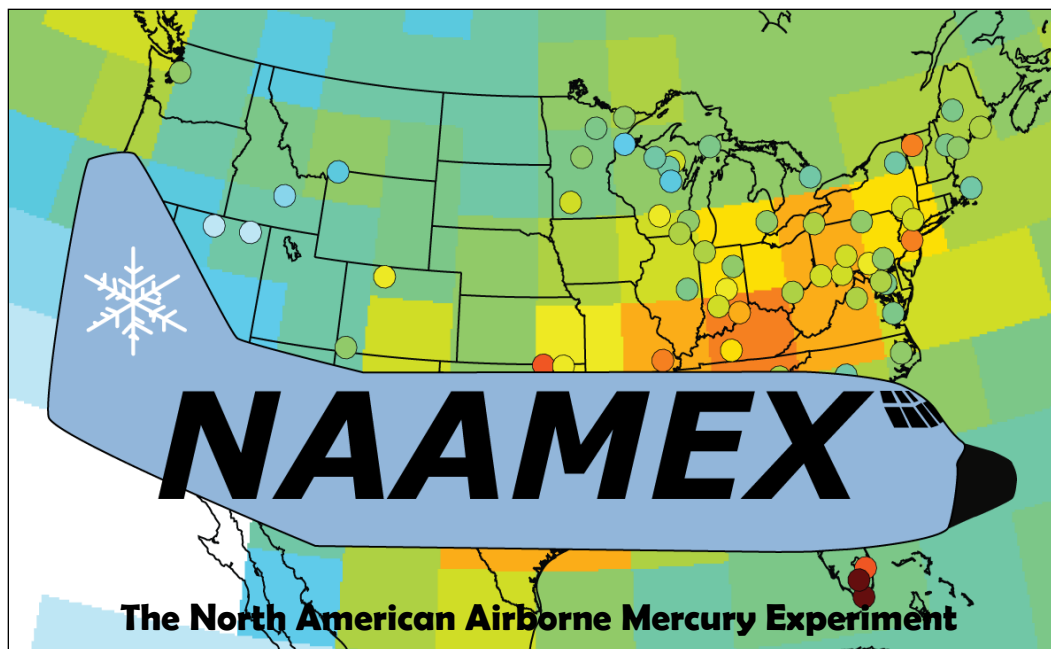


REQUEST FOR LAOF FACILITY SUPPORT
NORTH AMERICAN AIRBORNE MERCURY EXPERIMENT (NAAMEX)
FOR CONSIDERATION AT SPRING 2012 OFAP MEETING
Submitted Dec 1, 2011



Part I: General Information

A. Corresponding Principal Investigator(s)

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Additional C-130 investigators	Institution
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Chris Cantrell/Lee Mauldin	University of Colorado
Greg Huey/David Tanner	Georgia Institute of Technology
Andrew Weinheimer	NCAR
Teresa Campos	NCAR

B. Project Description

Project Title	North American Airborne Mercury Experiment (NAAMEX)
Location of Project	Dual bases: Western US and Eastern US
Start and End Dates of Field Deployment Phase	June 15, 2013- July 31, 2013
NSF Facilities requested	C-130 Aircraft
Funding Agency and Program Officer Name(s)	NSF, Alex Pszenny
Proposal affiliated with this request	Collaborative Research: North American Airborne Mercury Experiment (NAAMEX)
Proposal Status	Submission will occur in Dec 2011.
Do you expect other, non-NSF support? If yes, from whom?	Contributions in-kind from scientists in Canada and Europe on modeling and data analysis.
Is this a resubmission of a previous request?	Yes (originally submitted in Dec 2009)
Is this a multi-year request or continuation?	One time request for C-130

C. Abstract of Proposed Project (*one-page summary of NSF proposal*)

Intellectual merit

Mercury bioaccumulation is a problem of national significance, however there are large uncertainties in our understanding of the sources and atmospheric cycling of mercury. The North American Airborne Mercury Experiment (NAAMEX) is the first comprehensive airborne experiment focused on Hg in North America and will provide critical new data to understand the sources, chemical processing and deposition of Hg across the U.S. NAAMEX has two scientific goals:

- 1) Constrain emissions of mercury from major source regions in the United States
- 2) Quantify the distribution and chemical transformations of speciated mercury in the free troposphere.

To address these goals we will use the NCAR C-130 aircraft equipped with a suite of state-of-the-art instrumentation focused on Hg and related key compounds, including CO, CO₂, NO_x, O₃, HO_x, halogen species, hydrocarbons and halocarbons. Hg observations on the C-130 will include all of the major atmospheric forms of Hg: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) and particle-bound Hg (PHg). All of the primary instrumentation for NAAMEX has been successfully used previously on the C-130 or other aircraft. The experiment is proposed for the summer of 2013. During NAAMEX the C-130 will fly out of two bases, one in the Western US and one in the Eastern US. Using two bases is a critical component of NAAMEX in that it will allow the experimental team to examine a variety of scientific questions focused on understanding both the North American and global sources of Hg. Flight planning in the field will utilize state-of-the-art Hg forecasting models to optimize flight time. The C-130 observations will be integrated and analyzed after the mission using several chemical transport models that have been optimized to study and understand the global cycling of Hg.

Broader impacts

Human exposure to mercury is a significant health risk and a problem of national and global significance. The work described in this proposal will greatly add to our understanding of the distribution, sources and sinks of Hg. This will reduce the uncertainties concerning the source-receptor relationship and the relative contribution of domestic, foreign and natural sources of Hg in the U.S. NAAMEX will also provide critical new data on the horizontal and vertical distribution of Hg that has never before been possible. These data will yield significant improvements in global models of the Hg cycle; models that are used by regulatory agencies to set emission and exposure limits for Hg. The global nature of this problem is evident by the international participation in NAAMEX. In addition to a core of scientists from the US, NAAMEX will involve a collaboration with scientists from Canada, Italy, Germany and Russia. Thus NAAMEX will further international cooperation to study and understand the global cycle of Hg.

More details of the science objectives for NAAMEX are available in the NAAMEX White Paper available at <http://www.atmos.washington.edu/jaffegroup/modules/NAAMEX/>

D. Experiment Design

Scientific Objectives

NAAMEX is the first ever comprehensive airborne experiment focused on understanding the sources, sinks and chemical transformations of Hg across North America. It is a medium sized experiment, which will utilize the NCAR-C130 and a suite of Hg chemical transport models. The primary objectives for NAAMEX are:

- Goal 1: Constrain emissions of mercury from major source regions in the United States.
 - Specific questions: What is the spatial distribution of atmospheric mercury in the western US, Midwest, and Eastern US? To what extent do sources in the Eastern and Western U.S. contribute to elevated Hg concentrations? Are observations consistent with ground-based observations? Are the observations consistent with bottom up emissions inventories? Can we separate US sources from global sources to establish boundary conditions?
- Goal 2: Quantify the distribution and chemical transformations of speciated mercury in the free troposphere.
 - Specific questions: How does the speciation of mercury change with increasing height in the free troposphere over the western and eastern U.S.? How is this speciation affected by synoptic conditions, atmospheric oxidation, precipitation, cloud-processing, and stratospheric intrusions? Can we quantify the transport of GOM from the free-troposphere into the boundary layer? Can we use the spatial distribution of GOM to provide information on its production mechanism? How does the partitioning of mercury evolve downwind of major emission regions?

