer, Los Gatos Research Inc</td>
<td>1 s</td>
<td>CH4: 2.5 ppb; 1.3 ppm CO2: 0.7 ppm; 0.2 ppm</td>
<td>GV, GH</td>
<td>FAAM / U. Manchester O'Shea et al. (2013)</td>
</tr>
<tr>
<td>NO, NO2</td>
<td>Chemiluminescence with photolytic conversion for NO2, Air Quality Design Inc.</td>
<td>10 s</td>
<td>5 pptv for NO and 15 pptv for NO2 (at 10 s averaging)</td>
<td>GV</td>
<td>FAAM / U. York Lee et al. (2009)</td>
</tr>
<tr>
<td>Halocarbons (Whole air samples (WAS)): (DMS, CHBr3, CH2Br2, CHBrCl, CHCl3, CHBrCl2, CH2BrCl, HCl, CH3Br, CH3Cl, CH2Cl2, CHCl3)</td>
<td>TD-GC-MS, Markes</td>
<td>30 s fill time for WAS</td>
<td>Species dependent, typically 0.1 – 1 pptv.</td>
<td>GV, GH</td>
<td>U. York Andrews et al. (2013; 2016)</td>
</tr>
<tr>
<td>NMHCs (Whole air samples (WAS)): (C1–C7) NMHCs (alkanes, alkenes, aromatics); small α-VOCs (acetone, methanol, acetaldehyde, ethanol); DMS</td>
<td>GC-FID (flame ionization detector), Perkin Elmer</td>
<td>30 s fill time for WAS</td>
<td>Species dependent, typically 5 pptv</td>
<td>GV, GH</td>
<td>U. York Hopkins et al. (2003)</td>
</tr>
<tr>
<td>Halocarbons, VOCs (in situ)</td>
<td>GC-MS (Gas Chromatography – Mass Spectrometry), Agilent</td>
<td>300 s</td>
<td>Species dependent, typically 1 – 5 pptv.</td>
<td>GV</td>
<td>U. York</td>
</tr>
<tr>
<td>BrO, Br2, HOBr, BrCl, HCOOH (formic acid), HCN, CINO2, HNO3, N2O5, CH3COOH (acetic acid), CH3CH2COOH (propanoic acid), CH3CH2CH2COOH (butanoic acid)</td>
<td>Chemical Ionisation Mass Spectrometer (CIMS)</td>
<td>30 s</td>
<td>Species dependent, typically 0.3 – 5 ppt</td>
<td>GV</td>
<td>U. Manchester Le Breton et al. (2012)</td>
</tr>
<tr>
<td>IO</td>
<td>Broadband Cavity Enhanced Absorption Spectrometer (BBCEAS)</td>
<td>see text</td>
<td>see text</td>
<td>GV (IO remote sensing)</td>
<td>U. Cambridge Kennedy et al. (2011)</td>
</tr>
<tr>
<td>PAN</td>
<td>Dual column GC-ECD</td>
<td>90 s</td>
<td>3%, 10%</td>
<td>GV</td>
<td>U. York Whalley et al. (2004)</td>
</tr>
<tr>
<td>Black carbon</td>
<td>Soot particle photometer (SP-2)</td>
<td>10 s</td>
<td>None</td>
<td>U. Manchester, Liu et al. (2015)</td>
<td></td>
</tr>
</tbody>
</table>
Table 2: Measurements made at the ARM site at Manus, Papua New Guinea during CAST

