DFGAS (Difference Frequency Generation Absorption Spectrometer) for the Measurement of Formaldehyde - Instrument Description from Alan Fried

The new DFGAS instrument utilizes a new room temperature infrared (IR) laser source based upon non-linear difference frequency generation (DFG) in the measurement of $CH₂O$. This is in contrast to our previous DCALS instrument, which employed a liquid-nitrogen cooled tunable diode laser. Aside from the laser source, the measurement approaches in the two systems are essentially the same, but the new DFG instrument has the potential for significantly improved performance [Weibring et al., 2006, 2007].

Mid-IR laser light is generated in the DFG system by mixing the output of two near-IR room temperature laser sources (one at 1562-nm and the other at 1083-nm) in a periodically poled lithium niobate (PPLN) non-linear wavelength conversion crystal. The mid-IR difference frequency at 2831.6 cm⁻¹ (3.53-µm) is generated at the PPLN output and directed through a multipass astigmatic Herriott cell (100-m pathlength using \sim 4-liter sampling volume) and ultimately onto IR detectors employing a number of optical elements. A portion of the IR beam is split off by a special beam splitter (BS) before the multipass cell and focused onto an Amplitude Modulation Detector (AMD) to capture and remove optical noise from various components in the difference frequency generation process. A third detection channel from light emanating out the back of the beam splitter is directed through a low pressure $CH₂O$ reference cell and onto a reference detector (RD) for locking the center of the wavelength scan to the absorption line center. The mid-IR DFG output is simultaneously scanned and modulated over the CH2O absorption feature, and the second harmonic signals at twice the modulation frequency from the 3 detectors are processed using a computer lock-in amplifier [Weibring et al. [2006].

Fig. 1*:* **(Left Panel)** *Optical schematic of DFG system, which includes WDM (wavelength division multiplexer beam combiner), HF (hybrid fiber ball lens), FL (focusing lens), PPLN (periodically poled lithium niobate non-linear wavelength conversion crystal), BD (beam dump), Ge (germanium filter), IL (imaging lens), P (prism reflector), OS (optical shutter), BS (coated beam splitter), OAP (off axis parabolic focusing mirrors), RD (reference detector), CD (cell detector), MPC (multipass cell) and associated front mirror MPM, and AMD (amplitude modulation detector). A reference cell (RC), containing pure CH2O at low pressure precedes the RD.* **(Right Panel)** *DFG system mounted in the cabin of NOAA's P-3 aircraft during the TexAQS*

mission. The rack to the left contains; calibration and zero air generation systems, along with flow controllers and various power supplies. The rack to the right houses; operator monitor, service oscilloscope, control and data acquisition computer, spectrometer enclosure (lasers and DFG module, fiber amplifiers, multipass cell, detectors), and air-conditioning unit. The entire system, including the rack and vacuum pump weighs 640 pounds, which is ~ 110 pounds lighter than the TDLAS.

Ambient air is continuously drawn through a heated rear-facing inlet at flow rates around 9 standard liters per minute (slm), through a pressure controller, and through the multipass Herriott cell maintained at a constant pressure around 50-Torr. Ambient measurements are acquired in 1 second increments for time periods as long as 60 to 120-seconds (to be determined during the campaign), and this will be followed by 15-seconds of background zero air acquisition, using an onboard CH2O scrubbing unit. The zero air is added back to the inlet a few centimeters from the tip at flow rates ~ 2 to 3 slm higher than the cell flow. This frequent zeroing procedure very effectively captures and removes optical noise as well as residual outgassing from inlet line and cell contaminants. Retrieved CH₂O mixing ratios are determined for each 1-second ambient spectrum by fitting to a reference spectrum, obtained by introducing high concentration calibration standards (\sim 3 to 7-ppbv) from an onboard permeation calibration system into the inlet approximately every hour. The calibration outputs for the two permeation tubes employed are determined before and after the field campaign using multiple means, including direct absorption employing the Beer-Lambert Law relationship. The 1-second ambient $CH₂O$ results can be further averaged into longer time intervals for improved precision. However, in all cases the 1-second results are retained. This flexibility allows one to further study pollution plumes with high temporal resolution, and at the same time study more temporally constant background CH₂O levels in the upper troposphere using longer integration times.

During the TexAQS-2006 airborne study we achieved 95% data coverage over the entire field campaign, and determined mission ensemble median $CH₂O$ 2 σ limits of detection (LODs) of 232-pptv and 42-pptv for, respectively, 1-second and 1-minute measurements. During ARCTAS a number of additional improvements should improve the performance even further.

References

- Weibring, P., D. Richter, A. Fried, J.G. Walega, and C. Dyroff, *Ultra-High-Precision Mid-IR Spectrometer II: System Description and Spectroscopic Performance,* Appl. Phys. **B**, doi: 10.1007/s00340-006-2300-4, 2006.
- Weibring, P., D. Richter, A. Fried, and J.G. Walega, *First Demonstration of a High Performance Difference Frequency Spectrometer on Airborne Platforms,* in preparation for submission to Optics Express, 2007.