Collaborative Research:

CONvective TRansport of Active Species in the Tropics (CONTRAST)

Experimental Design Overview (EDO) Document

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Executive Summary

We propose here the CONvective TRansport of Active Species in the Tropics (CONTRAST) experiment to be conducted from the island of Guam (13.5N, 145E) during January-February, 2014. The main scientific objective of the project is to measure the chemistry and transport of reactive chemical species into the tropical Tropopause Transition Layer (TTL) over the Western Pacific warm pool region. During the boreal winter season, tropospheric air masses are preferentially transported into the lower stratosphere in this region. Thus, the sources, chemistry, and transport of trace gases and their degradation products in the region can substantially impact the chemistry of the lower stratosphere. Considerable attention is being given to the role of tropical convection on the delivery of reactive gases to the TTL. For example, uncertainties in the abundance of reactive halogen species and the fate of short lived organic halogen compounds, particularly bromocarbons, in the tropical upper troposphere leads to significant uncertainties in the photochemistry of stratospheric ozone, especially in the lowermost stratosphere. Despite the recognized importance of this region to shaping the chemical composition of the tropical upper troposphere and lower stratosphere, relatively few chemical measurements have been made to date, partially because of the lack of suitable airborne platforms and instruments to reach the required altitudes. This limitation is no longer a factor with the deployment of the NSF GV aircraft and the development of suitable instrumentation for that platform.

Detailed in-situ measurements of a suite of gases and particles as well as meteorological parameters from the GV during CONTRAST will elucidate the roles of active convection and long-range transport on the chemical composition of the tropical atmosphere, and the altitude variation of these processes. The western Pacific region is also found, from ozonesonde measurements or remote sensing platforms, to have extremely low ozone concentrations. CONTRAST will provide detailed in-situ measurements of a suite of chemicals to characterize the region's chemical environment. The unique low-ozone environments in the upper troposphere are expected to result in very small levels of OH radicals, which can be expected to increase the lifetime of reactive gases whose main loss is through OH radical oxidation. The simultaneous occurrence of deep convection and prolonged lifetime of organic compounds in the TTL can have significant impact on the stratospheric halogen budget. Because the chemical and radiative properties of this region are a key factor in the mechanisms that link climate forcing and atmospheric composition, the CONTRAST experiment will provide measurements necessary for diagnosing and constraining chemistry-climate models.

The proposed payload and operational range of the GV during CONTRAST will allow significant progress to be made in characterizing the chemistry of the tropical upper troposphere, including the altitude region of main convective outflow (12-14 km). Moreover, the timing of the CONTRAST experiment has been designed to take advantage of collaborations with two other airborne studies planned for the same time and geographic location. These are the NASA EV1 project ATTREX (Airborne Tropical Tropopause Experiment) and the European CAST (Coordinated Airborne Studies in the Tropics). With complementary instrument payloads, coordinated flights of the GV, the NASA Global Hawk and the UK Bae146 will provide an unprecedented examination of the full atmospheric column, from surface to >19 km, in the Tropical Western Pacific.

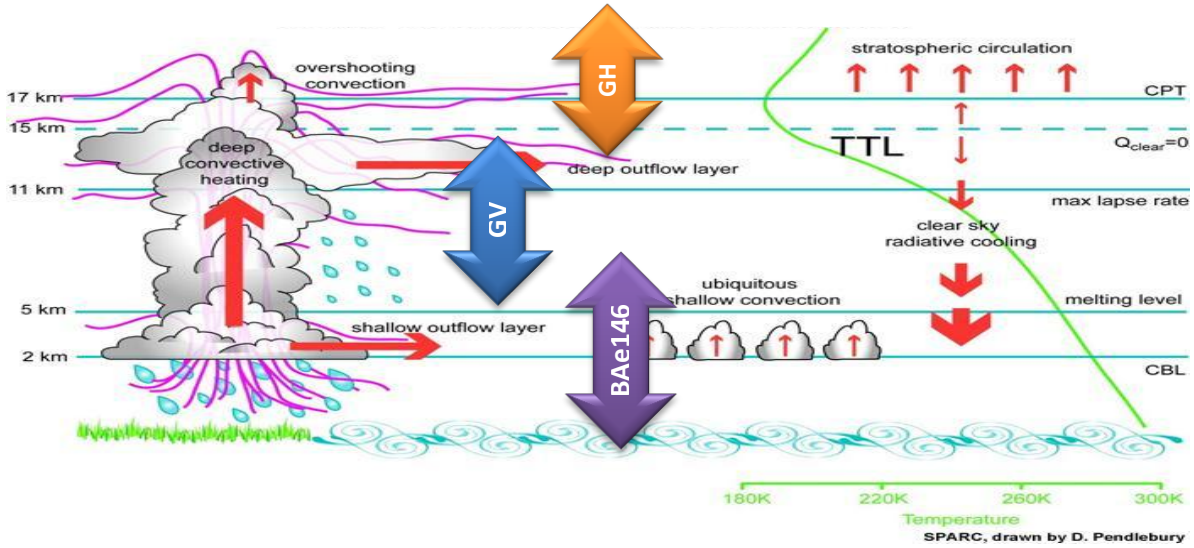
Program Rationale and Objectives

The upper troposphere and lower stratosphere (UTLS) is important for chemistry-climate coupling largely because of the significant radiative forcing caused by ozone, water vapor, clouds and aerosols in this region of the atmosphere [*Shepherd*, 2008] The tropical tropopause layer (TTL) is particularly important because it is the primary gateway to stratosphere [*Fueglistaler et al.*, 2009]. The vast majority of tropospheric air that enters the stratosphere originates from deep convection in the tropical western Pacific that detrains in the lower altitude portion of the TTL [*Bergman et al.*, 2012]. The overall transport of emissions from the boundary-layer to the stratosphere involves rapid transport by deep convection from the surface to the bottom of the TTL (time scales of minutes to hours), followed by slow ascent through the TTL (time scales of months). Despite its importance, the composition of air in the UTLS over the tropical western Pacific is not well characterized.

The convective transport process that rapidly redistributes chemical compounds between surface emission sources and the UT is especially important for the budget of short-lived chemicals, many of which play a significant role in the ozone production and loss processes in the troposphere and stratosphere. The contribution of convective transport to the chemistry of the upper troposphere is a high priority research area because of potential impacts of on chemistry/climate coupling [e.g., *Shepherd*, 2008].

We propose a field project, CONvective TRansport of Active Species in the Tropics (CONTRAST), using the NSF/NCAR research aircraft GV to investigate the role of convective transport in TTL chemistry and the vertical distribution of the short-lived species in the western Pacific region. The proposed flights will be based from Guam (13.48ºN, 144.8ºE) and will occur during January-February 2014. The timing is designed for coordination of the GV flights with flights of the Global Hawk at higher altitudes as part of the NASA Airborne Tropical TRopopause Experiment (ATTREX) program and of the BAe-146 at lower altitudes as part of the NERC Coordinated Airborne Studies in the Tropics (CAST) program. Combined measurements from the three aircraft would span the surface to the lowermost stratosphere, providing an unprecedented opportunity to assess the role of convection on atmospheric composition in the tropical western Pacific (Figure 1). CONTRAST has unique science goals and will also conduct extensive sampling of the TTL during January-February 2014, providing an important connection between the highest altitudes sampled by CAST (top of free troposphere) and the lowest altitudes probed extensively by ATTREX (top portion of the TTL and lower stratosphere).

Figure 1. Schematic showing the convective processes in relation to the TTL, and typical altitude regions sampled by the CONTRAST GV, the ATTREX Global Hawk (GH), and CAST BAe-146 aircraft.