Observations of GEM, GOM and PHg downwind of major source regions will provide critical data to quantify both natural and anthropogenic emissions, including the Hg speciation. Flights in specific source regions (e.g. the Ohio River Valley) can help constrain industrial emissions. Curtail flights, downwind of eastern North America, will provide concentration fields and total flux estimates. This will be possible because GEM is known to have a tropospheric lifetime of at least 6 months. In the western US, gold mining in northern Nevada is the largest source category, but the GEM emissions are not known with high accuracy. Flights perpendicular to plumes from the mining region will constrain the emission fluxes using both curtain flights and tracer ratios combined with modeling. A variety of Hg chemical transport models will be employed in NAAMEX and these will be used to derive emissions using, for example, inverse modeling (e.g. *Bocquet and Roustan, 2006; Pan et al., 2007*). Flights to address goal #1 will include flights from bases in the western and eastern U.S. Some extended sampling in the boundary layer is needed in both the western and eastern U.S. Two flights will do “curtain” sampling off the east coast to quantify the total flux of Hg leaving North America.

Goal 2 is focused on furthering our understanding of transformation of Hg between the three chemical forms: GEM, GOM and PHg. Observations of the spatial distribution of these species and potential GEM oxidants (e.g. OH, O₃ and/or Br radicals) in a variety of air mass types will provide new data to test various transformation mechanisms and model predictions.

For example the NAAMEX White Paper shows how the modeled vertical distribution of GOM in the troposphere (2-8 km) varies dramatically depending on the model used and the assumptions made about atmospheric processing. Once we locate regions of high GOM in the free troposphere (e.g. Swartzendruber et al 2006), we can follow this with airborne and surface observations, as well as models, as the GOM mixes into the boundary layer and contributes to surface deposition. This mechanism is thought to be the primary one delivering Hg from the global pool to North American ecosystems, yet it has never been directly observed. Flights to address goal #2 will include flights from both a western and eastern US base. We will want to sample in a variety of air mass types and do slow ascending and descending spirals to and from the maximum altitude.

Relationship to previous work.

Measurements of total gaseous Hg (TGM) or GEM from aircraft have been made by several groups. Figure 1 (from Swartzendruber et al 2009) shows mean vertical profiles from a number of campaigns (Radke et al., 2007; Swartzendruber et al., 2009; Swartzendruber et al., 2008; Banic et al., 2003; Friedli et al., 2004; Talbot et al., 2008; Ebinghaus and Slemr, 2000). While these flight data span many regions including Europe (Ebinghaus and Slemr 2000) and North America (Banic et al 2003), four of these campaigns took place in the North Pacific region (Friedli et al 2004; Swartzendruber et al 2006; 2008; Talbot et al 2008), thus it is somewhat surprising to see such a wide variation in mean concentrations. Unfortunately previous aircraft measurements have been ambiguous regarding the speciation. Some groups report that their data as Total Gaseous Mercury (TGM), while others report GEM for essentially the same inlet configuration. The inlet configuration is important because our recent shows that GOM compounds are readily lost to an unheated inlet (Gustin, Jaffe and Lyman 2011, manuscript in preparation). There are also questions about calibration methods used in some previous aircraft studies (Gustin and Jaffe 2010).

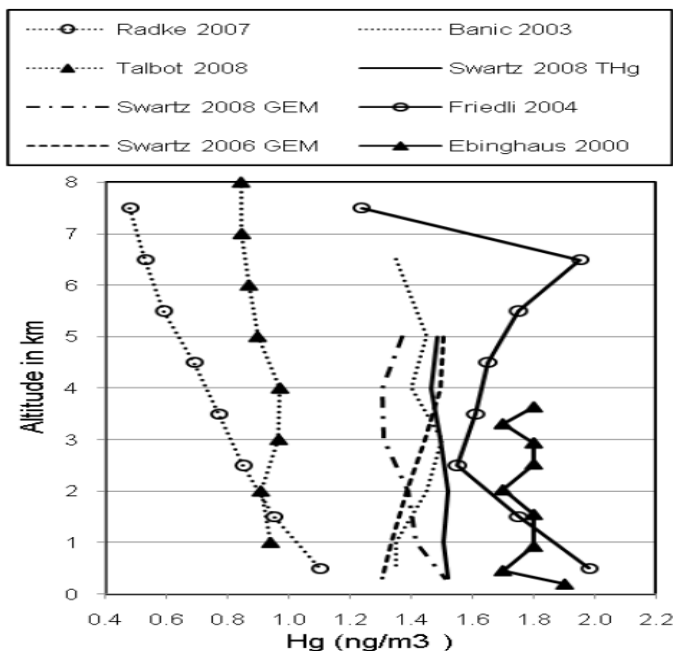


Figure 1. Published vertical profiles for TGM or GEM (from Swartzendruber et al., 2009).

The University of Washington (UW) group has focused on reducing these ambiguities and developing more robust methods for aircraft measurements of Hg and GOM (Swartzendruber et al., 2009; Lyman et al., 2011; Lyman and Jaffe, 2011). In Swartzendruber et al. (2009), two methods were used to determine the vertical distribution of GOM by aircraft: a high time response GOM measurement using the difference between TGM and GEM and a standard, low time resolution method based on collection of GOM on a KCl coated quartz denuder. With both methods the GOM vertical profile peaks in the middle troposphere, similar to the distribution of HO_x radicals (e.g. Cantrell et al., 2003) and reactive bromine compounds (Yang et al., 2005). While the correlation between the two GOM methods was good, there was a factor of 2 difference in concentrations due to problems with the KCl coated quartz denuder method (Swartzendruber et al 2009). The KCl coated quartz denuder method, used by many Hg researchers for more than 10 years, appears to be biased low when exposed to ozone (Lyman et al., 2010). The high time resolution method, based on thermal decomposition, does not have this problem.

The UW aircraft instrument, called the UW-Detector for Oxidized Hg Species (UW-DOhGS), was further improved for the Western Airborne Mercury Observations (WAMO) project, undertaken in 2010 on the NCAR C-130. The UW-DOhGS instrument features many of the elements of the original design, while improving the detection limit, reliability and ability to be calibrated with both GEM and GOM compounds (e.g. HgCl₂ or HgBr₂). The instrument consists of dual atomic fluorescence detectors that measure GEM and TGM. The difference is due to GOM+PHg. For the GEM channel, GOM compounds and PHg are removed using quartz wool. For the TGM channel, GOM compounds are decomposed to GEM using a high temperature furnace. The instrument has been extensively tested in the lab using GEM and GOM compounds as well as using potential interfering compounds. To our knowledge, the UW-DOhGS is the only instrument in the world that has these capabilities and has been tested as extensively. During the WAMO mission the UW group measured GEM and GOM reliably and obtained some exciting results. The most important result was the speciation of Hg in a stratospheric fold that was observed on a flight from Bangor Maine to Boulder Colorado on November 5th, 2010 (Lyman and Jaffe, Nature Geosciences, Available on-line 12/18/2011). The observations showed that GOM is more than half of the total Hg in the stratosphere, but that Hg⁰ is not totally depleted, as one group has reported (Talbot et al 2007).

Table 1 lists gas and aerosol measurements to be made on the C-130 for NAAMEX. Most of the major instrumentation has successfully flown previously on the C-130. The GIT CIMS instrumentation has previously flown on the GV and DC8 aircraft, but not the C-130. We will also deploy a low time resolution denuder/filter pack system for PHg which has not previously flown, but this system requires only a pump, flow controllers and valves to operate. Analysis on these filters will be performed off-line. Species like CO, CO₂, SO₂, NO_x have some of the same combustion sources as Hg and will be useful to identify anthropogenic sources as well as biomass burning sources. We will use Hg/CO and Hg/SO₂ correlations to constrain Hg sources. Reactive species (O₃, HO_x, BrO and Br₂) will provide information on the oxidative environment leading to the production of GOM. NO_x/HNO₃. NMHC ratios will serve as chemical clocks to track the ageing of air masses. Aerosol properties will help constrain the gas/particle partitioning of GOM and PHg, as well as characterize source properties.

