Airborne and ground-based measurements of atmospheric constituents by difference frequency generation absorption spectroscopy

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Mid-Infrared laser spectroscopy has been widely employed in the laboratory, on ground-based, airborne, and balloon-borne platforms to study atmospheric constituents, processes, and transformations. Traditionally, liquid-nitrogen cooled tunable lead-salt diode laser absorption spectrometers (TDLAS) have been employed [1], although more recent studies now also include spectrometers based upon quantum cascade laser sources [1]. In 2006 a new laser source based upon a difference frequency generation (DFG) mid-IR laser was deployed for the first time on an airborne platform by Weibring and colleagues at the National Center for Atmospheric Research [2]. As discussed in their publications, this new spectrometer has the potential to significantly improve the performance of airborne trace gas measurements over more traditional TDLAS-based systems. The DFG-based spectrometer was deployed in 2006 during three different airborne studies spanning more than 300 flight hours to measure the important trace gas formaldehyde (CH_2O). More recently this new spectrometer was deployed during the 2008 Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) campaign to measure CH_2O over the course of ~ an additional 170 flight hours. The DFG spectrometer was continuously improved between each study, and during ARCTAS we typically achieved 1σ minimum detectable line center absorbances of 3×10^{-6} and 4×10^{-7} for 1-second and 1-minute averages, respectively, when the aircraft cabin pressure was stable. This is an improvement of at least a factor of two compared to our past TDLAS system performance. In addition, ground-based measurements of CH_2O and methane were also acquired with this instrument for one month at Barrow, Alaska during the 2009 Ocean-Atmosphere-Sea Ice-Snowpack project (OASIS). Although an operator was on location, the latter measurements were essentially carried out autonomously once the instrument was set up and running. In fact, the instrument was operated remotely for several weeks using Virtual Network Connection.

The ARCTAS study had many objectives, and three areas where our CH_2O measurements will help to improve our understanding of atmospheric processes involve: transport and transformations of pollution to the arctic, halogen chemistry from the arctic snowpack, and the effects of fire emissions on ozone formation and hydrogen radicals as well as other trace gases like CH_2O downwind and in the upper troposphere. In the case of the latter, our 1-second CH_2O observations are essential, as typical plume intercepts are only a few seconds to 10's of seconds in duration.

The present talk will give a brief overview of the latest DFG spectrometer and the modifications that have been implemented for improved performance. This talk will then present some specific examples of airborne CH_2O measurements during the ARCTAS study and how these observations are being used to further our understanding of arctic halogen chemistry related to emissions of trace reactive gases from the snowpack as well as emissions of CH_2O from boreal forest fires.

References

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[2] P. Weibring, D. Richter, J. Walega, A. Fried: "First demonstration of a high performance difference frequency spectrometer on airborne platforms", Optics Express **15**, pp.13476–13495 (2007)