The scientific objectives of CONTRAST are to:

- Characterize the chemical composition at the level of convective outflow over the Western Pacific during the deep convective season
- Evaluate the budget of organic and inorganic bromine and iodine in the TTL
- Investigate transport pathways from the oceanic surface to the tropopause using the GV coordinated flights with BAe-146 and Global Hawk

The western Pacific during Boreal winter season is characterized by intense deep convection. This region makes the dominant contribution for the transport across the TTL into the stratosphere [e.g., *Fueglistaler et al.*, 2004; *Levine et al.*, 2007]. A recent study using trajectory analyses combined with convective influence calculations indicates that during Boreal winter most of the air reaching the tropical stratosphere originally detrained from deep convection in the western Pacific region [*Bergman et al.,* 2012]. Nonetheless, key questions persist:

- What is the impact of deep convection on the photochemical budget of O_3 in the UT and on the supply of halogens to the LS?
- What is the relative importance of typical deep convection detraining at 12 to 13 km versus extreme convection detraining above 15 km for the composition of air entering the stratosphere?
- How important is mixing from the extra-tropical stratosphere for TTL composition over the western Pacific?

Persistent deep convection creates a unique chemical environment of extremely low ozone (O_3) in the middle and upper troposphere of the western Pacific, though the rates of ozone loss and production in these layers has not been quantified. The absence of O_3 creates a distinct photochemical condition that potentially extends the lifetime of many very short-lived (VSL) halogen species [*Kley et al.*, 1996]. In addition, the low O_3 environment alters the partitioning of inorganic halogen species that have formed following the decomposition of these source species. Characterizing the composition of the western Pacific TTL region is critical for understanding the transport of halogens into the stratosphere. CONTRAST will focus on characterizing the photochemical environment of the convective outflow region and determining the abundance of organic and inorganic bromine and iodine species in the TTL, which is subject to considerable uncertainty (e.g., Section 1.3 of WMO, 2011).

The chemistry involving VSL halocarbons and the details of convective transport have been identified as a significant area of investigation for the process-oriented Chemistry-Climate Model (CCM) evaluation project, a joint project of the International Global Atmospheric Chemistry (IGAC) Project and the WCRP's (World Climate Research Programme) SPARC (Stratospheric Processes and their Role in Climate). As convective parameterization has been a significant challenge in climate modeling, accurately representing the impact of deep convection on chemistry is a significant challenge for global chemistry-climate models.

CONTRAST is designed to provide a comprehensive suite of measurements needed to characterize the effect of deep convection on the chemical environment of the tropical western Pacific TTL, which is a pre-requisite for testing and ultimately improving chemical models of the tropics. In addition to measurements of ozone, ozone precursors, and a variety of trace gases, CONTRAST will quantify the amount of bromine injected into the TTL by deep convection, the fate of VSL bromocarbons as air is lofted through the TTL, and the reason(s) for different behavior of bromine versus iodine bearing halogens. Importantly, CONTRAST will provide, for the first time, measurement of a complete set of bromine and iodine gases (inorganic and organic) needed to address the most prominent uncertainty described above.

Scientific Hypotheses of CONTRAST

To address the main objectives of CONTRAST a series of hypotheses and questions has been developed. These are summarized below, but they are more fully described in the companion Science Project Overview (SPO) document.

Objective 1. Characterize the influence of deep convection on the chemical composition and the photochemical budget of O_3 at the level of convective outflow over the western Pacific

Hypothesis: The photochemical budget of O_3 in the tropical TTL is determined by the strength of inputs of chemical precursors from convection and lightning. The simultaneous measurement of O_3, H_2O, CO, CH_4, and radiation will allow for model estimates of atomic O as well as OH and HO_2 (HO_x). Of course, volatile organic compounds (VOC) could supply HO_x to this region (Wennberg et al., 1998). Acetone (C_3H_6O), perhaps the most important VOC source of HO_x, will be measured both inside and outside regions of deep convective influence. Comparison of measured and modeled H_2CO will further help to evaluate the consistency of our photochemical calculations. We will calculate OH and HO_2 profiles, based on constraints for CONTRAST measurements, for air parcels *inside* and *outside* regions of deep convective influence. Measurements of NO, NO_2, BrO, and IO provide direct constraints on the abundance of radicals that participate in a series of O_3 production and loss reactions. Although we will not have direct measurements of ClO, the abundance of total inorganic chlorine (Cl_y) will be inferred from measurement of the nighttime reservoir of Br_y species (see below). Finally, the western TTL is a region of strong lightning occurrence and potential source of NO_x. Comparison of NO_x vs CO and other tracers of combustion will provide an empirical measure of the impact of lightning on NO_x. The CONTRAST measurements will be used to construct estimates of the photochemical loss terms of O_3 by $O+O_3$, HO_x, NO_x, and halogens as a function of altitude, for air parcels both *inside* and *outside* regions of deep convective influence, in order to test model representations of the photochemical production and loss of O_3, such as that shown in Figure 5 [from *Saiz-Lopez et al.*, 2011] and discussed in *von Glasow et al.* [2004].

Hypothesis: The low O_3 environment of air undergoing recent, deep convection will increase the atmospheric lifetime of halocarbons lost by reaction with OH. As noted above, the simultaneous measurement of O_3, H_2O and radiation will allow for model estimates of OH. Of course, non-methane hydrocarbons could supply OH to this region [*Wennberg et al.*, 1998]. A likely result of the low O_3 environment associated with recent deep convection is decreased values of calculated OH. CONTRAST measurements of a range of organic trace gases, including halocarbons such as $CHBr_3$, CH_2Br_2, and bromochloromethane (CH_2BrCl), will allow the effect of OH, on the lifetime of these gases, to be assessed.

Objective 2. Evaluate the budget of organic and inorganic bromine and iodine in the TTL

A suite of organic halocarbons are emitted from the tropical ocean by biological processes in near surface waters. Convection transports these gases to the upper troposphere, where they decompose. Past measurements in other parts of the tropics have provided a limited survey of the abundance of organic halocarbons, particularly bromine bearing compounds, at the top of the TTL (e.g., Figure 3, Auxiliary Material, *Salawitch et al.*, 2010). These data clearly establish that the major bromine bearing organic molecules to cross the tropopause in the tropics are CH_2Br_2 and CH_2BrCl, due to their long tropospheric lifetimes. All other bromocarbons are mostly lost in the TTL. Whether this distribution is also characteristic of the Western Pacific region, with significant surface emissions and potential for deeper convective penetration into the TTL, will be examined during CONTRAST. As noted above, there is

tremendous uncertainty regarding the fate of the organic products produced upon photochemical decomposition of CBr_y and CI_y species.

Hypothesis: CH_2Br_2, $CHBr_3$, and other VSL bromocarbons will be elevated in air parcels that have undergone recent deep convection. Model predictions (e.g., Figure 4 in SPO) suggest that air influenced by recent deep convection will have higher abundances of CH_2Br_2 and $CHBr_3$ than air in nearby air parcels. CONTRAST measurements will allow a quantitative estimate of the convective influence on VSL bromocarbons. Contrasting profiles will provide an important test for models such as CAM-Chem that was used to generate Figure 4.

Hypothesis: When CBr_y and CI_y species decompose, the resulting inorganic species remain as labile, gas phase species. The test of this hypothesis will be similar to many past studies that have examined atmospheric halogen budgets. A key aspect enabling this hypothesis to be tested, *for the first time during CONTRAST*, is the capability for aircraft measurement of concentrations of atomic Br and atomic I, in addition to BrO and IO and other major inorganic halogen species. The low O_3 environment of the tropical UT drives the partitioning of Br_y species towards atomic Br. During daytime, this photochemical model calculation indicates the vast majority of inorganic bromine should be present as Br. This species converts to either HOBr or BrCl at night, depending on ambient levels of inorganic chlorine, with a brief spike of $BrNO_3$ occurring just after sunset (See SPO Figures 6 + 7).