Table 1. Key C-130 Measurements during NAAMEX

Aircraft observations:	Detection limit	Time resolution	Who
GEM	0.02 ng/sm ³	2.5 minutes	Jaffe, Ambrose-UW
GOM	0.04 ng/sm ³	2.5 minutes	Jaffe, Ambrose-UW
GOM/PHg	0.01 ng/sm ³	~60 minutes (8/ft)	Jaffe-UW
CO	10 ppbv	1 sec	Campos-NCAR
CO ₂	0.5 ppmv	1 sec	Campos-NCAR
O ₃	3 ppbv	1 sec	Weinheimer-NCAR
NO, NO ₂	5/10 pptv	1 sec	Weinheimer-NCAR
BrO, Br ₂ , ClNO ₂ , HO ₂ NO ₂ , HNO ₃ , SO ₂	1 pptv	1 min	Huey, GIT
OH, HO ₂ , RO ₂ and H ₂ SO ₄	0.01 pptv	1 min	Cantrell, Mauldin-Univ of Colorado
Speciated NMHCs and Halocarbons	5 pptv	1 min	Blake, UCI
Aerosol number/size distribution	NA	10 sec	NCAR
Optical properties (scattering/absorption)	NA	10 sec	NCAR
Meteorology and other standard C-130 parameters	NA	1 sec	NCAR

Since all of the primary user-supplied instruments have previously flown, we include a very brief description of each major system and a few citations. The UW-DOhGS instrument was described above and in the supplemental section of Lyman and Jaffe (2011, available on-line 12/18/2011). The GT group will fly a CIMS instrument with two channels. The first will utilize I- chemistry to detect halogens (e.g. Liao et al., 2011). The second channel will utilize either SF₆- or SF₅- chemistry to measure SO₂, HNO₃, and HO₂NO₂ (e.g. Kim et al., 2004). The UCI group will measure NMHCs, organics and halocarbons in whole air samples and analyzed per Colman et al (2001). Approximately 60 samples will be collected per flight. The Univ. of Colorado group will measure a variety of HO_x and other species using CIMS. For this ambient air is sampled into two inlets, one for OH and H₂SO₄, and one for peroxy radicals. Within the inlets, reagent gases are added to convert the analytes into H₂SO₄ (sulfur-34 labeled for OH measurements). Sulfuric acid is ionized by reaction with negatively charged nitrate ions. The reagent and product ions are transported into a vacuum system where the ions are mass-separated and counted. The ratio of count rates of product to reagent ions is proportional to the ambient concentration of the target species (Mauldin et al 2003; Cantrell et al 2005; Hornbrook et al 2011).

Bases of operations and flight plans

Observations and models suggest that there are substantial differences in the sources, processing and deposition of Hg between the Western and Eastern US. Figure 2 shows the distribution of anthropogenic Hg emissions over North America as determined by the EPA's National Emission Inventory (NEI 2005) and the distribution of biomass burning, land and ocean sources calculated using a high resolution/nested-grid version of the GEOS-Chem chemical transport model (Zhang et al., 2011). Anthropogenic emissions dominate the NE US, especially over the Ohio River Valley. Over the Western U.S. there are fewer point sources, except for gold mining in Nevada and industrial sources in California, and natural geogenic sources become important.

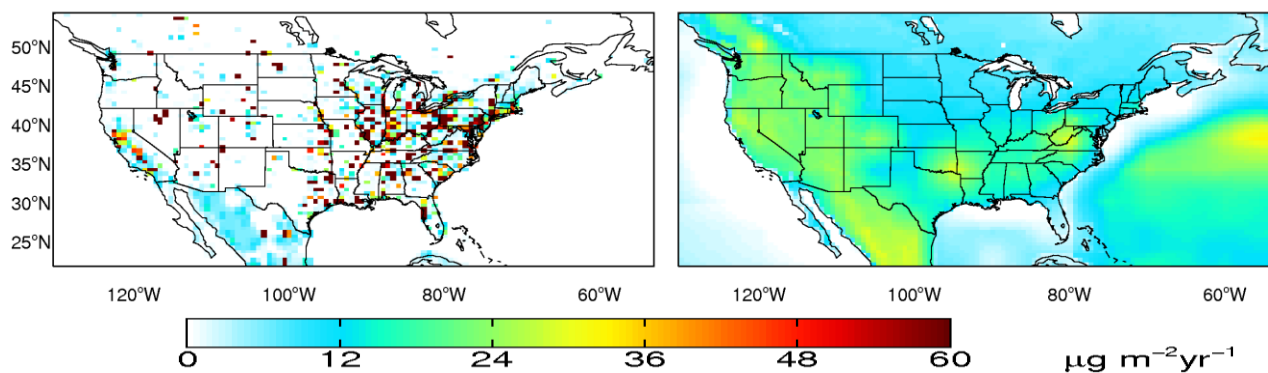


Figure 2. Distribution of mercury emissions from anthropogenic sources (left panel) and from all other sources (sum of biomass burning, soil, and ocean sources) (rightpanel). Anthropogenic sources are from the Environmental Protection Agency's (EPA) National Emission Inventory (NEI) for the year 2005 and the other emissions are from the GEOS-Chem chemical transport model in 2008 (from Zhang et al., 2011).

To intercept plumes from the Eastern U.S. we can use some of the strategies used previously to examine North American outflow. Figure 3 shows modeled North American outflow for CO for July 1998 (Li et al 2005). The outflow flux, calculated using the GEOS-CHEM model, shows a persistent maximum from the northeastern and mid-Atlantic regions, with flow headed towards the North Atlantic which is consistent with observations (e.g. Real et al 2008). Fang et al (2009) discuss the variability in the outflow and focus on the synoptic conditions that give rise to strong outflow of US pollution. Lee et al (2011) use observations from the ICARTT experiment to identify anthropogenic plumes from New York City. Figure 4 shows a model calculation of an outflow event originating in New York City that was verified

with observations using the WP-3D aircraft (Figure 5) These outflow events are similar to those we would expect to intercept during NAAMEX. These flights will provide a wealth of new data on the consistency of U.S. Hg emission inventories with atmospheric observations.

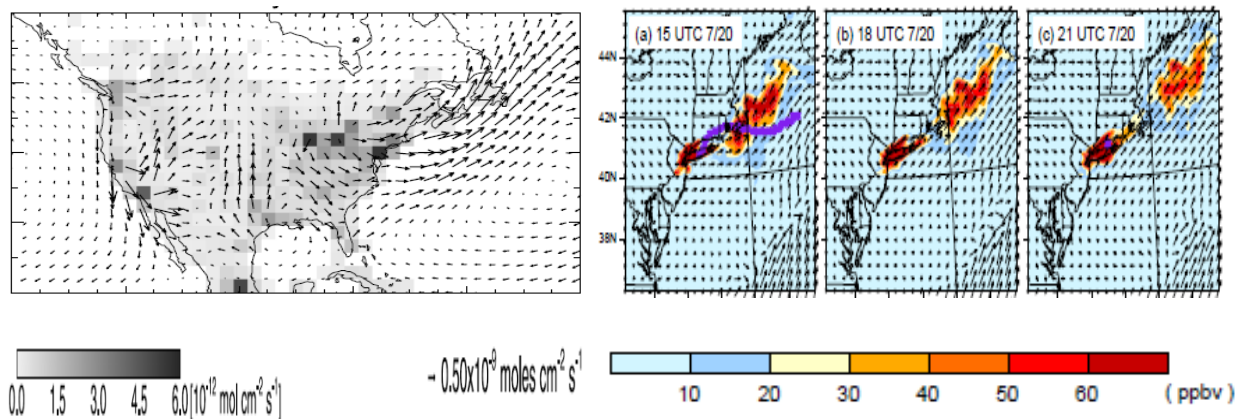


Figure 3 (left). CO emissions (grey background) and North American outflow flux (surface-700 mb, arrows) for July 2000 (Li et al 2005).

