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Quantifying Methane Fluxes Simply and Accurately: The Tracer Dilution Method

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Methane is an important atmospheric constituent with a wide variety of sources, both natural and anthropogenic, including wetlands and other water bodies, permafrost, farms, landfills, and areas with significant petrochemical exploration, drilling, transport, or processing, or refining occurs. Despite its importance to the carbon cycle, its significant impact as a greenhouse gas, and its ubiquity in modern life as a source of energy, its sources and sinks in marine and terrestrial ecosystems are only poorly understood. This is largely because high quality, quantitative measurements of methane fluxes in these different environments have not been available, due both to the lack of robust field-deployable instrumentation as well as to the fact that most significant sources of methane extend over large areas (from 10's to 1,000,000's of square meters) and are heterogeneous emitters – i.e., the methane is not emitted evenly over the area in question. Quantifying the total methane emissions from such sources becomes a tremendous challenge, compounded by the fact that atmospheric transport from emission point to detection point can be highly variable.

In this presentation we describe a robust, accurate, and easy-to-deploy technique called the tracer dilution method, in which a known gas (such as acetylene, nitrous oxide, or sulfur hexafluoride) is released in the same vicinity of the methane emissions. Measurements of methane and the tracer gas are then made downwind of the release point, in the so-called far-field, where the area of methane emissions cannot be distinguished from a point source (i.e., the two gas plumes are well-mixed). In this regime, the methane emissions are given by the ratio of the two measured concentrations, multiplied by the known tracer emission rate. The challenges associated with atmospheric variability and heterogeneous methane emissions are handled automatically by the transport and dispersion of the tracer.

We present detailed methane flux results from four different landfills in the United States, using a commercially available Cavity Ringdown Spectroscopy (CRDS) dual-species (methane – acetylene) analyzer. This instrument, because of its high precision, mobility, and ease-of-use, enables quantification of the methane flux from a variety of extended area sources. The instrument was operated off of batteries and was mounted in a four-wheel drive vehicle. A high-precision GPS and two-dimensional self-aligning anemometer were integrated directly with the instrument. Concentration data on methane and acetylene were collected every second, and, together with the wind and GPS data, were processed to provide quantitative measurements of total methane fluxes, on a time scale of just minutes.

The landfills studied varied widely in their size, location, topography, and physical access. Data were collected using three variants of the method: the Mobile Transect Method, in which the dual-species analyzer is transported rapidly through the plumes in the far-field; the Stationary Plume Method, in which the analyzer is situated in a fixed location downwind of the release point; and a new method called the Mid-Field Stationary Method, in which the instrument is located at a fixed location at a closer distance than the true far-field, where the plume overlap is not ideal. The resulting methane fluxes varied over a wide range of values, from just a few kg methane / minute, to over 20 kg methane / minute. Finally, we describe how these methods can be used to quantify methane emissions from other natural and anthropogenic extended-area sources, such as wetlands.