The CONTRAST flights will include *both daytime and nighttime flights* that, coupled with measurement capability for HOBr and BrCl, will provide definitive quantification of the partitioning of inorganic bromine species. The CONTRAST data analysis will benefit greatly from the collaborative aircraft measurements. The CAST will provide data defining the range of expected values for total Br and total I because, near the surface, the vast majority of the molecular mass should be present as organic molecules. The ATTREX flights will provide a connection to stratospheric input: as the GH approaches the lower stratosphere, O_3 levels rise, and the titration switches back to BrO, which instruments on the GH will measure.

Objective 3. Investigate transport pathways from the oceanic surface to the tropopause using coordinated flights with CAST BAe-146 and ATTREX GH

Limited understanding of both the efficiency of deep convective transport and the rates of transport through the TTL contribute to uncertainties in our ability to predict the composition of air entering the stratosphere. Within the TTL, photochemical reactions and competing physical processes are important for chemical species whose lifetime is comparable to the ~2 months that it takes for slow ascent to traverse the TTL. In-mixing of air from midlatitudes will increase the age of the air in the TTL and could reduce the input of halogens to the stratosphere . Such in-mixing can also reduce TTL relative humidity and cloud formation [*Fujiwara et al.*, 2009].

Transport of boundary layer air into the TTL is poorly understood, both in terms of the height distribution of convective detrainment in the TTL and in terms of dilution of convective cores by entrainment throughout the free troposphere. Mid-oceanic convergent zones have convective tops that rarely exceed 13.5 km, while a significant percentage of western Pacific boreal winter convection (~3%) reaches the tropopause. Entrainment of free tropospheric air into deep convection updrafts is important, particularly for the relatively small convective cores that occur in the tropical maritime regions. Thus, the composition of air deposited in the TTL at the tops of convection will be a combination of boundary-layer composition and free tropospheric composition.

Science Goal: **Quantify the relative importance of the following three pathways for trace gas transport from surface to the stratosphere, based on observations in the tropical TTL:**

1) deep convective injection directly into the stratosphere [*Danielsen, 1982; Dessler and Sherwood,* 2003];

2) convection detrainment into the TTL followed by a slow ascent into the stratosphere [*Holton and Gettelman*, 2001]; and

3) the effect of intrusion from midlatitude lower stratosphere into the tropical TTL, also known as "in-mixing" [*Waugh and Polvani*, 2000; *Konopka et al.*, 2010].

These pathways are expected to have distinct chemical signatures that can be diagnosed with tracer/tracer correlation analysis [e.g., *Marcy et al.,* 2004; *Pan et al.,* 2004,2007*; Ridley et al.,* 2000*; Avery et al.,* 2011] that will take advantage of measurements from all three aircraft. A combination of tracers with different sources and lifetimes are expected to provide the necessary diagnostics to evaluate different transport pathways. Mixing timescales can also be attached to the different transport processes. We have already noted the use of short-lived iodine species as a significant indicator of rapid convective transport to the UT. Mixing time scales of convective transport can be inferred from the measured distribution of methyl iodide. Other species with marine origins, such as methyl nitrate and various bromocarbons, provide additional constraints on the transport timescales. Profiles of these species through the TTL should allow us to evaluate the relative importance of detrainment at different altitudes of convection. Similarly, measurements of CO_2 have been shown to provide a useful "clock" for diagnosing transport rates in the TTL [*Park et al.,* 2007; 2010]. These rates have also been estimated from gradients in hydrofluorocarbon and hydrofluorocarbon mixing ratios. Calculated transport rates are then extremely valuable to assess loss rates and mechanisms for VSLS in the TTL region.

Collaborative Project Science Goals

The rationale for coordinating the GV CONTRAST mission with ATTREX and CAST is related to common scientific goals between the projects. Given common scientific objectives, it is not surprising that the instrument payloads are also complementary. To provide some perspective on the collaborative aspects of the proposed missions, we briefly discuss objectives of CAST and ATTREX next.

From its inception, the CAST mission included the collaboration with ATTREX in order to relate the CAST measurements in lower to mid-troposphere to the higher altitude conditions that could be accessible to the GH. Thus, the major objectives of the CAST project were identified to provide an improved and more quantitative understanding of: 1) transport processes in the Tropical Tropopause Layer (TTL); and 2) VSL compounds in the tropical atmosphere, including their fluxes from the ocean, their atmospheric chemistry, and their transport up into the TTL.

The CAST measurements will allow a more complete characterization of chemical properties in the marine boundary layer and lower free troposphere than would be possible with just GV measurements, and the GV will extend the chemical characterization into the upper troposphere to the lower TTL and at levels of the main convective outflow which are not routinely accessible by the GH. Thus we are able to obtain a more complete picture of the inflow and outflow of Western Pacific convection.

ATTREX focuses on the upper troposphere and lower stratosphere, but will benefit from measurements in the lower TTL and in the main convective outflow region which is below the normal operating range of the Global Hawk. The combined measurements from the Global Hawk and the GV will provide excellent definition of chemical gradients and transport through the altitude range of the TTL. The broad goals of the ATTREX mission are to improve understanding of how deep convection, slow large-scale ascent, waves, and cloud microphysics control the humidity and chemical composition of air entering the

stratosphere, and to improve global-model predictions of feedbacks associated with future changes in TTL cirrus, stratospheric humidity, and stratospheric ozone in a changing climate.

To meet these broad goals, a range of specific questions were identified in the ATTREX plan. These include questions that relate to the control of stratospheric humidity and temperature structure in the TTL. Of more direct relationship to the CONTRAST objectives are questions regarding TTL radiation and transport and TTL chemistry. For example some of the specific questions addressed by ATTREX in these areas that are closely aligned with CONTRAST (as discussed above) are:

- What is the relative importance of typical convection detraining at ~13 km versus extreme convection detraining above 15 km for the humidity and composition of air entering the stratosphere?
- What are the mechanisms for transport from the midlatitudes, and how important are they?
- What is the vertical distribution of BrO and short lived halogen compounds in the TTL and how does it vary seasonally and geographically?
- Are TTL O_3 and halogen observations consistent with photochemical models?

Experimental Design and Observational Requirements

LOCATION AND TIME PERIOD.

The experiment will be based in Guam (13.5N, 144.8E) and will take place during the winter of 2014. The location was specifically chosen to be out of the main convective region. Based on examination of long term meteorological conditions at Guam, we have determined that local conditions are favorable for flight operations. Importantly, the location is well within the range of the tropical warm pool convection to the south and the subtropical jet to the north (See Fig 2). Access to both of these areas is necessary to address important scientific objectives of the mission.

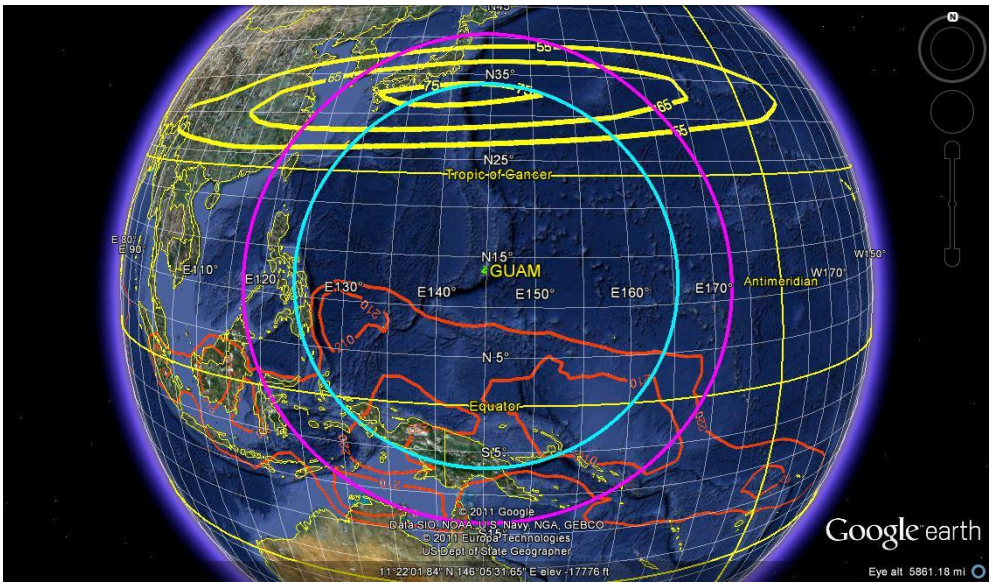


Figure 2. Range of NCAR GV in the CONTRAST study area (cyan (3hr) and purple (4hr)) from base in Guam; Also shown is contour of OLR (Red) to demonstrate good access to region of major tropical convection and windspeed (yellow) to indicate access to region of the subtropical jet and mixing gradients between tropics and extratropical air.