Figure 4 (right). Modeled contribution to CO between ~140-600 meters asl from sources in New York City for July 20, 2004 using the WRF-Chem model with a high-resolution nested grid (Lee et al 2011).

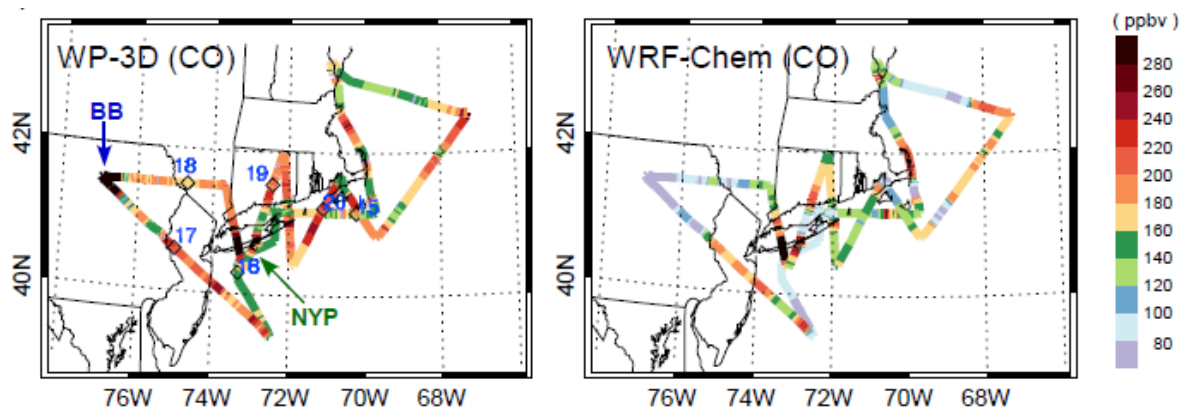


Figure 5. Measured (left panel) and modeled CO (right panel) for July 20, 2004 during the ICARTT campaign (Lee et al 2011). The left panel indicates CO contributions from biomass burning (BB) and the New York plume (NYP).

Flight Schedule

Because the oxidative potential of the atmosphere (and its potential to produce GOM) is greatest during summer, we propose to begin C-130 flights on June 15, 2013. This will increase the probability that we will detect high mixing ratios of GOM.

Alternative Flight Plans/Schedule

NAAMEX could start as late as July 15, 2013 with minimal impact on our scientific objectives. A start later than this would be less than ideal as more frequent storms and reduced photochemistry would reduce the value of the experiment.

Collaboration with other missions

During NAAMEX there is also the possibility for cooperation with other airborne experiments happening at the same time. **While this collaboration is not essential** for NAAMEX, it would contribute to meeting the scientific goals of the experiment. This includes

- 1) The NASA Discover-AQ project, which will be operating aircraft near Houston in the same time frame (summer 2013);
- 2) The European CARABIC program, which has measurements of Hg on commercial flights and may operate between Europe and a US East coast location during NAAMEX;
- 3) The European Global Mercury Observation System (GMOS) which may operate an aircraft over Europe and the North Atlantic during NAAMEX.

While the details of each of these flight programs is not yet final, it would be desirable to coordinate day to day flight operations with one or more of the missions. As NAAMEX and these other missions move forward it will be useful to communicate our plans to each other and develop flight plans that build on the synergy of these multiple missions. A letter of collaboration from Dr. Nicola Pirrone, Director of GMOS is included at the end this facility request.

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Past publications by PI (D.Jaffe) from use of EOL facilities:

The P.I. used the Wyoming King Air in 1999. This project resulted in 7 publications. In addition many of the other NAAMEX PI's have been involved in C-130 experiments and these have resulted in many additional publications. From the WAMO flights in 2010, one paper has already been accepted for *Nature Geosciences* (Lyman and Jaffe 2011) and a second paper is in preparation.

E. Educational Activities

Numerous students (both graduate and undergrad) will be associated with the project from the respective institutions of the various PIs on the project. These students will be exposed to research in atmospheric mercury, and receive training and exposure to large scale aircraft experiments. Because of the policy and health implications there is a great deal of interest in Hg research from the general public and news media. For this reason it is important to carefully plan out a significant outreach component associated with NAAMEX. The outreach will focus on the specific NAAMEX science objectives, how these will help improve national mercury policies and specific health and dietary information (e.g. fish consumption) as provided by the EPA (see <http://www.epa.gov/mercury>). In addition we will work with NCAR to involve high school students and teachers in the mission.

PART II – OPERATIONAL CONSIDERATIONS & LOGISTICS

Approx. how many people will be involved in the field campaign? <i>Please specify number of participants and location(s).</i>	~15 scientists to support user supplied instrumentation, including 7-8 on each C-130 flight. This includes an on-site modeling group of ~2-3 scientists.
What other facilities/platforms outside the EOL suite will be deployed? Are any of them non-US facilities?	None.
Are complex inter-facility or inter-agency permissions required for flight operations and/or other facility operations that would benefit from EOL leadership and experience?	No
Is there a need for integrated diplomatic arrangements? (e.g., customs, immigration, focal point with local hosts/governments)	No
If there are multiple instrumentation/operations sites, is there a need for operational coordination?	No
What kind of real-time data display and project coordination needs do you anticipate?	We will need real-time display of ozone, CO, aerosols, and standard meteorological data
Is forecasting support required for project operations?	No-we will have our own on-site modeling team.
What kind of communications capabilities do you expect on site? (e.g., bandwidth)	Std. high speed internet access.
Will operations center and real-time display and coordination services be required? ¹	Yes.
Will you require work space? (e.g., office, lab and storage space)	Yes, ~60-80 m ² total needed for post-flight chemical analysis of samples, storage, work stations, etc.
Will you require system administration support on site?	No
Is there a need for coordinated shipping, lodging or transportation?	No
Will you be shipping hazardous/radioactive material?	Yes. See detailed sheets from each individual instrument.
Will you be shipping expendables? (e.g., radiosondes to local NWS offices)	No
Do you require assistance with various activities/services?	Not exactly sure what this question means.

