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Continuous in field measurements of N_2O concentration and its isotopologue and isotopomer ratios, with a field deployed mid–infrared, wavelength scanned, cavity ring-down spectroscopy instrument

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In recent years wavelength scanned cavity ring-down spectroscopy (WS-CRDS) has proven to be a highly stable and reliable method for doing molecular concentration and isotopologue measurements in the field, with high precision even under strenuous conditions such as the Greenland Ice sheet. These instruments use molecular absorption of light in the near infrared (NIR) wavelength region 1 - 1.6 μm from the vibrational overtone transitions, to quantify concentration and isotope ratios. Even with the strong signal enhancement provided by the ring-down method, measurements are often limited by the low molecular concentration. This problem can be overcome by performing WS-CRDS in the mid infrared (MIR, 3-8 μm) at the fundamental vibrational transition of the molecule. This provides absorption strengths being several orders of magnitude stronger than in the NIR. However the limited available optical materials and the increased thermal noise of the MIR, makes MIR-WS-CRDS a demanding task.

MIR-WS-CRDS provides possible enhanced sensitivity to several species of molecules, (e.g. CO_2, CH_4, NO, NO_2 and isotopologues are a few) and can be combined with various sample preparation systems, to study substances not being on gas form. In this work N_2O is being addressed, as this is a potent greenhouse gas, which is increasing in atmospheric concentration by 0.25%/yr, and is not yet fully understood. Although the sensitivity is not as good as IRMS ($\pm$ 0.1 ‰), MIR-WS-CRDS has the vantage of being easy to use and is able to directly distinguish the isotopomers of N_2O which is not possible with conventional IRMS. A comparison measurement between the presented MIR-WS-CRDS and a GC-IRMS is presented, along with continuous isotopologue N_2O measurements from an agricultural landsite in the western part of Denmark. Finally will, proceedings toward applying MIR-WS-CRDS to N_2O in prehistoric atmospheric samples captured in air bubbles of the Greenland ice sheet, be addressed.

This work presents the first continuous infield measurements, with a standalone MIR-WS-CRDS instrument. The presented instrument is based entirely on thermo electrically cooled technology, enabling the system to be run unattended for extended periods of time. The light source is a Mode-Hop-Free CW Quantum Cascade Laser centered at 4.55 μm, with a narrow line width and power exceeding 100 mW, operating at room temperature. A fast low noise TE cooled MCT detector is used to record the $\sim$10 μs optical decay from the ring-down cavity, which provide an effective absorption path length of numerous kilometers. Ambient atmospheric gas samples may be introduced directly into the instrument achieving a precision in N_2O concentration < 0.1 ppbv in only few seconds of data acquisition time, while the isotopologues ^{15}N and ^{18}O of N_2O are analyzed at < 1 ‰ precision within 30 sec. The mobility of the instrument makes it ideal for future studies such as N_2O measurements in the Troposphere and in remote locations such as tundra peat bogs.