The timing of the mission is based on the significant convective activity that occurs over the tropical warm pool during the boreal winter. It is important to get into this region during the winter to be able to characterize the transport and chemistry associated with the deep convection that occurs in the region, and

to provide a contrast to the nearby tropical upper troposphere. The other obvious important factor in the timing is the ability to coordinate flights with scientific collaborators in the ATTREX and CAST missions. The combined capabilities of the different aircraft and their complementary instrument payloads provide a remarkable synergy to characterize convective transport and chemical/radiative impacts in this region.

Several potential complications to the mission timing and location can be identified. One potential complication to the mission is the Madden Julian Oscillation. As part of this oscillation, which is most pronounced in the Eastern Hemisphere during Northern Hemisphere winter, the outgoing long wave radiation, and hence the deep convective clouds, undergo a 30-60 day oscillation (*Madden and Julian*, 1994). There is a chance, therefore, that convection would be suppressed during a significant portion of a mission. However, analysis already done by the ATTREX team has shown that, because of the long range of the GV (and the Global Hawk), this is not a major issue for CONTRAST.

Another potential deployment issue is ENSO, which causes shifts in the pattern of convection. This is only a significant issue if there is a strong El Nino event and a large eastward shift of the region of strong convection (during La Nina or "normal" years the strong convection is within the GV range from Guam). Fortunately, these are predictable some time in advance, and a new locale may need to be considered. CONTRAST will carefully coordinate any attempt to move the locale of operation with the coordinated ATTREX and CAST projects.

OBSERVATIONAL REQUIREMENTS

To meet the objectives of the CONTRAST mission requires the altitude capabilities and long-range flight operations of the GV aircraft and an instrument payload of both in-situ and remote sensing measurement capabilities. Planned measurements from the GV (Table 1) include a range of gases and reactive species necessary to address CONTRAST science goals. The relationship of the proposed measurements to the specific project objectives is also indicated in Table 1.

The main objectives cover three main scientific areas, with overlapping measurement requirements in most cases. The broad objectives can be defined as: 1) Chemical characterization of the main convective outflow, and definition of the photochemical environment; 2) Budget and partitioning of halogen compounds in the TTL region; and 3) Evaluation of transport pathways to and through the TTL in the Western Pacific atmosphere during boreal winter.

Measurement of trace gases with different source emissions, lifetimes (including VSL's), and temporal trends will be used to evaluate transport time scales and mixing processes (Objectives 1 and 3). These measurements will be done with a combination of whole air sampling, in-situ GC/MS, and high resolution instrumentation for CO, CO_2, and CH_4. GC/MS and whole air sampling provide complementary measurements of trace gases. TOGA can measure a number of significant oxygenated VOC that are not well-behaved in whole air canisters. The AWAS can measure a full suite of NMHC, halocarbons, organic nitrates, etc. but with lower spatial resolution. A number of trace gases can be measured from both systems to provide good overlap and comparison. Measurements of radiation and relevant reactive gases will define the photochemical environment of the tropical UT to examine the impact of convective inputs of low ozone and elevated marine emissions to the UT. These measurements include actinic flux, ozone, formaldehyde, NO and NO_2, and halogen radical species (Objectives 1 and 2). The NO_x measurements also provide information on the input of lightning-produced nitric oxide in this area of deep convection. The budget and partitioning of bromine in the TTL will be evaluated with measurements of the organic bromine precursors and inorganic bromine species (Objective 2). The organic bromine precursors will be measured from whole air sampler and GC/MS. The inorganic halogen species will be measured by in-situ instruments (chemical ionization mass spectrometry (CIMS) and resonance enhanced fluorescence

(ROFLEX)) as well as remote sensing (MAXDOAS). The MAXDOAS will provide a link to comparable measurements on the GH and BAe-146 aircraft. Contrast of daytime and nighttime partitioning of inorganic bromine species will test the proposed mechanisms of halogen partitioning that are relevant to the low ozone environment in this region. Finally, aerosol measurements will identify cloud aerosols and aerosol size distributions that may be significant for heterogeneous chemical processing. Complementary measurements from ATTREX and CAST are indicated in Table 1, with detailed information provided in Appendix 1.

TABLE 1. Observational Requirements and Potential Instruments for CONTRAST. Comparable instrumentation on the GH (= Global Hawk) and BAe (= BAe-146) aircraft is indicated. Relationship to scientific objectives: 1=chemical characterization, O3 budget; 2=halogen budget; 3= transport and mixing.

Observation	Requirement	Instrument Source & Status	Objective	GH	BAe
O3	1 ppbv; 10 s	Facility (Fast O3)	1,2,3	Yes	Yes
H2O Vapor	1 – 1000 ppmv; 1 s	Facility (VCSEL)	1,2	Yes	Yes
CO	5%; 10 s	ACD (VUV)	1,3	Yes	Yes
CH4	5 ppbv; 10 s	ACD (Picarro)	1,3	Yes	Yes
CO2	0.3 ppmv; 10 s	ACD (Picarro)	1,3	Yes	Yes
H2CO	25 pptv; 30 s	CU (Laser DFG)	1,2,3	No	No
NO, NO2	5 pptv; 10 s	ACD (Chemiluminescence)	1,2	No	Yes
BrO, HOBr, Br2 (in situ)	2 pptv; 10 s	Facility (CIMS)	1,2	No	Yes
BrO, IO, H2CO (remote)	2/1/100 pptv; 10 s	CU-AMAX (DOAS)	1,2	Yes	No
Br, I	2 pptv; <1 min	CIAC (Spain) (ROFLEX)	2	No	No
NMHC, including short lived tracers, HCFCs, halocarbons	Various	Facility (AWAS)	1,3	Yes	Yes
Oxygenated VOC, VOC	Various ; 2-4 min.	Facility (TOGA)	1,3	No	No
Aerosol (number, size, distribution)	Various	Facility (USHAS)	1,2	No	No
Cloud detection		Facility (CDP, 2D-C)	1,2	Remote	No
Microwave Temperature Profiler	2 K 6 km above / below aircraft	Facility (MTP)	3	Yes	No
Radiation (UV/VIS)		Facility (HARP)	1,2	Yes	Yes

Payload Risks and Uncertainties.

With one exception, all of the instruments identified for the CONTRAST mission have been tested and deployed on the GV in other missions prior to CONTRAST. Thus, there is an experienced and capable payload available for the mission. The single exception is the atomic-halogen resonance fluorescence instrument (ROFLEX). The instrument has been used for ground-based deployments, even in rugged and remote locations [*Mahajan et al.,* 2011; *Gomez-Martin et al.*, 2011]. Plans have already begun to design the instrument for airborne operation. Given the successful use of the instrument in ground-based

studies, we are confident that the transition to a research-grade airborne instrument can be accomplished prior to the deployment in 2014. We recognize the risk in this projection, but think the unique reward is worth it. Even without the ROFLEX, significant advances could be made in evaluating the halogen budget and partitioning with the other instruments available on the aircraft (MAXDOAS, CIMS, AWAS, TOGA), but the measurement of atomic halogens from ROFLEX most directly and uniquely provides the information necessary to test some of the photochemical model predictions.

