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Spectroscopic Uncertainty
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Long-term monitoring of long-lived greenhouse gases (ghgs) requires sub-percent (0.1%) accuracy, and observations sho
TEXT
Long-term monitoring of well-mixed greenhouse gases (ghg) requires instruments that can measure at 0.1% accuracy (K
The accuracy of ghg measurements is limited by both instrument capabilities and the accuracy of ghg spectroscopy. Ma
Recently, Dual-Comb Spectroscopy (DCS) has emerged as a candidate to augment the NOAA network, because it can
These capabilities have been demonstrated in field-deployments. Reiker et al, 2014, was the first to deploy DCS to mea
Although DCS is capable of measuring highly resolved spectra with absolute frequency stability, accurately measuring
For example, Waxman et al, 2017, also found that retrieved methane concentrations disagreed when using different spe
Motivated by achieving long-term monitoring capabilities, We compare ghg retrievals from different spectroscopic datal
Dual-Comb Spectroscopy Technique Dual-Comb Spectroscopy Technique The technique of DCS employs laser frequenc
To achieve laboratory-level accuracy, the frequency combs need to have long-term frequency stability and maintain com
Field Setup In this study, our DCS generates light between 6,000 and 6,400 cm^{-1} (1560 - 1660 nm) at 80,000 distinct a
The DCS design is outlined in Sinclair et al, 2015. In summary, both of the frequency combs are powered by a 10 mW
The DCS was mounted atop a building at the NIST facility between 21 September to x November, 2016 in Boulder, CC
A commercial cavity ring-down spectrometer (Picarro Model 3012) was also deployed to the field, alongside a pressure
Although the point sensor is acting as a reference for our field experiment, it should be noted that the point-sensor and
Pressure and temperature measurements are also taken on this tower. These measurements also benchmark our pressur
Retrieval Approach Problem Statement The retrieval problem for an open path system is that the depth of the measur

$$[\text{ghg}] = \text{ghg}_{cd} \frac{}{total_c d - H_2O_{cd}}$$

In Eq. **??**, $ctotal_{cd}$ is the column density of air, ghg_{cd} is the column density of ghg molecules, and H_2O_{cd} is the column

$$\rho_{dry} = \frac{p(1-[H_2O])}{rT}$$

$$dry_{cd} = \rho_{dry} \delta x$$

Eq. **??** provides the relationship between the number density of dry air, denoted ρ_{dry}, and the atmospheric state, which
Here, we will quantify the biases induced in retrieving greenhouse gas concentrations, for both the retrieval of greenhou
Retrieval Algorithm In order to retrieve the ghg concentrations from the measured spectra, we performed a non-linear

$$\tau = \sum_i^n [ghg] cd_{dry} \sigma$$

$$I = exp^{-\tau}$$

The resulting modeled transmission is scaled by a Legendre polynomial basis set, which approximates the underlying lo

Evaluations of the forward model map the chemical and thermo-dynamic state (e.g., concentrations, pressure, and temp

Here, x_i is our state vector at the ith iteration, and it includes the vertical column density (vcd) of each of the gases be
A crucial aspect of our retrieval is that our state vector contains the column density (total number of trace gas molecu
Spectral line-lists Since we are retrieving pressure and temperature from the shape of the absorption lines, it is necessa
The temperature, pressure, and wavelength absorption features unique to each molecule are calculated from spectral pa
Table **??** displays the line-lists being used in our study. We use the HITRAN 2008, 2016, and 2020 line-lists, in additio

Field Results Methane retrievals disagree more than CO_2
Regional-scale gradients of CO_2 in the atmosphere are about 0.25% (1 ppm). Inferring CO_2 sources at this scale requir
Fig **??** shows the retrieved time series for a two-week period in our study. Our algorithm retrieves CO_2 over the windo
Fig. **??**C displays the ratio between the CO_2 concentrations retrieved from the DCS and the CO_2 measurements from
Despite the fact that the DCS is solely relying on information from the spectroscopic databases, the ratio between the
co2$_timeseries.pdf Retrieved CO_2$ concentrations with line-lists outlined in Table **??** from our DCS field deployment in Bould
On the other hand, there is considerable disagreement for retrieved methane. Methane's spectroscopy is continuously e
We used the Hitran 2008, 2016, and 2020 line-lists , in addition to the TCCON line-list, to retrieve methane concentrat
Fig. **??**C shows the ratio between the retrieved methane from the DCS and the measured concentrations from the Pica
ch4$_timeseries.pdf Retrieved CH_4$ from our DCS field-deployment. The figure is arranged as in Fig. **??**.
vcd$_timeseries.pdf Retrieved variables required to calculate the dry column density$ (dry$_{cd}$). This includes, from top to bottom, c
Tracing error sources from field retrievals

Ch_4 Correlations				
Variable	Hitran 2008	Hitran 2016	Hitran 2020	TCCON
H_2O amount				
H_2O error				
pressure error				
temperature error				
CO_2 correlations				
H_2O amount				
H_2O error				
pressure error				
temperature error				

Error correlations for our retrievels from the field-deployed D