<table>
<thead>
<tr>
<th>Species / parameter</th>
<th>Method / instrument details</th>
<th>Operation</th>
<th>Precision, accuracy</th>
<th>Affiliation, reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>O$_3$ (profile)</td>
<td>Ozone sonde, ENSCI model Z from DMT</td>
<td>Daily</td>
<td>see Newton et al. (2016)</td>
<td>U. Manchester, NCAS Newton et al. (2016)</td>
</tr>
<tr>
<td>O$_3$ (surface)</td>
<td>Thermo-49 analyser</td>
<td>Continuous (10 sec)</td>
<td>± 1 ppbv, precision-limited</td>
<td>NCAS, Atmospheric Measurement Facility</td>
</tr>
<tr>
<td>CO$_2$, CH$_4$</td>
<td>Picarro G2401 CRDS analyser</td>
<td>Continuous (5 sec)</td>
<td>CO$_2$ precision 0.05 %, accuracy 0.05 % (±1sd); CH$_4$ precision 0.05 %, accuracy 0.1 % (±1sd);</td>
<td>U. Cambridge Crosson (2008)</td>
</tr>
<tr>
<td>Halocarbons: (CHBr$_3$, CHBr$_2$Cl, CH$_3$I, CH$_2$ICl, CCl$_4$)</td>
<td>Custom-built GC-ECD</td>
<td>Continuous (~50 minutes)</td>
<td>Species dependent, typically 0.1 – 1 pptv.</td>
<td>U. Cambridge Gostlow et al. (2010), Robinson et al. (2014)</td>
</tr>
</tbody>
</table>