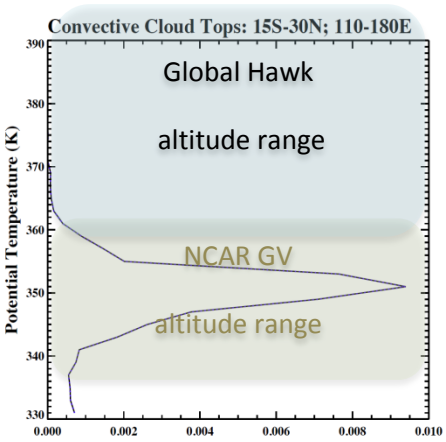
Another potential risk factor relating to the payload is the need for on-site analysis of whole air samples collected from the GV. The currently available complement of sample modules is 18 (and up to 24 if 6 additional are made available by NOAA). Nominally, 5 modules per flight (60 samples) will be collected, which means that canisters need to be recycled to allow for 12 or more flights in Guam. (We note that the CAST payload has similar requirements for on-site analysis). Fortunately, a field analytical instrument for whole air samples is under development for use with whole air samples collected on the GH for ATTREX (Atlas-PI). There will be enough capacity to accommodate the GV samples as well as the GH samples. Though it requires some additional personnel in the field, on-site analysis offers many advantages, including more rapid availability of the data and more convenient and immediate comparisons with others measuring trace gases on the different platforms. An alternate approach to on-site analysis is to transfer samples from the AWAS modules to other canisters for later analysis back in the laboratory. This approach was used successfully during the START08 mission, but it is not a first choice. A second alternative, if logistics prohibit on-site analysis, would be to consider the use of the NOAA flask sampler that was deployed during the recent HIPPO missions, if sufficient capacity is available for the mission requirements.

FLIGHT PLANS AND RATIONALE

We anticipate approximately 12 flights of ~ 8 hours each based out of Guam over 6 weeks in the field, plus science ferry flights that will provide measurements in the central and eastern Pacific atmosphere (Table 2 and Figure 4). Many of the flights will be coordinated with the collaborating missions (ATTREX and CAST) to achieve individual and common scientific objectives. The flight duration and altitude ranges of the aircraft involved in the mission are highly complementary for research of tropical convection. The NCAR GV has access to the main altitude of convective outflow altitude centered around 350 K potential temperature (12 – 14 km altitude) (see Figure 3) and can define conditions in the lower altitudes of the TTL. Furthermore, the GV has the altitude range to reach the lower stratosphere at higher latitudes. The upper altitude limit of the GV (about 14.5 km) leaves off where the lower altitude level of the Global Hawk picks up (about 14 km), with an eventual altitude ceiling near 19 km. With these two aircraft, we would be able to measure a detailed profile of chemical constituents throughout the TTL and into the lower stratosphere. The UK BAe-146 aircraft has a more limited duration and lower altitude ceiling (about 9 km). This aircraft has a payload designed to characterize chemical emissions and inputs within the marine boundary layer and the lower to mid free troposphere. (See Appendix 1 for details of the CAST and ATTREX payloads). Current plans for CAST include multiple short flights/day as well as island hops to get the aircraft deep into the intertropical convergence zone inflow region.

As illustrated in Figure 4 and summarized in Table 2, we anticipate several basic flight plans to meet CONTRAST objectives. Of course, meteorological conditions will dictate the details of each flight plan, but we envision five basic sampling scenarios. Sampling during transit flights (#5) from Colorado, through Hawaii, and to Guam will allow a good characterization of the longitudinal variations in the chemistry of the background tropical and subtropical upper troposphere. If the transit flights use Hawaii (e.g., Kona) as a stop, we would request a flight south from Hawaii (#1) to characterize the TTL in the Central Pacific region, which should have conditions considerably different from those that will be encountered in the Western Pacific (and also from those in the Eastern Pacific which will be studied

during the TORERO mission). Furthermore, these flights would nicely complement the extensive HIPPO measurements recently completed. The CONTRAST flights would help characterize the upper tropospheric region of the Central Pacific that could not be sampled in detail during the recent HIPPO campaign. Flights to the north of Guam (#2) to the region of the Subtropical Jet Stream will be used to examine chemical gradients and potential mixing pathways between the TTL region and the lowermost stratosphere. The remaining bulk of the flights will be to the south of Guam to probe different convective regions and vertical structures within the TTL. Flights that will profile the TTL will be targeted for



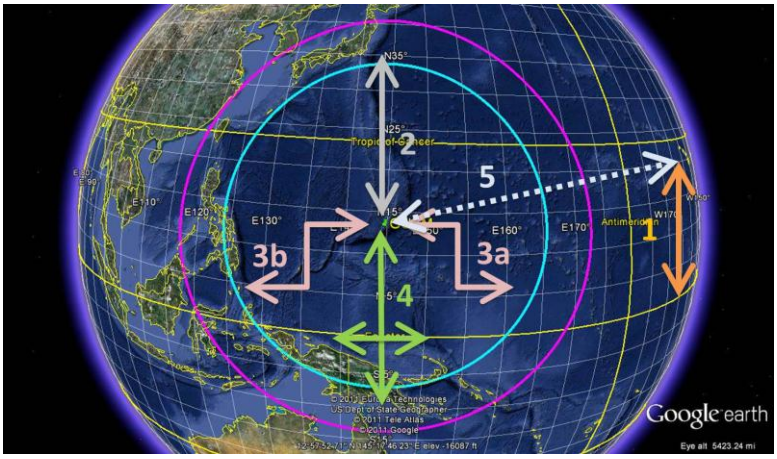


Figure 3. (left) Plot of convective cloud top frequency vs. potential temperature for the CONTRAST study region for DJF season based on TRMM data. Overlay of the approximate altitude ranges of the GV and the GH shows that the GV will be able to sample in the main convective outflow (lower TTL) and the GH will cover higher altitudes.

Figure 4. (right) Schematic flight tracks for GV during CONTRAST. Track numbers and descriptions are in Table 2. Range rings are indicated in cyan (3hr) and purple (4 hr).

TABLE 2. Nominal flight types and descriptions for CONTRAST GV.

Number of flights	Description	Flight type	Coordination with other aircraft
2	Profiles of central Pacific troposphere and lower stratosphere to be done flying south from Hawaii during transits to/from Guam. Characterize upper troposphere in mid Pacific region.	1	None anticipated
2	Sample across the subtropical jet to obtain composition information in the lower stratosphere; define latitudinal gradients; isentropic mixing	2	GH
3	Profile tropopause cold pool region, including night flights for halogen partitioning; locate and characterize chemistry of low ozone features.	3a,b	GH + BAe
2	Inflow/Outflow sampling from recent/older convection. Samples background condition and residual impact of past convection; evaluate longitudinal gradient and long-range transport.	3a,b	GH + BAe
3	Deep convection. Sample entrainment and detrainment from a convective system. Evaluate tracer relationships and effect on vertical distributions in the TTL.	4	GH + BAe
4	Transit flights (Broomfield-Hawaii/Hawaii-Guam)	5	None anticipated

conditions that include recent convection with outflow near 350K (#3a,b; 4), areas outside of recent convective activity (#3a,b), and regions of the TTL impacted by deep convection (>350K) (#4). Though we do not expect to probe active convective cells, we are interested in penetrating the anvil downstream of convection to compare with adjacent clear air chemical conditions.

