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Summary Report For: NAG 1 1767

Measurement of Reactive Nitrogen Compounds over the Tropical Pacific

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Introduction

This report covers the Tropospheric Trace Gas and Airborne Measurement Group's (TTGAMG) participation in the NASA Global Tropospheric Experiment (GTE) Pacific Exploratory Mission: Tropics A (PEM-TA) field measurement program. With other university and NASA centers, the Georgia Institute of Technology Airborne Laser Induced Fluorescence Experiment (GITALIFE) was deployed on the NASA Ames DC-8 aircraft for the measurement of NO and NO_2. One of the PEM-TA objectives was to collect the data and interpret the fast photochemical processes taking place in the remote and relatively pristine environment over Hawaii, Tahiti, Easter Island, New Zealand, and Fiji. The upload at NASA Ames Research Center started on June 26th 1996, continued with 17 data flights, that cumulated on October 11th with the download.

General Overview

The time span of PEM-TA encompasses both the finest hour and lowest point of my career at the Georgia Institute of Technology. With the integration of the new seeded Nd:YAG and Master Oscillator Power Oscillator (MOPO) solid state lasers on an airborne platform for the first time and the ability to land at our first base of operation with a plot of the first flight's data will never be matched. That is not to say all was in the bank as we departed Georgia Tech to participate in PEM-TA.

That milestone has been a long time in the making and is nice to have it behind us. We were able to collect NO data on all 17 data and transit flights and NO_2 on 12 of the flights. Our data set is showing well as modelers and their calculated fast photochemical parameters over all altitudes are in very close agreement with our measurements. Our data set is far better than most ground based data sets that we are aware of at this time. The question that is continually asked about why becomes difficult to answer. We have changed both the flow line and photolysis system and they can not be easily separated. Both oral presentations (Spring AEAP meeting) and publications (Photofragmentation Two-Photon Laser-Induced Fluorescence Detection of NO_2 and NO: Comparison of Measurement with Model Results on Airborne Observations During PEM-Tropics A) have discussed this new state of the art trace gas sensor which will make its next deployment on the NASA Dryden DC-8 in PEM-TB.

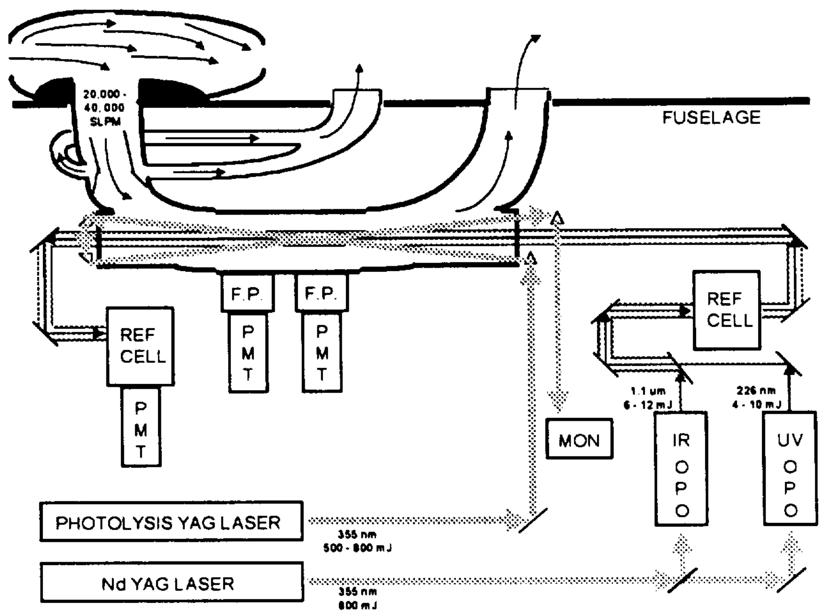
Aircraft Integration and Instrument Layout

There are two major changes to TTGAMG's Laser Induced Fluorescence (LIF) system that have been implemented since the previous NASA GTE field programs. First, the laser systems incorporated new technologies into the proven LIF sensor for measuring NO in order to improve sensitivity and specificity (freedom from potential interferences). The second major change came in the size of the diffuser style inlet and flow line. Nether of these modifications were trivial and the engineering effort substantial.

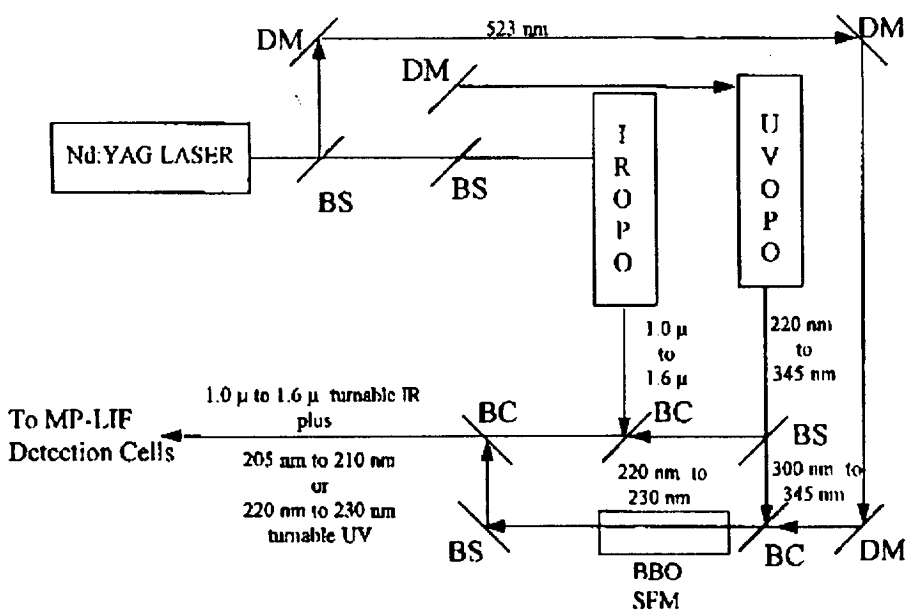
The laser modifications to the system have two major points. The first is the integration of the Master Oscillator Power Oscillator (MOPO) solid-state lasers. The independently tunable near-IR (1.1 micron from a Spectra-Physics MOPO-730) and deep UV (226 nm from a Spectra-Physics MOPO-730 FDO) solid state laser system created a narrow-linewidth and nearly ten-fold higher energies than were achievable with the previous dye laser based system. This is where the improved sensitivity and specificity was gained. The narrow line width allows the system for the first time to distinguish the ^{14}NO and ^{15}NO isotopes.