Information about the meteorological measurements from Manus can be found at [http://www.arm.gov/sites/twp/C1/instruments](http://www.arm.gov/sites/twp/C1/instruments).
<table>
<thead>
<tr>
<th>Flight no.</th>
<th>Date</th>
<th>Route</th>
<th>Flight description and observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>B823</td>
<td>18/1/14</td>
<td>Kota Kinabalu - Palau - Guam</td>
<td>Measurements on last part of leg from KK to Palau. Flight mainly at low levels (in boundary layer) on Palau to Guam leg. O&lt;sub&gt;3&lt;/sub&gt; and CO decreasing further North (O&lt;sub&gt;3&lt;/sub&gt; 30-12 ppb), higher (&gt;35ppb) above boundary layer (BL).</td>
</tr>
<tr>
<td>B824</td>
<td>22/1/14</td>
<td>Guam – Guam</td>
<td>Heading SE from Guam. 4000 m then 2000 m, flight aborted early due to aircraft technical problem. GV followed around 30 minutes later. O&lt;sub&gt;3&lt;/sub&gt; 15 ppb near Guam, falling to 10 ppb at 7°S.</td>
</tr>
<tr>
<td>B825</td>
<td>24/1/14</td>
<td>Guam – Chuuk</td>
<td>Mixed altitudes (lowest 300 m), mainly within BL. O&lt;sub&gt;3&lt;/sub&gt; dropping from 15 ppb to 8 ppb towards Chuuk. CO ~105 ppb on whole flight. SE flow.</td>
</tr>
<tr>
<td>B826</td>
<td>25/1/14</td>
<td>Chuuk – Chuuk</td>
<td>Due South from Chuuk on 152°E to 2°N, back on 153°E. Start at 6000 m then step down to 300 m. O&lt;sub&gt;3&lt;/sub&gt; constant (~15 ppb) in boundary layer, 25 ppb above BL. Largely SE flow in BL, W-NW in free troposphere.</td>
</tr>
<tr>
<td>B827</td>
<td>26/1/14</td>
<td>Chuuk – Chuuk</td>
<td>Due South from Chuuk on 152°N to 1°N then return on same track. In BL to 1°N, 4000 m on return North. Well mixed boundary layer. 20 ppb O&lt;sub&gt;3&lt;/sub&gt; to 1°N. BrO and CH₂Cl₂ observed. Largely SE flow in BL, W-NW in FT.</td>
</tr>
<tr>
<td>B828</td>
<td>26/1/14</td>
<td>Chuuk - Guam</td>
<td>Circled atoll at 100 m and 1500 m; then mixed altitude down to 300 m on way back to Guam. CO 100ppb round atoll in BL, O&lt;sub&gt;3&lt;/sub&gt; 15 ppb. O&lt;sub&gt;3&lt;/sub&gt; 10-13ppb as head North towards Guam.</td>
</tr>
<tr>
<td>B829</td>
<td>29/1/14</td>
<td>Guam - Palau</td>
<td>Mixed levels in BL down to 300 m. Low O&lt;sub&gt;3&lt;/sub&gt; (12 ppb) observed around island of Yap. Easterly flow.</td>
</tr>
<tr>
<td>B830</td>
<td>29/1/14</td>
<td>Palau – Palau</td>
<td>Flight East along 7°N; mixed altitude down to 300 m; 4 stacked runs above each other at easterly end. Profile of BrO observed on stacked runs - higher at surface. Same CO and O&lt;sub&gt;3&lt;/sub&gt; profile at all levels so well mixed BL. 45 ppb O&lt;sub&gt;3&lt;/sub&gt; and some NOx (25 ppt) seen at 4000 m. Higher N&lt;sub&gt;2&lt;/sub&gt;O at higher altitudes. Largely SE flow.</td>
</tr>
<tr>
<td>B831</td>
<td>30/1/14</td>
<td>Palau – Palau</td>
<td>Flight SE to Indoneisan airspace (4°30’N, 141°30’E) then due South to 3°N. Mainly in BL, down to 300 m at most Southern point where O&lt;sub&gt;3&lt;/sub&gt; was 25-30 ppb. Westerly flow so some Asian outflow observed (CO &lt; 100 ppb).</td>
</tr>
<tr>
<td>B832</td>
<td>30/1/14</td>
<td>Palau – Guam</td>
<td>Low level runs in BL crossing day/night terminator. 30m in early part of flight before hitting low level convection. Above BL towards Guam. 15 ppb O&lt;sub&gt;3&lt;/sub&gt; during sunset - very constant as heading North. NW flow.</td>
</tr>
<tr>
<td>B833</td>
<td>1/2/14</td>
<td>Guam – Guam</td>
<td>1&lt;sup&gt;st&lt;/sup&gt; part of day/night chemistry flights. Stacked legs to E of Guam: 6000, 3000, 1500, 1000, and 300 m. NE flow. Followed GV for first half of flight (~30 minutes behind).</td>
</tr>
<tr>
<td>B834</td>
<td>1/2/14</td>
<td>Guam – Guam</td>
<td>2&lt;sup&gt;nd&lt;/sup&gt; part of day/night chemistry flights. Stacked legs to E of Guam: 6000, 3000, 1500, 1000, and 300 m. NE flow.</td>
</tr>
<tr>
<td>B835</td>
<td>4/2/14</td>
<td>Guam – Chuuk</td>
<td>Fast transit to Chuuk above BL. 25 ppb O&lt;sub&gt;3&lt;/sub&gt;, 85 ppb CO at 6000 m, then O&lt;sub&gt;3&lt;/sub&gt; lower as dropping down to Chuuk (~13 ppb).</td>
</tr>
<tr>
<td>Flight Number</td>
<td>Date</td>
<td>Route</td>
<td>Description</td>
</tr>
<tr>
<td>--------------</td>
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<td>---------------------------------------------------------------------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>B836</td>
<td>4/2/14</td>
<td>Chuuk – Chuuk</td>
<td>Head S along 152°E at 7000 m, some low flying in BL to southernmost point (1°S) before intermediate height (2000 – 4000 m) back to Chuuk. 18 ppb O₃ above BL to 1°N. Then profile down and less O₃ in BL (13 ppb), CO 70 ppb. At 1°S O₃ 9 ppb in NE flow.</td>
</tr>
<tr>
<td>B837</td>
<td>5/2/14</td>
<td>Chuuk – Chuuk</td>
<td>Low level flying in BL to southernmost point (~1°N) to complement B836), then climb back and return at ~5000 m. O₃ decreasing in BL as head South. 20 ppb at 7°N, 11 ppb at 1°N. All in NE flow.</td>
</tr>
<tr>
<td>B838</td>
<td>6/2/14</td>
<td>Chuuk – Chuuk</td>
<td>Round Chuuk atoll at 3 altitudes in BL (150, 500, and 1000 m). CO higher to East of Islands (Easterly flow). Could be storms over the islands bringing elevated CO to the upwind side.</td>
</tr>
<tr>
<td>B839</td>
<td>12/2/14</td>
<td>Chuuk – Guam</td>
<td>SE of Guam at low level (500 m in BL), then above BL (5000 m) before descending down at lower levels in BL into Guam. O₃ spikes in profiles up to 7500 m (Asian outflow). 75 ppb seen at 7000 m.</td>
</tr>
<tr>
<td>B840</td>
<td>13/2/14</td>
<td>Guam – Palau</td>
<td>Start in FT (~6500 m), then low nearer Palau (1500 m); head to 4°N, 137°E before heading NW; same region as GV and GH. O₃ 30 ppb in FT, 12 ppb in BL, very stable. Easterly flow.</td>
</tr>
<tr>
<td>B841</td>
<td>14/2/14</td>
<td>Palau – Palau</td>
<td>Flight to SW of Palau with stacked legs in BL parallel to ATC boundary. O₃ 15 ppb in BL. Easterly flow.</td>
</tr>
<tr>
<td>B842</td>
<td>14/2/14</td>
<td>Palau - Guam</td>
<td>Reverse flight to B840. Similar flow and O₃.</td>
</tr>
<tr>
<td>B843</td>
<td>16/2/14</td>
<td>Guam – Guam</td>
<td>S from Guam to 7°N then E towards Chuuk before heading back to Guam; low latitude parts at low altitudes (&lt;1000 m in BL) under convective band. O₃ 10-15 ppb in BL (E flow), elevated at higher levels (70-90 ppb), concurrent with elevated NO (30 ppt) (N flow).</td>
</tr>
<tr>
<td>B844</td>
<td>17/2/14</td>
<td>Guam – Guam</td>
<td>SSE from Guam to fly under convective band (to 4°N) with low level runs (&lt; 1000 m in BL). GV and GH flying nearby. Layers of elevated O₃ and NOₓ at ~6000m (westerly flow).</td>
</tr>
<tr>
<td>B845</td>
<td>17/2/14</td>
<td>Guam – Guam</td>
<td>S from Guam to be West of convective band (to 6°N). Low level legs (&lt; 1000 m in BL) at Southern end. Layers of elevated O₃ and NOₓ at ~6000 m (westerly flow).</td>
</tr>
<tr>
<td>B846</td>
<td>18/2/14</td>
<td>Guam - Palau</td>
<td>Start in FT (~6500 m), then low nearer Palau (1500 m); head to 4°N, 137°E before heading NW; same region as GV and GH. O₃ 30 ppb in FT, 12 ppb in BL, very stable. Easterly flow.</td>
</tr>
<tr>
<td>B847</td>
<td>18/2/14</td>
<td>Palau – Kota Kinabalu</td>
<td>Steady ascent toward KK. Some Asian outflow observed on initial ascent (CO ~ 140 ppb). Westerly flow.</td>
</tr>
</tbody>
</table>
Figure Caption List