PART III: DATA MANAGEMENT

What operational data do you need? (e.g., satellite, upper air, radar, surface, oceanographic, hydrological, land characterization, model products)	Hg forecast models will be run operationally in the field. Standard meteorology, satellite and radar products will be used to identify synoptic patterns, fronts, etc (e.g. GOES Vis, IR, water vapor). Other data such as MODIS aerosol products will also be needed.
Do you have any specific real-time data needs to aid in your data collection activities?	No.
Is there a requirement for a local satellite receiver to acquire local or real time polar orbiter or high resolution geostationary satellite data?	No.
Beyond the EOL dataset, will you or your Co-PIs provide additional research data to the project?	Yes. Numerous data-streams will be generated by the multiple PIs involved.
What data analysis products will you provide during the deployment?	Varies by instrument. See specific information below.
What other research data and products do you need?	No other needs beyond those given here.
Is an EOL Field Catalog needed to provide real-time information management, reporting, decision dissemination, data exchange and resource monitoring?	Yes
Do you plan on moving a large amount of data back to your home institution during the project?	No
What arrangements have been made for a comprehensive data archive, including the management and distribution of data from non-EOL platforms?	Data from all user-supplied instruments will be merged with EOL aircraft data at appropriate time scales. Data can be archived by both EOL and the University of Washington.
Do you intend to request restricted data access? ²	EOL data will have no restrictions. Initially only PI's will be able to access the <u>complete</u> dataset for QC and initial analyses. Following a short time interval (6-12 months), the data will be freely available to all.

² Please note that EOL policy will make all EOL data publicly available once the data are quality controlled. If a PI wants to have exclusive access to these data for the first year, s/he has to officially request such a restriction via email from the EOL Division Director (wakimoto@ucar.edu) eight weeks prior to the start of an experiment. The burden will fall on the requesting PI to request the restriction and also to "police" data distribution and access to the data once the restrictions are in place.

PART IV: NSF/NCAR C-130

Contact: Dr. Jorgen Jensen

Email: bj@ucar.edu, Phone: (303) 497-1028

<http://www.eol.ucar.edu/instrumentation/aircraft/C-130>



Operational Considerations:

Preferred flight period	June-July, 2013
Total number of research flight hours requested	96 hours
Total number of flights requested	12
Estimated duration of each flight	8 hours
Total number of flights per week	Approx. one every three days
Particular part(s) of day for flights	Flights will occur during daytime.
Do you plan to fly night missions?	No.
Preferred base of operation	Dual bases required: Western and Eastern US. Ideal locations would be Sacramento in the western US and Maryland in the eastern US.
Alternate base	We are flexible, esp for Eastern base.
Is JeffCo Airport (near Boulder) acceptable as your operations base?	No
Average flight radius from base	~1000 km for most flights
Desired flight altitudes(s)	Extensive time in boundary layer over Eastern US and vertical profiles to max C-130 altitudes
Will there be operations in foreign or military airspace?	No
Number of scientific observers for each flight (max is 15)	7-8
Will you require air to ground communications?	No (other than chat)
Will you require satellite communications above base level? (see Appendix IV)	No

Description of desired flight pattern(s), priorities, and estimate number of flights

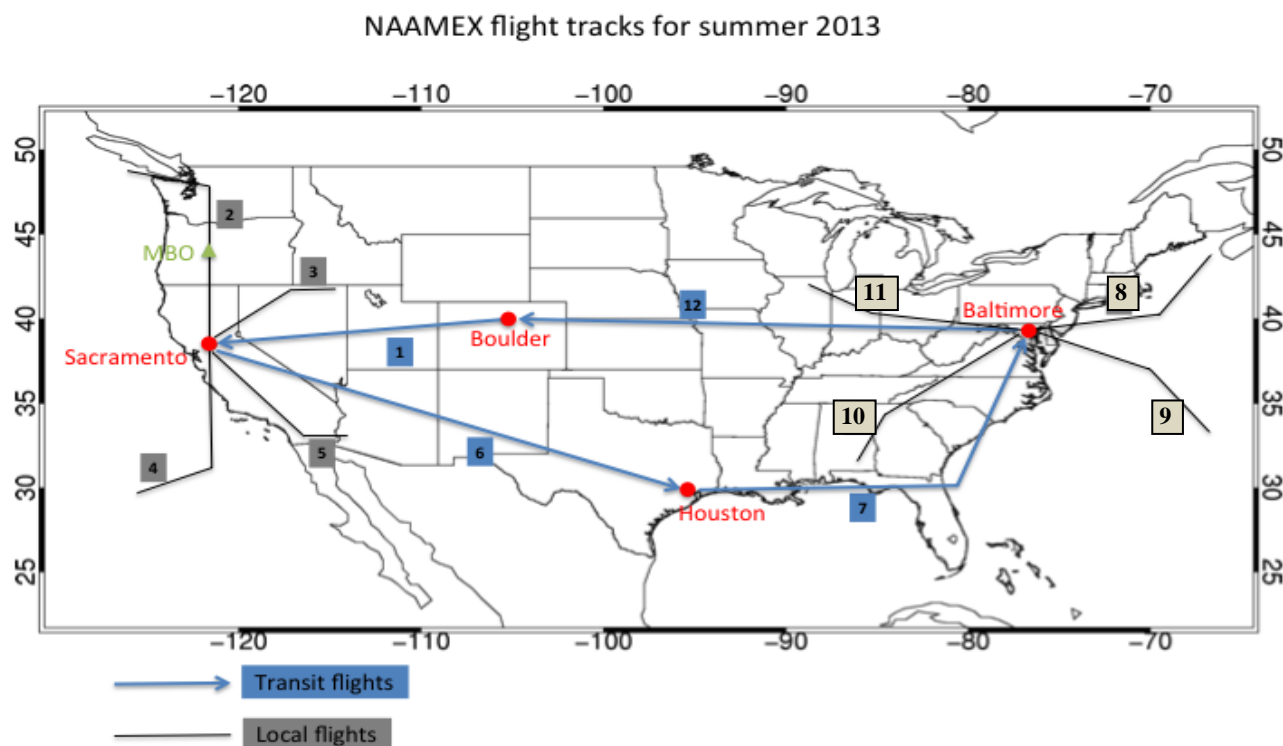


Figure 5. Map of proposed flight tracks for NAAMEX during summer 2013. We assume here that the bases of operation will be Sacramento in the Western U.S. and Baltimore in the eastern U.S. although other locations are possible. Proposed tracks for transit flights are indicated in blue, while local flights are in black.

Flight	Location	Objectives
#1	Transit from Boulder to Sacramento	Nevada natural and anthropogenic sources; Tropospheric profiles of Hg speciation; Free tropospheric influence of surface GOM; Large-scale distribution of Hg
#2	Local flight Sacramento: North transect	Characterization of Hg boundary conditions; Vertical profiles; Mt Bachelor intercomparison
#3	Local flight Sacramento: Nevada sources	Nevada sources; Boundary-layer/Free troposphere exchange; Profiles
#4	Local flight Sacramento: South transect	Characterization of Hg inflow; Vertical profiles; Influence of oxidation;
#5	Local flight Sacramento: California sources	California sources and speciation; Large-scale characterization; Influence of oxidation.
#6	Transit to from Sacramento to Houston	Large-scale distribution; Vertical profiles; Texas sources; Influence of PBL/FT exchange on surface deposition.
#7	Transit from Houston to Baltimore	Southeast large-scale horizontal and vertical distribution; Convective scavenging; Marine boundary layer chemistry.
#8	Local flight Baltimore: NA outflow and North	Evolution of N. American pollution outflow; Large-scale characterization; Marine boundary layer chemistry

	transect	
#9	Local flight Baltimore: NA outflow and South Transect	Evolution of N. American pollution outflow; Large-scale characterization; Marine boundary layer chemistry
#10	Local flight Baltimore: SE sources	SE anthropogenic sources; Convective scavenging; Influence of PBL/FT exchange on surface deposition;
#11	Local flight Baltimore: Ohio River valley sources	Large-scale anthropogenic source characterization; Vertical profiles
Flight #12	Transit from Baltimore to Boulder	Large-scale Hg distribution; west-east evolution of distribution; Vertical profiles.