Flights will be designed to study the vertical structure and relationships of chemical tracers and dynamical background in the TTL. For example, it is known that the tropical ozone profile has a characteristic "S" shape. Low ozone at ~14 km is an indication of convective outflow [*Folkins et al.*, 1999] that contains surface layer air depleted in ozone. Using a multi-year record of ozonesonde data from Fiji, we can see this behavior for the DJF time period (Figure 5). Also shown in Figure 5 are the ozone profiles and.

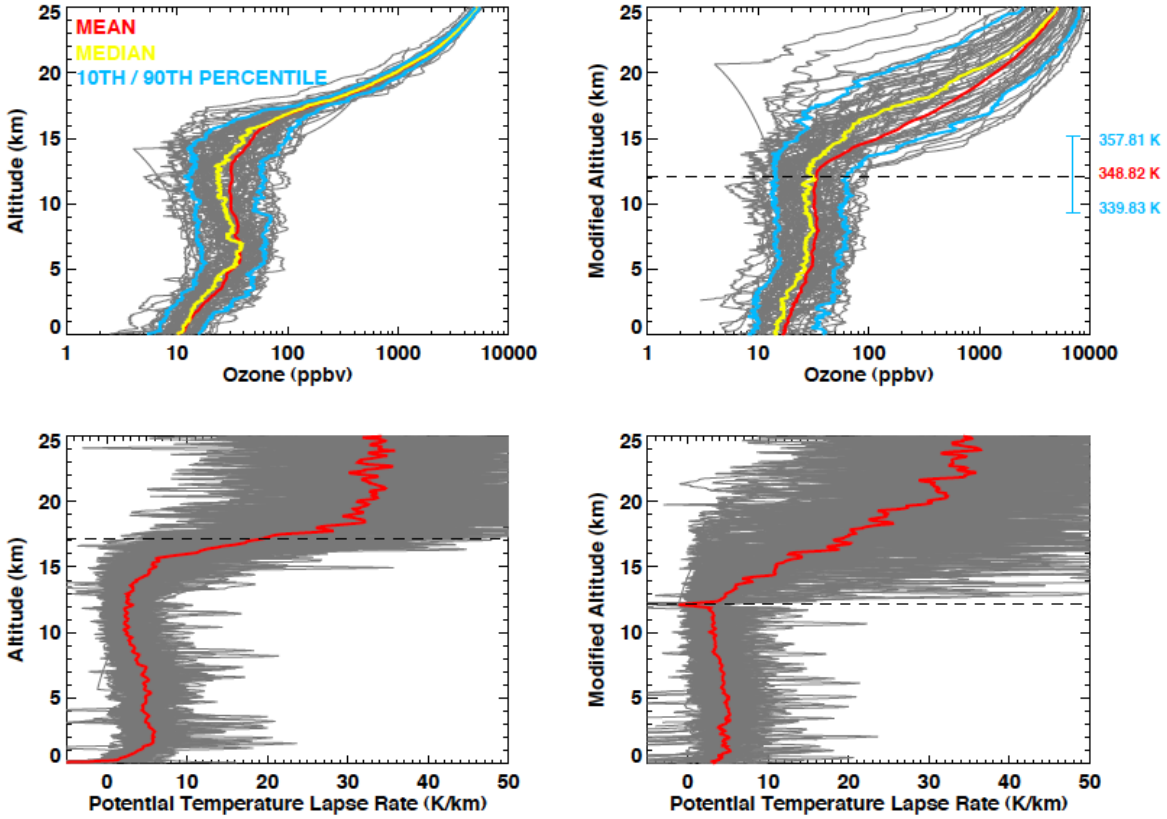


Figure 5. Multi-year record of ozonesonde profiles from Fiji vs altitude (Top-left) and in modified altitude, i.e., altitude relative to the level of minimum static stability (also referred to as the lapse rate minimum (LRM)) calculated from temperature profiles and adjusted to the mean level of minimum stability (dashed line) (top-right). Red=mean; Yellow= median; Blue =- 10th and 90th percentile. The range of variability in level of LRM is marked by the blue vertical bar on the dashed line (top-right). The potential temperature lapse rate profiles associated with the ozonesondes are shown in the bottom panels in the same vertical coordinates.

associated lapse rate profiles in altitude relative to the level of lapse rate minimum (LRM), which is also referred to as the level of minimum static stability. The sharp increase of both ozone and stability at the level of LRM shows that LRM is a critical level of convective influence and serves as the lower boundary of the TTL [*Gettelman and Foster*, 2002]. In Figure 6, we show a calculation, using 4 years of COSMIC GPS temperature profile data, of the distribution of LRM in CONTRAST region with the GV range. The average LRM height shows the contrast in the level of convective influence between the southern and northern part of the CONTRAST domain. The LRM is above 12 km in the southern domain, with a higher frequency of occurrence above the average. We plan to investigate the behavior of

chemical tracers of different lifetimes in relation to the TTL boundary and contrast the behavior between the southern and the northern domain. In both sides of the domain, the typical LMR is within the GV altitude capability. This relationship will provide a good diagnostic for the signature of convective influence in the chemical species of interest. Utilizing this relationship, a sampling strategy will be designed to obtain complete vertical profiles through the troposphere using coordinated flights. The level of convective influence will be characterized using both temperature and ozone profiles, and additional detailed chemical measurements at this level will provide totally new information for the transport into TTL.

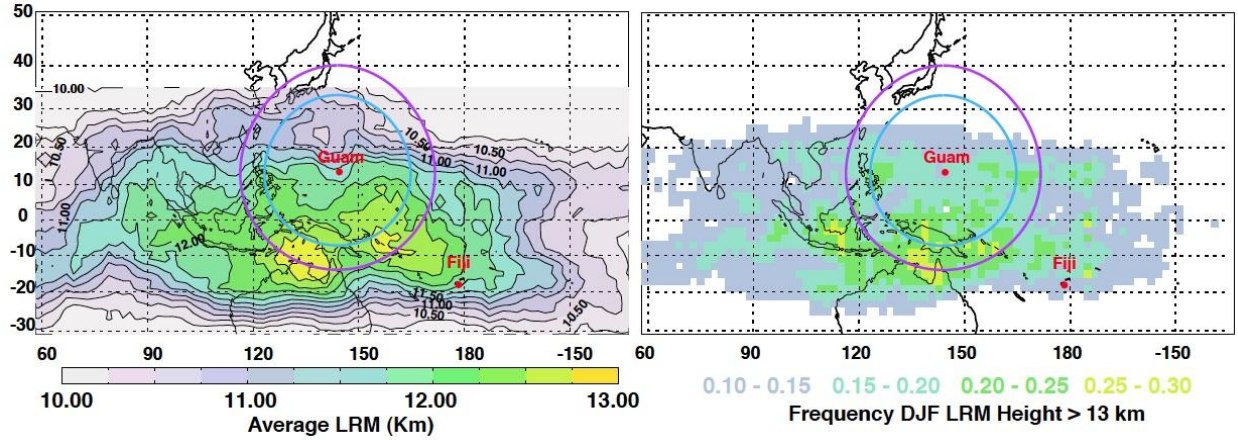


Figure 6. (left) Seasonal average of LRM based on four-year COSMIC GPS satellite temperature profiles. Calculation is done in 2x2 degree Lat/Lon bins. The highest average shown is 13 km. (Right) Frequency of occurrence of LRM greater than 13 km. The GV range is shown by the blue and purple rings.

Flight Coordination with Global Hawk and BAe-146

As noted, the complementary objectives, capabilities, and instrument payloads of CONTRAST, CAST, and ATTREX present a unique opportunity for understanding the convective atmosphere of the Western Tropical Pacific during a period of active and deep convection. Conceptual flight plans of the GH for ATTREX are shown in Figure 7, with an overlay of the GV operational range. The overlap region presents excellent opportunities for meeting common scientific objectives (compare Figure 4). However, detailed logistics and planning of a multi-aircraft operation also presents challenges. Fortunately, Principal Investigators of both CONTRAST and ATTREX will have experience in the planning and conduct of such multi-platform missions (e.g., SEAC4RS). In addition, mission meteorologists will work closely and will be cognizant of the different capabilities and objectives of the different aircraft as they develop forecasts for flight planning.

