Contents lists available at ScienceDirect

Remote Sensing of Environment

journal homepage: <www.elsevier.com/locate/rse>

Airborne visualization and quantification of discrete methane sources in the environment

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article info abstract

Article history: Received 12 May 2014 Received in revised form 1 July 2014 Accepted 6 August 2014 Available online xxxx

Keywords: Methane emissions TIR hyperspectral imaging

1. Introduction

1.1. Methane and climate change

Current climate trends augur for considerable increases in atmospheric methane abundance from anthropogenic sources, such as fossil fuel production, and natural sources such as the release of gas sequestered in and below permafrost due to warming trends [\(Henriet &](#page--1-0) [Mienert, 1998; Zhuang et al., 2007](#page--1-0)). Although carbon dioxide is significantly more abundant than methane and has a considerably longer atmospheric residence time, the relative potency of methane as a greenhouse agent is approximately 100 times that of carbon dioxide on decadal timescales [\(Solomon et al., 2007\)](#page--1-0). Because of its short lifetime, and the current dominance of anthropogenic sources, methane has greater potential return on investment for regulatory efforts to mitigate greenhouse warming [\(Shindell et al., 2012](#page--1-0)). Despite the likelihood of progressively increasing future natural emissions due to global warming feedbacks ([Rigby et al., 2008](#page--1-0)), and anthropogenic activities [\(Kirschke et al., 2013; Wunch, Wennberg, Toon, Keppel-Aleks, &](#page--1-0) [Yavin, 2009\)](#page--1-0), large uncertainties exist in current estimates from many sources with greater uncertainty in future trends. These uncertainties fuel the current critical need to develop robust methane measurement methodologies from diurnal to decadal time scales and decameter to continental size geographic scales on a global basis.

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Airborne thermal-infrared (TIR) imaging spectrometry techniques have been used to detect and track methane and other gaseous emissions from a variety of discrete sources in diverse environmental settings, and to enable estimation of the strength of each plume. The high spatial resolution (1–2 m) permits attribution of chemical plumes to their source, while the moderate spectral resolution (44 nm across the 7.5–13.5 μm TIR band) enables identification and quantification of the gaseous plume constituents, even when one is present in considerably greater concentration than the others. Raw imagery was quantitatively analyzed using matched filtering and adaptive coherence techniques. Experiments under controlled conditions demonstrated successful detection of methane point sources at release rates as low as 2.2 kg/h $\left(\sim 1 \text{ dm}^3\right)$ at NTP).

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1.2. Methane regional and global measurements

Although the ground-based monitoring network has grown rapidly in recent years, many of the Earth's important methane sources including wetlands, permafrost, husbandry, fossil fuel industrial (FFI) emissions, and megacity urban centers, lie logistically and politically beyond the reach of surface networks as well as airborne monitoring. Atmospheric inverse modeling based on observed methane concentration is commonly used to infer methane source strengths (e.g., [Jeong](#page--1-0) [et al., 2012; Kirschke et al., 2013; Kort et al., 2008; Zhao et al., 2009](#page--1-0)). However, this approach relies on the availability of sufficient proximal measurements to achieve the desired level of fidelity. Consequently, inversion model-derived emission estimates tend to be prone to large uncertainties. Moreover, the significant discrepancies between inventorybased emission estimates and more rigorous local-scale measurements [\(Karion et al., 2013; Miller et al., 2013](#page--1-0)) suggest that reliance on reported methane emissions alone is insufficient. In particular, recent studies indicate that natural gas and oil system emissions in the U.S. are severely underestimated by approximately a factor of two [\(Brandt et al., 2014;](#page--1-0) [Miller et al., 2013; Pétron et al., 2014\)](#page--1-0), yet these underestimates were not identified previously in inversion models. This highlights why inversion model a priori initialization requires higher data density than is currently available.

Given adequate meteorological information and data, source strength can be derived from in situ surface and airborne measurements. For example, inverse plume modeling and mass balance approaches have been applied to quantify source strength from

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remotely-sensed column data and in situ airborne methane measurements ([Krings et al., 2011, 2013](#page--1-0)). This approach involves optimally fitting the observed spatial distribution to a Gaussian plume model based on measured or inferred meteorological information, primarily winds but also boundary layer depth, with the caveat that the plume is well described as Gaussian and exhibits steady emissions and transport. Remote spectral imaging has significant potential to improve on in situ measurements by providing an altitude-integrated measurement and a denser dataset.

1.3. Monitoring methane sources

Important methane contributions to global budgets arise from both anthropogenic and natural sources [\(Stocker et al., 2013](#page--1-0)). With respect to evaluating and monitoring these sources by various approaches – including remote sensing – strong localized sources such as FFI facilities can be characterized by anomaly analysis and plume inversion modeling [\(Krings et al., 2011, 2013](#page--1-0)). For strong sources, this can involve stationary or mobile data collection. Station data collection involves using wind direction and concentration to derive emissions from relatively nearby sources [\(Bradley, Leifer, & Roberts, 2010; LaFranchi et al.,](#page--1-0) [2013](#page--1-0)). However, this requires an a priori emission distribution, knowledge of the prevailing meteorology over the area of interest, and assumes steady-state emissions to de-convolve transport variability from source emission variability. Uncertainty therefore increases with distance from source(s), lending greater uncertainty to interpretations. Multiple stations allow significant improvement through triangulation, but in this case station placement is key. Mobile measurements typically involve aircraft in situ, surface in situ, or airborne remote sensing instruments and provide significant advantages over stationary measurements. Specifically, they can collect "snapshot" data on a sufficiently rapid time scale to ostensibly eliminate the temporal component from transport and emissions interpretation. In addition, they can characterize meteorology over the area of interest accounting for phenomena such as wind veering due, for example, to topography, differential heating, etc.