Airborne Scientific Instrumentation

a) Standard EOL/RAF Measurements as listed on EOL website and in C-130 handbook.

Standard measurements provided automatically when the C-130 is allocated for a project are listed in Appendix II.

b) C - 130 Instrumentation by Request

	Data		Rack	Priority
Description	Rate(s)	Location	Space	0 - 1 - 2
HIMIL Chemistry Inlet (std)	N / A	std aperture	0	1
HIMIL Chemistry Inlet (heated)	N / A	std aperture	1/4	1
Priority code: (0 = un-necessary; 1 = required; 2 = desired but optional)				

c) C - 130 Instrumentation by Special Request

			Rack	Num	Priority
Description	Inlet	Location	Space	Oper	0 - 1 - 2
Fast Ozone (CARI)	simple	cabin w/ aperture	1	1	1
Cabon Monoxide (CARI)	simple	cabin w/ aperture	1	0	1
NO-NOX (CARI)	special	cabin w/ aperture	1	1	1
Mission Coordinator Station	N/A	cabin	1	1	1
Priority code: (0 = un-necessary; 1 = required; 2 = desired but optional)					

d) C - 130 Instrumentation Under Development

Carbon Dioxide (CARI)	simple	cabin w/ aperture	1/4	0	1
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e) User-supplied scientific payload ³

Summary of user-supplied instrumentation:

Investigator	Compounds measured	Weight (kg)	Rack space (bays)	Kwatts@ 60 hz	Kwatts@ 400 hz
Jaffe/Ambrose	GEM, GOM (high time res)	180	2	1.9	
Jaffe	PHg (filter method)	60	1	0.7	
Cantrell/Mauldin	OH, HO ₂ , RO ₂ , H ₂ SO ₄	635	2	2	some
Huey	BrO, Br ₂ , ClNO ₂ , HO ₂ NO ₂ , PANs, HNO ₃ , SO ₂	250	2	2.5	1
Blake	Speciated NMHCs (~60 samples/flight)	127	1	1.2	
Totals		1252	8	8.3	

Jaffe/Ambrose, GEM, GOM, PHg:

Instrument Name:	UW Dual-channel Oxidized Hg System (DOhGS)+Filter pack system for PHg
Primary Contact Name:	Dan Jaffe/Jesse Ambrose
Primary Contact Institution:	University of Washington-Bothell
Primary Contact Phone:	425-352-5357 or 352-3533
Primary Contact Email:	jlambros@u.washington.edu
Individual weight of all components:	DOhGS: 180 kg (not including carrier gases and rack) Filter pack system: 60 kg
Complete size dimensions of all components:	Entire system (minus carrier gases) will fit in two standard C-130 19" racks (one double-wide rack)
Rack-mountable 19" panel space required (Note: depth beyond 25" will overhang in back):	DOhGS: 99 inches (two 19" racks) Filter pack system: 48" (one rack)
Supplying your own 19" rack (yes/no): (Note: racks must survive 9G crash load.)	No
Hazardous material required:	The system will include small quantities of liquid mercury (<1 mL) in sealed, inaccessible permeation tubes within the analyzers and calibrator. A tank of compressed argon will be used as carrier gas for the analyzers; a tank of compressed nitrogen will be used as

³ All user-supplied equipment must meet RAF safety and design specifications. Refer to RAF Bulletin No. 3, No 13 and the Design Guide RAF-DG-00-001 (<http://www.eol.ucar.edu/Bulletins/>).

	carrier gas for the calibrator.
Radioactive sources or materials:	None
Power required (watts, volts, amps):	DOhGS: 1900 total Watts of 120V AC power on two 15 Amp and one 20 Amp circuits Filter pack system: 700 watts of 120 VAC on one 20 Amp circuit
Type of power (DC, 60 Hz, 400 Hz):	60 Hz AC power
External sensor location (if any):	DOhGS: Shrouded duct inlet for reactive gases will be installed in standard aperture plates outside of aircraft (e.g. Zondlo et al 2003). Filter pack system: Rear-facing (sub-micron) non heated metal inlet.
Are signal(s) to be recorded on RAF's Aircraft Data System (yes/no)?	Yes, up to four
If yes: Signal format (digital, analog, serial):	Analog
Full-scale Voltage:	5 V
Range:	0–5 V
Resolution:	16 bit
Sample Rate (1, 5, 250 sps):	1 hz
Need real-time, in-flight, RAF-measurement, serial data feed (RS-232, RS422)?	No
Need IRIG time-code feed?	No
Special sensor calibration service required?	No
Need full-time operator during flight?	2 UW scientists will be on board
Number of lap-top computers for on-board use:	2
Will EOL support be required in preparing the instrument(s) for use on the aircraft (other than inspection, installation and power hook-up)? If so, specify type and lead time.	Some support may be necessary to prepare the system to meet C-130 specifications (i.e. help with certification of wiring, structural strength). Most of this was already done during WAMO, but minor modifications since then.
Will you be using your own recording system?	Yes w/ backup signal to EOL
What additional recording capability is needed? Please provide details on the number of signals, their characteristics, format, synchronous, fire-wire, ethernet, etc.	TBD
If nonstandard output formats and/or data rates are required, how often are the measurements needed? <i>Note: The standard format for processed, RAF output data is net CDF. The standard output media are CD/DVD and ftp transfer.</i>	N/A

Payload ground support needs for user-supplied instrumentation:

	Preflight needs	Postflight needs	Routine Maintenance
	On flight days	On flight days	On non-flight days
Access (hrs)	2	1	5
Power (hrs)	2	1	5
Special support needs			

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Cantrell/Mauldin:

Instrument Name:	
Primary Contact Name:	Chris Cantrell/Lee Mauldin
Primary Contact Institution:	NCAR/University of Colorado
Primary Contact Phone:	303-497-1479
Primary Contact Email:	cantrell@ucar.edu / lee.mauldin@gmail.com
Individual weight of all components:	1400 lbs
Complete size dimensions of all components:	Approx 1 double bay rack (we use a custom rack) – vacuum system mounts on outboard side of rack
Rack-mountable 19” panel space required (Note: depth beyond 25” will overhang in back):	Custom rack
Supplying your own 19” rack (yes/no): (Note: racks must survive 9G crash load.)	Yes – the same rack that flew on TOPSE and PASE
Hazardous material required:	Propane, 0.5% SO2 in N2, 4% SO2 in N2, 1.5% NO in N2, N2, O2
Radioactive sources or materials:	Americium-235 (one for OH and one for HO2/RO2)
Power required (watts, volts, amps):	2000 watts
Type of power (DC, 60 Hz, 400 Hz):	60Hz and 400Hz
External sensor location (if any):	Front left (custom probe plate)
Are signal(s) to be recorded on RAF’s Aircraft Data System (yes/no)?	No
If yes: Signal format (digital, analog, serial):	
Full-scale Voltage:	
Range:	
Resolution:	
Sample Rate (1, 5, 250 sps):	
Need real-time, in-flight, RAF-measurement, serial data feed (RS-232, RS422)?	Yes
Need IRIG time-code feed?	No (We don’t have an IRIG translator card; NTP would be preferred)
Special sensor calibration service required?	No
Need full-time operator during flight?	Yes – 2