Figure 1: Examples of trajectory-based forecast products used for multi-aircraft flight planning. These plots are for February 13 2014 when all three aircraft were in the same region (see Figure 7 in Pan et al. (2016)). The three panels show the location of air parcels which had been below 1 km altitude in the preceding 12 days at (a) 16-18 km; (b) 14-16 km; and (c) 12-14 km. The number in each box is the percentage of parcels in that box from below 1 km in the preceding 12 days. During the campaign, they were available as 1, 3 and 5 day forecasts for flight planning, and the NAME model was driven by analyses and forecasts from Met Office operational model run at 25 km horizontal resolution.

Figure 2: Map of FAAM BAe-146 flight tracks during January and February 2014. The flights tracks are coloured by altitude. The islands of Guam, Palau and Chuuk are marked. The background shows Jan-Feb averaged Chlorophyll-a concentrations, measured by the MODIS satellite (NASA Giovanni: Acker et al., 2007). The inset shows an enlarged area around Chuuk Atoll.

Figure 3: Ozone and carbon monoxide mixing ratios measured in all CAST flights as a function of latitude and altitude (left). The means and associated 2 standard deviations of ozone and carbon monoxide are shown as a function of altitude (right). See text and Table 1 for instrumental details.

Figure 4: Plots of O₃ against CO coloured by (upper) NO and (lower) relative humidity (10 s averaged data).