Airborne in situ sampling missions have limitations due to the potential for sampling artifacts related to where sampling occurs in relationship to the boundary layer (e.g., [Gentner et al., 2014](#page--1-0)) and source(s), which may be transient. Air sampling above the boundary layer generally is unrepresentative of surface sources, while flight restrictions over populated and other restricted areas, along with safety concerns, often prevent airborne collection within the boundary layer. For airborne in situ measurements collected at altitude, modeling may enable de-convolving the sources assuming adequate meteorology information for the boundary layer is available, but as above, a priori initialization can bias model-derived emission strengths.

1.4. Methane remote sensing of facility emissions

Only satellite observations have the coverage to monitor methane emissions on a global scale at synoptic timescales. However, current satellite systems have coarse spatial resolution or do not characterize lower atmospheric methane ([Buchwitz et al., 2013; Schepers et al.,](#page--1-0) [2012; Schneising et al., 2009; Stephan et al., 2011; Worden et al.,](#page--1-0) [2012; Xiong et al., 2010, 2013\)](#page--1-0). Resolving and characterizing the localscale variability of methane for compact source emissions requires decameter to sub-kilometer scale measurements. Currently, this capability lies beyond planned satellite observation missions; however, airborne measurements can address this need.

A key consideration is the calibration and validation of derived methane source strengths from satellite data using airborne and surface data for real sources. This is a primary objective of the $CO₂$ and Methane EXperiment (COMEX) field campaign, scheduled for summer 2014 [\(Leifer, Tratt, Egland, & Melton, 2013\)](#page--1-0). COMEX combines airborne, surface, thermal-infrared (TIR), and shortwave-infrared (SWIR) imaging spectroscopy with high resolution, non-imaging spectroscopy to validate plume inverse-model derivation of greenhouse gas source emissions [\(Bergamaschi et al., 2009; Monteil et al., 2013](#page--1-0)).

1.5. Study motivation

In this work we demonstrate the utility of airborne TIR hyperspectral imaging spectroscopy as a new data approach for inverse source strength estimation. The ability of airborne imaging spectrometry to characterize sources lies in its ability to collect snapshot column abundance data and has advantages for mass balance assessment with respect to strong localized sources. Given methane's large source uncertainties and importance to greenhouse warming, our focus emphasizes detection and mapping of strong methane sources. However, airborne TIR imaging spectroscopy also can detect and characterize non-methane hydrocarbons and other organic/inorganic trace gases, whether in isolation or as components of an admixed plume.

1.6. Strong anthropogenic methane emission sources

Strong sources produce plumes amenable to anomaly detection, and inverse modeling for emission strength derivation. Some important strong methane sources relate to landfills, FFI, and enteric fermentation [\(Kirschke et al., 2013](#page--1-0)), releasing 28%, 24%, and 27% respectively of the estimated 330 Tg yr^{-1} anthropogenic emissions based on a bottomup approach [\(Kirschke et al., 2013\)](#page--1-0). Each of these is discussed below.

1.6.1. FFI

FFI sources incorporate emissions from production, refining, and transportation. These sources are variously regarded as the most important ([Brandt et al., 2014\)](#page--1-0) or second most important ([EPA, 2013](#page--1-0)) component of the global methane budget. By definition, production introduces pipes and other infrastructure into a natural geological system characterized by migration, typically along faults and fractures ([Abrams,](#page--1-0) [2005](#page--1-0)). This process is termed seepage and can be natural, anthropogenic, or a mixture of both. Thus, production-related emissions can occur by infrastructure or through natural pre-existing migration pathways or potentially by opening new geologic migration pathways. Production emissions can be notoriously difficult to assess for bottom-up emission estimates as they often are transient [\(Chambers, Strosher, Wootton,](#page--1-0) [Moncrieff, & McCready, 2008](#page--1-0)). Although new production tends to use natural pressure, long-exploited reservoirs often require repressurization both to prevent reservoir collapse and damage as fluids are removed (creating unsupported open spaces), and to enhance migration to wells. The latter is termed enhanced oil recovery (EOR) and can involve re-injecting a gas such as nitrogen, a technique used at the Elk Hills oil field in the southern San Joaquin Valley (SJV), California [\(Alvarado & Manrique, 2010](#page--1-0)). EOR also can involve steam or other fluid re-injection as is practiced at the Kern River oil field, also in the southern SJV.

After FFI production, hydrocarbons are transported to refineries and then distributed to residential and commercial consumers through extensive networks of pipelines, which can produce significant emissions. For example, studies in U.S. and eastern German cities have determined that distribution leakage was the main source of urban methane [\(Shorter et al., 1996; Wennberg et al., 2012](#page--1-0)). Trunk lines can also leak; [Leifer et al. \(2013\)](#page--1-0) identified a large methane plume in an unpopulated woodland area that was interpreted as originating from such a source. Refineries also can produce significant emissions [\(Leifer, Tratt, Egland,](#page--1-0) [& Melton, 2013](#page--1-0)).

Legacy FFI sources have important implications for future emission trends as current FFI sources become depleted and abandoned, yet remain poorly characterized for a number of reasons. Legacy emissions arise from un-extracted hydrocarbons in reservoirs. Reservoir recharge by migration from deeper sources along faults and fractures [\(Leifer,](#page--1-0) [Kamerling, Luyendyk, & Wilson, 2010\)](#page--1-0) then can lead to migration

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