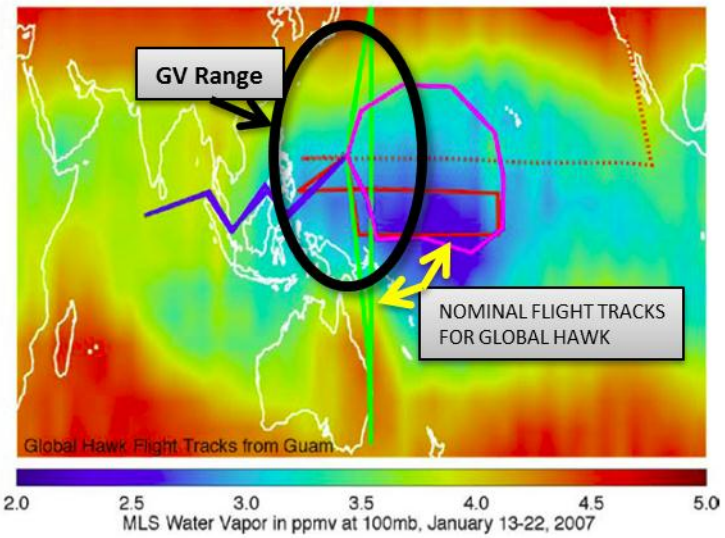


Figure 7. Nominal flight tracks of the ATTREX Global Hawk aircraft for missions from Guam. The flights are designed to get long transects and detailed information in and over the TTL region over the Tropical Warm Pool. The blue color represents the low water vapor conditions characteristic of cold tropopause in the region. The operational range of the GV indicates excellent opportunities for coordinated flights (see also Figure 4).

The day-to-day details of flight coordination will need to be worked out during the mission. Clearly one factor that will need to be considered is how to optimize the flight coordination of the long duration Global Hawk flights (typically 24 – 28 hours), with the normal flight durations of the GV (about 8 – 10 hours) and the BAe-146 (about 5 - 6 hours). With these constraints, we expect several periods of back-to-back flights of the GV to be able to interact with the GH on outbound and return legs. Similarly, the CAST plan calls for occasional days with multiple flights per day, and for island hopping ("suitcase") flights to extend the regional coverage.

Project Management, including Management in the Field

Project management of CONTRAST is outlined in Figure 8 below. The Mission Scientists (Atlas, Pan, and Salawitch) will be responsible for meeting the overall scientific objectives of the project. They will coordinate closely with EOL Project Management to communicate scientific objectives and plans and will work with EOL to decide how to best meet mission objectives within the normal constraints of safe aircraft operations.

The CONTRAST science team is composed of all principal investigators in measurement, theory, and modeling, and forecasting activities, and includes the appropriate NSF program manager(s). The science team is responsible for design of the experimental investigation, modeling and analysis of data, and publication and presentation of results.

Mission planning in the field will be coordinated by L. Pan (NCAR). In consultation with the co-principal investigators and the CONTRAST Science team, she will work with CONTRAST mission meteorologist and forecaster (J. Bresch), with collaborating mission forecasters (L. Pfister and colleagues (ATTREX); N. Harris and colleagues (CAST)), and with GV pilots and project manager to define individual mission flight tracks. Dr. Pan has led flight planning in the past (START08) and will play a similar role in the upcoming SEAC4RS mission (which is also a coordinated multi-aircraft campaign). This experience will be very relevant to the CONTRAST mission planning. Mission scientists Pan, Atlas, and Salawitch will rotate as Flight Scientists on board the GV during the CONTRAST study. Atlas will be the coordinator for the GV measurements, and will work with counterparts in ATTREX and CAST to develop and implement appropriate activities for measurement comparisons between instrument payloads.

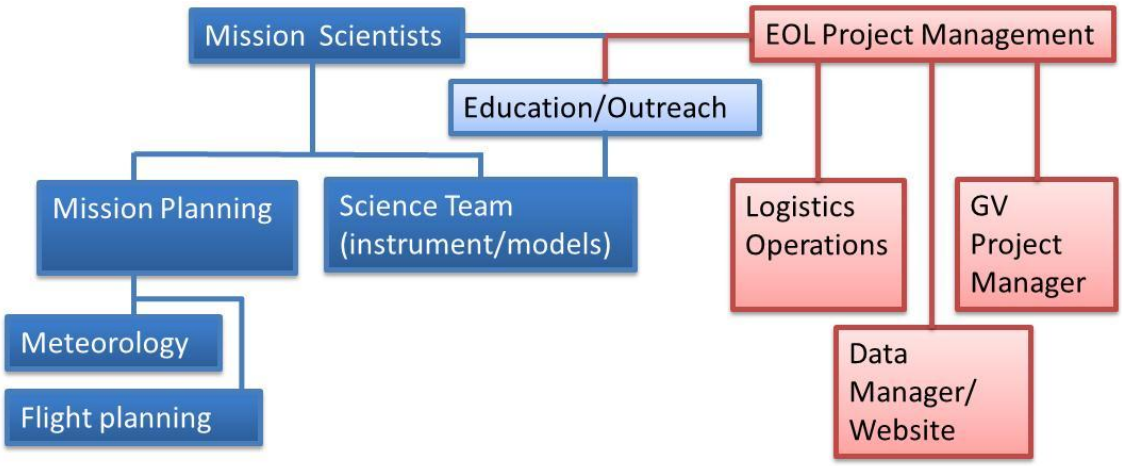


Figure 8. CONTRAST Project management structure.

Flight operations for CONTRAST, ATTREX, and CAST will be co-located in Guam, which will help in the organization and planning of joint operations. Mission meteorologists will provide daily briefings where opportunities for different flight scenarios will be discussed.

The education and outreach activities of CONTRAST will be coordinated by the Mission Scientists with appropriate personnel from EOL. The education and outreach activities of the recent HIPPO missions will serve as a model for CONTRAST. These activities included a Facebook page with educational clips and interviews, instrument descriptions, scientific background, and images from the mission. Visits to the aircraft by students and teachers will be part of the education and outreach activity. In addition, individual CONTRAST investigators supported by NSF may also propose education and outreach activities from their home institution.

Data Management Plan

Data management plans for atmospheric chemistry (and other) missions of the NSF GV aircraft have been developed during several studies that have taken place in the last few years (e.g., START08, HIPPO, etc.), and we plan to follow this successful model of data management for CONTRAST. The basic data management plan includes use of a web-based Field Catalog, and a separate data exchange website.
Flight planning discussions and supporting meteorological, satellite and modeling products will be made available on a CONTRAST project Field Catalog that will be setup in collaboration with NCAR/EOL/CDS (Computing Data Services). Project scientists will work with CDS programmers and staff to establish the necessary files and information to be made available during and after the mission. Typically a Field Catalog will archive daily meteorological forecasts and discussions, operational satellite products for the study area, mission instrument status, flight reports, etc. These reports and products are necessary and valuable during the mission, and they serve as complete documentation for post-mission analysis. Similar field catalogs have been used with great success in past campaigns.

A separate website will be established for flight instrument data exchange. This will be a password protected site where first-look data from the aircraft and from the instrumentation on board the GV will be located. We will request data from each instrument be submitted to the first-look archive within 24 hours after each flight. This data is necessary to identify potential instrument problems, to evaluate the success of meeting mission objectives, and to help plan subsequent missions. A data protocol will be established with collaborators on ATTREX and CAST to share the data in the field and also after the mission. We already have agreement in principle for data sharing with our ATTREX and CAST partners, and a detailed data exchange/sharing protocol will be established and agreed upon by each Science Team. After the mission is complete, the data exchange site will be used as a location to place final data after appropriate quality control checks. One year after the mission is complete, the data will migrate to a publicly available website that is managed by EOL (e.g., data.eol.ucar.edu/codiac).

