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Study of Differential Column Measurements for Urban Greenhouse Gas Emission Monitoring

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Urban areas are home to 54% of the total global population and account for $\sim$ 70% of total fossil fuel emissions. Accurate methods for measuring urban and regional scale carbon fluxes are required in order to design and implement policies for emissions reduction initiatives.

In this paper, we demonstrate novel applications of compact solar-tracking Fourier transform spectrometers (Bruker EM27/SUN) for differential measurements of the column-averaged dry-air mole fractions (DMFs) of CH_4 and CO_2 within urban areas. Our differential column method uses at least two spectrometers to make simultaneous measurements of CO_2, CH_4 and O_2 column number densities. We then compute the column-averaged DMFs X_G for a gas G and the differences ΔX_G between downwind and upwind stations. By accurately measuring the small differences in integrated column amounts across local and regional sources, we directly observe the mass loading of the atmosphere due to the influence of emissions in the intervening locale. The inference of the source strength is much more direct than inversion modeling using only surface concentrations, and less subject to errors associated with modeling small-scale transport phenomena.

We characterize the differential sensor system using Allan variance analysis and show that the differential column measurement has a precision of 0.01% for $X_{\mathrm{CO_2}}$ and $X_{\mathrm{CH_4}}$ using an optimum integration time of 10 min, which corresponds to standard deviations of 0.04 ppm, and 0.2 ppb, respectively. The sensor system is very stable over time and after relocation across the contiguous US, i.e. the scaling factors between the two Harvard EM27/SUNs and the measured instrument line function parameters are consistent.

We use the differential column measurements to determine the emission of an area source. We measure the downwind minus upwind column gradient $\Delta X_{\mathrm{CH_4}}$ ($\sim$ 2 ppb, 0.1%) across dairy farms in the Chino California area, and input the data to a simple column model for comparison with emission strengths reported in the literature. Our model assumes that air parcels within the air column are transported with a mass-enhancement-weighed horizontal wind velocity U, which is estimated using surface wind speeds measured at nearby airports and assuming a wind profile power law up to the mixing height, to which CH_4 emissions are transported vertically by turbulent flow. The emission estimate using differential column measurements is dominated by the uncertainty in the transport i.e. U, not the differential column measurements themself. Furthermore, we derive spatial column gradient ratios $\Delta X_{\mathrm{CH_4}}/\Delta X_{\mathrm{CO_2}}$ across Pasadena within the Los Angeles basin, and determine values that are consistent with regional emission ratios from the literature.

Our precise, rapid measurements allow us to determine short-term variations (5 to 10 minutes) of $X_{\mathrm{CO_2}}$ and $X_{\mathrm{CH_4}}$ in side-by-side measurements at Caltech and Harvard. Both Harvard EM27/SUNs capture these fluctuations simultaneously, which represent geophysical phenomena, not noise as might be assumed.

Overall, this study helps establish a range of new applications for compact solar-viewing Fourier transform spectrometers.