A second component related to the introduction of these solid state lasers was the need for the seeded Nd:YAG lasers (Spectra-Physics model GCR 290) to pump the MOPOs. A second Nd:YAG was also installed to generate the photolysis wavelength need for the measurement of NO_2. The NO_2 is probed on every other 10 Hz laser shot and it is occurring in the center of the 4-inch flow line. The ability to quantitatively photofragment NO_2 into detectable NO using a rapid (30 - 50 microsecond) paired photolysis/probe lasers pulse approach produces an effective temporal "freezing" of the ambient sample.

The second major change to the system was the incorporation of the larger diffuser inlet and 4 inch flow line. The flow diffuser-style nozzle acts to slightly pressurize the inlet (e.g. ~80 mbar) and segregate large aerosols. The purpose of which is to decrease the residence time in the sampling manifold and therefore decrease the wall-induced memory or chemical effects. Flow rates of 35,000 l/min provides a residence time in the entire manifold of 50 milliseconds was achieved. Figure 1 is a depecition on the pseudo-wall-less system in cartoon form



and schematically in figure 2.



RESULTS

The improvements to the system are obvious when comparing measurements with modeled calculations and comparing PEM-TA with other field campaigns. (See J. Bradshaw et al. 1999 and Schultz et al. 1998) Figure 3a and 3b depicts the difference in measured and modeled results from Pacific Exploratory Mission: West A (PEM-WA) and PEM-TA. Note the divergence in measured and modeled NO_2 values as the altitude is increased for figure 3a. The median value for the ratio of $(NO_2)_{meas}/(NO_2)_{calc}$ was 3.36 for PEM-WA (Crawford et al. 1996) and 0.93 for PEM-TA (Bradshaw et al. 1999).

Figure 3a

NO2expt/NO2calc vs Altitude
PEM-West A

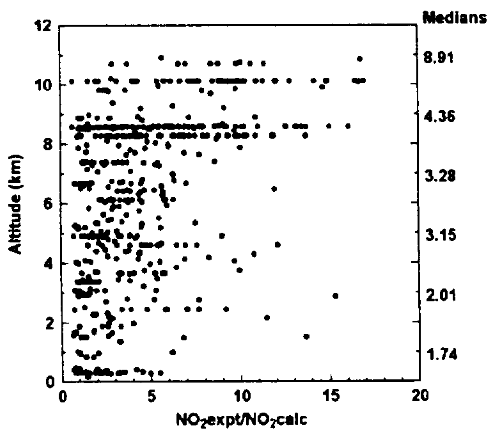
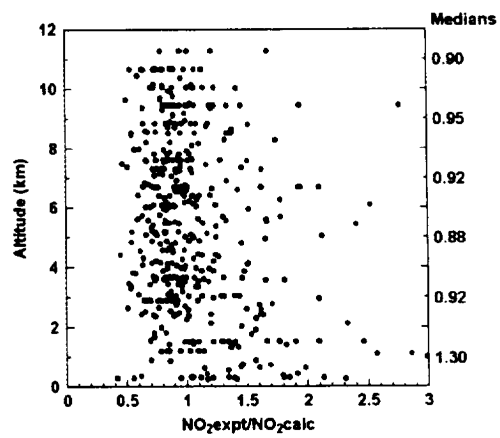


Figure 3b

NO2expt/NO2calc vs Altitude
PEM-Tropics A



All of the data that was collected and used for these modeling results is available to the general public through the DAAC archive release of the GTE PEM-TA data. A paper was also presented at the spring AEAP Meeting, as well as, a publication submitted to Geophysical Research Letters titled "Photofragmentation Two-Photon Laser Induced Fluorescence Detection of NO_2 and NO: Comparison of Measurements with Model Results Based on Airborne Observations During PEM-Tropics A"

References

"Photofragmentation Two-Photon Laser Induced Fluorescence Detection of NO_2 and NO: Comparison of Measurements with Model Results Based on Airborne Observations During PEM-Tropics A," J. Bradshaw, D. Davis, J. Crawford, G. Chen, R. Shetter, M. Muller, G. Gregory, G Sachse, D. Blake, B. Heikes, H. Singh, J. Mastromarino, S. Sandholm, Geophysical Research Letters (accepted).

"Photostationary state analysis of the NO_2-NO system based on airborne observations from the western and central North Pacific," J. Crawford, D. Davis, G. Chen, J. Bradshaw, S. Sandholm, G. Gregory, G. Sachse, B. Anderson, J. Collins, D. Blake, H. Singh, B. Heikes, R. Talbot, and J. Rodriguez, Jour. Geophys. Res., 101, 2053-2072, (1996).

"On the Origin of Tropospheric Ozone and NO_x over the tropical South Pacific," M. Schultz, D. Jacob, J. Logan, Y. Wang, D. Blake, J. Bradshaw, G. Gregory, G. Sachse, B. Heikes, R. Shetter, H. Sing, and R. Talbot, Jour. Geophys. Res., (submitted).