Number of lap-top computers for on-board use:	0
Will EOL support be required in preparing the instrument(s) for use on the aircraft (other than inspection, installation and power hook-up)? If so, specify type and lead time.	No
Will you be using your own recording system?	Yes
What additional recording capability is needed? Please provide details on the number of signals, their characteristics, format, synchronous, fire-wire, ethernet, etc.	None
If nonstandard output formats and/or data rates are required, how often are the measurements needed? <i>Note: The standard format for processed, RAF output data is net CDF. The standard output media are CD/DVD and ftp transfer.</i>	N/A

Payload ground support needs for user-supplied instrumentation:

	Preflight needs	Postflight needs	Routine Maintenance
	On flight days	On flight days	On non-flight days
Access (hrs)	1 hour	½ hour	8 hours
Power (hrs)	1 hour	½ hour	8 hours
Special support needs			

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Huey, CIMS (two):

Instrument Name:	
Primary Contact Name:	Greg Huey
Primary Contact Institution:	Ga. Tech
Primary Contact Phone:	404-894-5541
Primary Contact Email:	greg.huey@eas.gatech.edu
Individual weight of all components:	Two CIMS: weight – 250 lbs each for a total of 500 lbs, external 75 lb cylinder needed
Complete size dimensions of all components:	Approximately two racks are needed – but may be able to fit into smaller
Rack-mountable 19” panel space required (Note: depth beyond 25” will overhang in back):	120”
Supplying your own 19” rack (yes/no): (Note: racks must survive 9G crash load.)	no
Hazardous material required:	Compressed gases (nitrogen, dilute CH ₃ I in nitrogen, SF ₆ in Nitrogen, dilute SO ₂ in nitrogen), nitric acid permeation tube
Radioactive sources or materials:	Yes – polonium 210 ion source for both CIMS
Power required (watts, volts, amps):	2.5 kW - @ 110 VAC
Type of power (DC, 60 Hz, 400 Hz):	60 or 400 Hz (60 preferred)
External sensor location (if any):	Inlet is needed for both CIMS

Are signal(s) to be recorded on RAF's Aircraft Data System (yes/no)?	no
If yes: Signal format (digital, analog, serial):	
Full-scale Voltage:	
Range:	
Resolution:	
Sample Rate (1, 5, 250 sps):	
Need real-time, in-flight, RAF-measurement, serial data feed (RS-232, RS422)?	Needed no – preferred yes
Need IRIG time-code feed?	No – NTP server is preferred
Special sensor calibration service required?	no
Need full-time operator during flight?	Yes it is preferred but can cross train
Number of lap-top computers for on-board use:	two
Will EOL support be required in preparing the instrument(s) for use on the aircraft (other than inspection, installation and power hook-up)? If so, specify type and lead time.	no
Will you be using your own recording system?	yes
What additional recording capability is needed? Please provide details on the number of signals, their characteristics, format, synchronous, fire-wire, ethernet, etc.	
If nonstandard output formats and/or data rates are required, how often are the measurements needed? <i>Note: The standard format for processed, RAF output data is net CDF. The standard output media are CD/DVD and ftp transfer.</i>	

Payload ground support needs for user-supplied instrumentation:

	Preflight needs	Postflight needs	Routine Maintenance
	On flight days	On flight days	On non-flight days
Access (hrs)	2	0.5	3
Power (hrs)	2	0	3
Special support needs			

Blake, Whole air sampling for NMHCs and halocarbons:

Instrument Name:	Whole Air
Primary Contact Name:	Don Blake
Primary Contact Institution:	UCI
Primary Contact Phone:	949 824 4195
Primary Contact Email:	drblake@uci.edu
Individual weight of all components:	280 lbs
Complete size dimensions of all components:	One C-130 rack with 13" clearance on both open sides of the rack.
Rack-mountable 19" panel space required (Note: depth beyond 25" will overhang in back):	One C-130 rack with 13" clearance on both open sides of the rack.

Supplying your own 19" rack (yes/no): (Note: racks must survive 9G crash load.)	Yes
Hazardous material required:	No
Radioactive sources or materials:	No
Power required (watts, volts, amps):	1200 watts, 120 volts and 10 amps running
Type of power (DC, 60 Hz, 400 Hz):	60
External sensor location (if any):	Window or other type of forward facing 1/4" to 3/8" stainless steel tubing.
Are signal(s) to be recorded on RAF's Aircraft Data System (yes/no)?	no
If yes: Signal format (digital, analog, serial):	
Full-scale Voltage:	
Range:	
Resolution:	
Sample Rate (1, 5, 250 sps):	
Need real-time, in-flight, RAF-measurement, serial data feed (RS-232, RS422)?	RS-232
Need IRIG time-code feed?	no
Special sensor calibration service required?	no
Need full-time operator during flight?	yes
Number of lap-top computers for on-board use:	1
Will EOL support be required in preparing the instrument(s) for use on the aircraft (other than inspection, installation and power hook-up)? If so, specify type and lead time.	No
Will you be using your own recording system?	Yes
What additional recording capability is needed? Please provide details on the number of signals, their characteristics, format, synchronous, fire-wire, ethernet, etc.	Ethernet if possible
If nonstandard output formats and/or data rates are required, how often are the measurements needed? <i>Note: The standard format for processed, RAF output data is net CDF. The standard output media are CD/DVD and ftp transfer.</i>	No


Payload ground support needs for user-supplied instrumentation:

	Preflight needs	Postflight needs	Routine Maintenance
	On flight days	On flight days	On non-flight days
Access (hrs)	1	1	0
Power (hrs)	1	1	0
Special support needs			

End user supplied instrument list.

Attachments

- 1) First page for accepted Nature Geosciences (Available on-line 12/18/2011).
- 2) Letter from Nicola Pirrone, Director of European GMOS program.



LETTERS

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Formation and fate of oxidized mercury in the upper troposphere and lower stratosphere

Seth N. Lyman* and Daniel A. Jaffe

1 Mercury contamination affects many aquatic ecosystems¹. The
2 atmosphere is the main transport route for this toxicant².
3 According to aircraft measurements, the upper troposphere
4 and lower stratosphere are depleted in gaseous elemental
5 mercury^{3,4} but enriched in oxidized, particle-bound mercury^{5,6}.
6 It is therefore assumed that mercury is oxidized in the strato-
7 sphere, and then incorporated into stratospheric aerosols⁶.
8 However, direct evidence for mercury oxidation in the strato-
9 sphere is missing. Here, we present simultaneous measure-
10 ments of elemental and oxidized mercury concentrations in air
11 of stratospheric origin, collected during an aircraft campaign
12 over North America and Europe in 2010. We show that levels of
13 oxidized mercury are strongly correlated with tracers of strato-
14 spheric air. Concentrations of total and elemental mercury, in
15 contrast, are negatively correlated with these tracers. Together,
16 the findings indicate that elemental mercury is oxidized in
17 stratospheric air masses. We develop a numerical model of
18 atmospheric mercury, based on the assumption that mercury is
19 oxidized in the upper troposphere and lower stratosphere.
20 The resultant vertical profiles—which depict a rapid decline in
21 mercury concentrations with increasing stratospheric height—
22 resemble those seen in other studies, and indicate that mercury
23 has a relatively short stratospheric lifetime. We suggest that
24 following oxidation, mercury is removed from the stratosphere
25 by sedimentation and entrainment processes common to all
26 stratospheric particles.