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I. Table of Participants, Activities, and Estimated Costs

	Institution	Function	Cost Estimate
GV Facility and Aircraft Support	NCAR	Deployment pool costs for aircraft and associated activities	$1,950 K
EOL/CDS/FPS	NCAR	Data management, field catalog, field planning logistics, education/outreach activities	$450K
Elliot Atlas^	University of Miami	Co-Principal Investigator + Whole Air Sampler (HAIS AWAS)	Uncertain ($500K)
Ross Salawitch	University of Maryland	Co-Principal Investigator + Modeling/Theory Investigations	Uncertain ($500K)
Laura Pan	NCAR/ACD	Co-Principal Investigator + Flight Planning/Modeling+ Data Management	ACD
Alan Fried	University of Colorado	Formaldehyde measurement	Uncertain ($640K)
Alfonso Saiz-Lopez	Lab. for Atmos. and Climate Science; Toledo, Spain	Br/I measurement: ROFLEX	Supported by PI funds
Dan Riemer^	University of Miami	VOC measurement: HAIS TOGA	Uncertain ($380K)
Greg Huey	Georgia Institute of Technology	In-situ: BrO, BrCl, HOBr/Br2: HAIS CIMS	Uncertain ($500K)
Mark Zondlo^	Princeton University	Water Vapor: HAIS VCSEL	Uncertain ($400K)
Rainer Volkhamer	University of Colorado	**BrO +… MAXDOAS:** GV measurement	Uncertain ($500K)
Eric Apel	NCAR/ACD	VOC measurement: HAIS TOGA	Included
Frank Flocke	NCAR/ACD	CH4/CO2 measurement: PICARRO CRD	ACD
Teresa Campos	NCAR/ACD	CO measurement: VUV	Included
Sam Hall	NCAR/ACD	HARP: GV Radiation measurements	Included
Andy Weinheimer	NCAR/ACD	NO/NO2/O3: GV measurement	ACD
Facility support	NCAR	Aerosols and Cloud instrumentation	included
Julie Haggerty	NCAR/RAF	Microwave Temperature Profiler	Included
John Bergman	NCAR/ACD	Modeling/Meteorology	ACD
Bill Randel	NCAR/ACD	Modeling/Satellite	ACD
Doug Kinnison	NCAR/ACD	Modeling/WACCM	ACD
Jean-Francois Lamarque	NCAR/ACD	Modeling: Chemistry Climate	ACD
	Project	Additional Shipping PI support	$40K
	Project	Contingency	$200K
Lennie Pfister	NASA	ATTREX Principal Investigator + meteorology	No cost
Eric Jensen	NASA	ATTREX Principal Investigator	No cost
Neil Harris	University of Cambridge	CAST Principal Investigator	No cost
		TOTAL ESTIMATE	
		Deployment + Special Costs	$2,400 K
		Investigator Costs*	$3,420 K
		Additional	$ 240 K

*If not estimated by individual, assumes $500K total for 3 year project.

^Affiliated with requested HAIS instruments (AWAS, TOGA, VCSEL); PI's to be involved in mission planning, deployment, and post mission science. Partial TOGA support included in RAF estimate. No support for WAS or VCSEL included in RAF cost estimate.

APPENDIX 1.

Instrument payloads on BAe-146 (CAST) and NASA Global Hawk (ATTREX).

CAST – Bae-146 aircraft

Parameter	Instrument	Performance	Institution
Ozone	TE49C	1 minute integration time, 1ppb detection limit (dl)	FAAM
Water vapour	General Eastern 1011 & Buck CR2		FAAM
Carbon Monoxide	Aerolaser 5002	1 minute integration time, 2 ppb dl	FAAM
Nitrogen oxides	Air Quality Designs	1 Hz, dl is 10 pptv for NO and 20 pptv for NO_2	FAAM + York
VSL Halocarbons: $CHBr_3$, CH_2Br_2, $CHBr_2Cl$, CH_3I, CH_2BrCl, $CHBrCl_2$, C_2H_5I, CH_2ICl, CH_2IBr, CH_2I_2, CH_2Cl_2, $CHCl_3$	In situ Agilent GC-MS with Markes dual TD	3-4 min sampling *in situ*, 2-3 min via WAS bottles < 0.01-0.05 ppt dl.	York
Whole Air Samples NMHC (C_1-C_6), small OVOCs, DMS	Perkin Elmer GC-FID (WAS bottles, ~2-3 min sampling)	2-3 min sampling. 2.5, 1 pptv dl for C_2-C_4 and >C_4 respectively	York
CO_2, CH_4	Los Gatos	5 sec integration precision ±σ CH_4, 1.0 ppb; CO_2, 200 ppb. Max rate 10 Hz.	FAAM + Manchester
N_2O, H_2O	Aerodyne QCLAS	N_2O precision @ 1 Hz ±1σ, 0.2 ppbv. Max sampling rate 20 Hz.	Manchester
BrO	CIMS	2.6 pptv ± 3σ @ 4 s integration	Manchester
Black Carbon	SP2	Black carbon mass size distribution, 1 Hz	Manchester

ATTREX – NASA Global Hawk

Acronym	Weight (lb)	Power (W)	Measurement	Sampling Rate	Precision	Accuracy
CPL	366		Aerosol/Cloud Backscatter	1 Hz	10-15% backscatter	15-25% extinction
O_3	40	200	O_3	2 Hz	1.5 x 10^{10} molecules cm^{-3}	5% + precision
AWAS	200	300	~60 tracers with lifetimes of 1 week to years	80 samples per flight	Various, typically 1-10%	Various, typically 2-20%
UCATS	60	250 (450)[a]	O_3	10 s	> 1 ppb or 2%	> 2 ppb or 3%
			H_2O	1 s	2-3%	3-5%
			CH_4	140 s	0.4-0.8%	1%
			N_2O	70 s	0.2-0.5%	1%
			CO	140 s	2-5%	1%
			H_2	140 s	2-3%	1%
			CFC-11*	70 s	0.3-0.6%	1%
			CFC-12*	70 s	0.3-0.6%	1%
			Halon-1211*	70 s	0.5-0.8%	1%
			SF_6	70 s	0.2-0.5%	1%
PCRS	45	370	CO_2	5 s	200 ppbv	150 ppbv
			CO	5 min	3 ppbv	15 ppbv
			CH_4	5 s	2 ppbv	1 ppbv
ULH	24	260	H_2O vapor	1-40 Hz	> 0.05 ppmv or 1%	10%
DLH	50	280	H_2O vapor	100 Hz	1% or 50 ppbv	10%
Hawkeye	135	3200	Ice crystal size distributions, habits	1 Hz	20%	50%
SSFR	40		Radiative Fluxes	20 Hz	0.1%	3%
MMS	65	135	Temperature	20 Hz	0.01 K	0.3 K
			Pressure	20 Hz	0.1 mbar	0.3 mbar
			Horizontal wind	20 Hz	0.01 m/s	1 m/s
			Vertical wind	20 Hz	0.01 m/s	0.1 m/s
MTP	24	51	Temperature Profile	1 prof/15 s	<1 K	<0.05 K
Mini-DOAS	33	28	BrO	50 s	0.9 pptv	8%
			O_3		80 ppbv	2%
			NO_2		20 pptv	5%
			OClO		4.5 pptv	12%
			IO		0.4 pptv	25%
			OIO		0.4 pptv	55%

**Requires replacing H_2-CO-CH_4 channel*