Figure 5: CHBr₃ mixing ratios (colours) sampled on the FAAM aircraft during CAST using the whole air sampler (squares) and the on-board GC-MS (circles). Panel (a) contains all measurements made at altitudes less than 1 km, and the enlarged inset (bottom left) shows the values around the Chuuk Atoll. The lines associated with each measurement in the inset indicate the instantaneous wind speed measured by the aircraft. Panel (b) contains the measurements at altitudes greater than 1 km, and the inset shows the vertical profile of all measurements.

Figure 6: Surface observations of wind, O₃, CO₂, CH₄, C₂Cl₄, CHBr₃ and CH₃I at the ARM Facility on Manus Island, Papua New Guinea (2.07°S, 147.4°E) from February 1-24,
2014. The time shown in the x-axis is Universal Time. The shading indicates the local time, with the darker bands representing night-time.

Figure 7: Daily observed ozone profile in Manus (left) and corresponding MACC forecast with a lead time of 1 day (middle) and 4 days (right).

Figure 8: AIITS scattering patterns recorded from ice particles in the UTLS, at altitudes of ~ 16 km and temperatures of ~ -80°C. The pictures are indicative of (left) a smooth quasi-spherical ice particle, (middle) a columnar crystal, and (right) a pristine hexagonal plate.

Figure 9: Flight path of the NASA Global Hawk on 10th March 2015 (blue). OCO-2 (green) and GOSAT (red) soundings are shown which coincide temporally with the flight leg between 25°N, 127°W and 18°N, 125°W. MODIS cloud fraction data (Platnick et al., 2015) coincident with the OCO-2 overpass at 2140 UTC is plotted in grayscale, showing the largely cloud-free conditions encountered during this leg of the flight.
Figure 7. Examples of trajectory-based forecast products used for multi-aircraft flight planning. These plots are for February 13 2014 when all three aircraft were in the same region (see Figure 7 in Pan et al. (2016)). The three panels show the location of air parcels which had been below 1 km altitude in the preceding 12 days at (a) 16-18 km; (b) 14-16 km; and (c) 12-14 km. The number in each box is the percentage of parcels in that box from below 1 km in the preceding 12 days. During the campaign, they were available as 1, 3 and 5 day forecasts for flight planning, and the NAME model was driven by analyses and forecasts from Met Office operational model run at 25 km horizontal resolution. NB Only a fraction of the trajectories are shown in each plot, so the density of dots is not comparable at different altitudes.
Figure 2: Map of FAAM BAe-146 flight tracks during January and February 2014. The flights tracks are coloured by altitude. The islands of Guam, Palau and Chuuk are marked. The background shows Jan-Feb averaged Chlorophyll-a concentrations, measured by the MODIS satellite (NASA Giovanni, Acker et al., 2007). The inset shows an enlarged area around Chuuk Atoll.
Figure 3: Ozone and carbon monoxide mixing ratios measured in all CAST flights as a function of latitude and altitude (left). The means and associated 2 standard deviations of ozone and carbon monoxide are shown as a function of altitude (right). See text and Table 1 for instrumental details.
Figure 4: Plots of O₃ against CO coloured by (upper) NO and (lower) relative humidity (10 s averaged data).
Figure 5: CHBr₃ mixing ratios (colours) sampled on the FAAM aircraft during CAST using the whole air sampler (squares) and the on-board GC-MS (circles). Panel (a) contains all measurements made at altitudes less than 1 km, and the enlarged inset (bottom left) shows the values around the Chuuk Atoll. The lines associated with each measurement in the inset indicate the instantaneous wind speed measured by the aircraft. Panel (b) contains the measurements at altitudes greater than 1 km, and the inset shows the vertical profile of all measurements.
Figure 6: Surface observations of wind, O$_3$, CO$_2$, CH$_4$, C$_2$Cl$_4$, CHBr$_3$ and CH$_3$I at the ARM Facility on Manus Island, Papua New Guinea (2.07°S, 147.4°E) from February 1-24, 2014. The time shown in the x-axis is Universal Time. The shading indicates the local time, with the darker bands representing night-time.
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