27 Although previous studies have measured atmospheric mercury
28 by aircraft, our instrument differentiates between elemental
29 and oxidized forms. The National Center for Atmospheric
30 Research (NCAR) C-130 has a typical cruising altitude of 6,000–
31 7,000 m and is usually well below the tropopause. However,
32 during the October–November 2010 Western Airborne Mercury
33 Observations campaign aboard the NCAR C-130, we intercepted
34 stratosphere-influenced air on a few occasions. The strongest
35 of these events was during a flight from Bangor, Maine, to
36 Broomfield, Colorado, on 5 November (Fig. 1). During this
37 flight, oxidized mercury (Hg(II)); gaseous + particle-bound) was
38 positively correlated with stratospheric tracers (ozone and potential
39 vorticity (PV)) and negatively correlated with CO ($R^2 = 0.91$),
40 indicating that Hg(II) increased with increasing stratospheric
41 influence. The Global Forecast System model confirms that the
42 flight altitude was at or near the tropopause during these events (see
43 Supplementary Information).

44 We observed a negative linear correlation between gaseous
45 elemental mercury (Hg(0)) and Hg(II) in stratosphere-influenced
46 air (Fig. 2), providing evidence that Hg(0) is converted to Hg(II) in
47 the stratosphere^{3,4,6}. The slope for this relationship was -0.53 ± 0.08
48 (95% confidence interval; $R^2 = 0.89$; that is an Hg(0) loss of
49 1 pg m^{-3} was associated with an Hg(II) gain of 0.53 pg m^{-3}), greater
50 than the slope of approximately -1 that would be expected if no loss

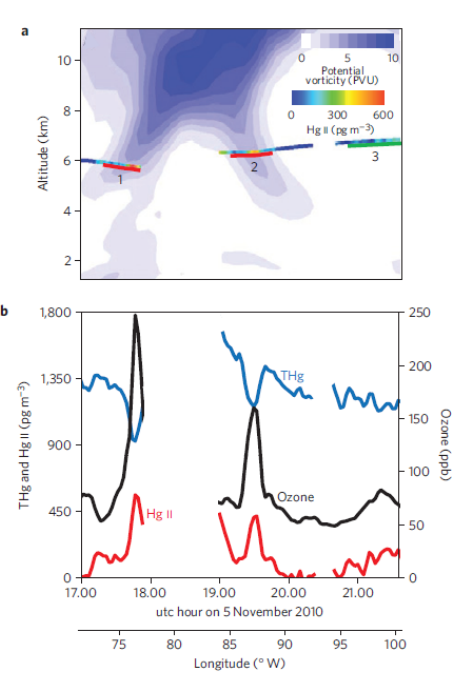


Figure 1 | Mercury and ozone during a 5 November 2011 flight through stratosphere-influenced air. **a**, Flight track, coloured by Hg(II) concentration, along 40° to 44° latitude. The background is coloured by potential vorticity. Solid red lines indicate stratosphere-influenced air ($PV > 1.5 \text{ PVU}$; Events 1 and 2) and the solid green line indicates high-Hg(II) air that was not recently influenced by the stratosphere (Event 3). **b**, Concentrations of THg, Hg(II) and ozone. The x axes correspond to **a** and **b**. The data gap from 18:00 to 19:00 exists owing to diplomatic restrictions on sampling in Canadian airspace.

mechanism existed for Hg(II) (ref. 7). An air mass with elevated Hg(II) that did not show recent stratospheric influence ($O_3 < 80 \text{ ppb}$, $PV < 1 \text{ PVU}$; green in Figs 1a and 2) was encountered on the same flight and had a more negative slope of -0.93 ± 0.25 ($R^2 = 0.62$). These two slopes were significantly different ($p = 0.003$).

The Hg(0)–Hg(II) slope in stratosphere-influenced air was probably greater than -1 because total atmospheric mercury (THg)

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Page 24



Dr. Dan Jaffe, Professor
University of Washington-Bothell
18115 Campus Way NE
Bothell, WA 98011-8246
USA

Subject: letter of support for NAAMEX

Dear Dan:

We are very much looking forward to collaborating with you on your project "The North American Airborne Mercury Experiment (NAAMEX)".

As Coordinator of the European Global Mercury Observation System (GMOS) project (www.gmos.eu), I can say that NAAMEX is an excellent focus for collaboration between the US and European Hg researchers.

GMOS is a five year project (2010-2015), funded by the European Commission and it is aimed at the establishment of a worldwide observation system for the measurement and modeling of atmospheric mercury species, in ambient air and precipitation samples, on regional and global scales. GMOS includes ground-based monitoring stations, shipboard measurements over the Pacific, Atlantic and Indian Oceans and major European Seas, as well as aircraft-based measurements in the UTLS (CARIBIC) and in mid-troposphere (ETMEP). GMOS involves 23 partners from all over the world and is in the process of establishing bi-lateral agreements with existing regional programs in the USA and Canada (NADP) and also in Asia.

As part of GMOS we are now pursuing advanced Hg modeling and observation activities, and as such the NAAMEX project, with reference to its objectives and expected results, is both timely and important for the overarching goal of establishing a globally coordinated mercury observation systems by merging state-of-the-art observations and modeling capabilities available in leading research and university institutions worldwide.

I have discussed NAAMEX with the GMOS Steering Committee and we can commit to the following collaborative research with the NAAMEX project:

- 1) Use the NAAMEX observations to improve global models and better understand the sources and sinks of Hg. The group involved in WP7 (Global Mercury Modeling) of GMOS, led by Dr. Oleg Travnikov, MSC-East in Moscow, to which a number of institutions contribute (i.e., CNR-IIA, Dr. Ian M. Hedgecock; HZG, Dr. Volker Matthias), is particularly enthusiastic to collaborate with you on NAAMEX both pre- and post-mission.
- 2) Use the NAAMEX observations in regional models as boundary conditions and to improve the parameterizations of Hg. The group involved in WP8 (Regional Mercury Modeling), led by Dr. Volker Matthias, HZG in Hamburg, to which a number of other institutions contribute (i.e., CNR-IIA, Dr. Ian M. Hedgecock; MSC-East Dr. Oleg Travnikov), is very interested to collaborate in terms of data sharing and validation of regional and global scale models. The regional and global modeling activities will be based on state-of-the-art atmospheric models (i.e., ECHMERIT, GLEMOS, WRF-Chem, CMAQ), that already participated in the UNECE-TF HTAP modeling intercomparison.



- 3) Collaboration with the European Tropospheric Mercury Experiment Program (ETMEP), which is part of the WP5 (Aircraft-Based Observations) of GMOS, led by Prof. Ralf Ebinghaus (HZG in Hamburg) with contributions from CNR-IIA (Prof. Nicola Pirrone) and MPI in Mainz (Prof. Franz Slemr), and which is now in the planning phase for a series of aircraft missions similar to NAAMEX. As part of WP5 we also have the contribution of the CARIBIC program which will provide mercury measurements in the UTLS and will allow to validation of ETMEP observations. We strongly recommend a close cooperation of NAAMEX with WP5 scientists.

In addition, as Chair of the UNEP Global Mercury Fate and Transport Partnership (www.unep.org) charged with supporting the preparation of the future international treaty on global mercury contamination, I would like to mention that NAAMEX would also be an excellent contribution to the over-arching goals of the F&T partnership that is in charge of contributing to the next synthesis report for the UNEP Governing Council planned for 2013.

In closing, the GMOS consortium, that represents nearly 50 distinguished and well known scientists worldwide, feels that NAAMEX is an exciting opportunity for US-Europe collaboration to improve our understanding of the global Hg cycle.

We look forward to working with you and the entire NAAMEX team.

Sincerely yours,

A handwritten signature in blue ink that reads 'Nicola Pirrone'.

Prof. Nicola Pirrone
(Director)