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Retrieval techniques for airborne imaging of methane concentrations using high spatial and moderate spectral resolution: application to AVIRIS

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Abstract

Two quantitative retrieval techniques were evaluated to estimate methane (CH_4) enhancement in concentrated plumes using high spatial and moderate spectral resolution data from the Airborne Visible/Infrared Imaging Spectrometer (AVIRIS). An iterative Maximum a Posteriori Differential Optical Absorption Spectroscopy (IMAP-DOAS) algorithm performed well for an ocean scene containing natural CH_4 emissions from the Coal Oil Point (COP) seep field near Santa Barbara, California. IMAP-DOAS retrieval precision errors are expected to equal between 0.31 to 0.61 ppm CH_4 over the lowest atmospheric layer (height up to 1.04 km), corresponding to about a 30 to 60 ppm error for a 10 m thick plume. However, IMAP-DOAS results for a terrestrial scene were adversely influenced by the underlying landcover. A hybrid approach using Singular Value Decomposition (SVD) was particularly effective for terrestrial surfaces because it could better account for spectral variability in surface reflectance. Using this approach, a CH_4 plume was observed immediately downwind of two hydrocarbon storage tanks at the Inglewood Oil Field in Los Angeles, California, with a maximum near surface enhancement of 8.45 ppm above background. At COP, the distinct plume had a maximum enhancement of 2.85 ppm CH_4 above background and was consistent with known seep locations and local wind direction. A sensitivity analysis also indicates CH_4 sensitivity should be more than doubled for the next generation AVIRIS sensor (AVIRISng) due to improved spectral resolution and sampling. AVIRIS-like sensors offer the potential to better constrain emissions on local and regional scales, including sources of increasing concern like industrial point source emissions and fugitive CH_4 from the oil and gas industry.

1 Introduction

Atmospheric methane (CH_4) is a long-lived greenhouse gas with an instantaneous radiative forcing 21 times greater than carbon dioxide (CO_2) on a per molecule basis

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(IPCC, 2007). In the late preindustrial Holocene (1000 to 1800 A.D.), mean concentrations were 695 ppb (Etheridge et al., 1998) and global concentrations have increased to around 1800 ppb in 2013 (NOAA, 2013). While anthropogenic sources made up an estimated 4 to 34 % of pre-industrial emissions (IPCC, 2007; Houweling et al., 2000), between 60 and 70 % of emissions are presently anthropogenic (Lelieveld et al., 1998). Further, ice core records have indicated CH₄ concentrations are closely tied to atmospheric temperature records, while present-day concentrations have not been observed in the previous 420 000 yr (Wuebbles and Hayhoe, 2002).

While the global CH₄ budget is relatively well constrained ($550 \pm 50 \text{ Tg CH}_4 \text{ yr}^{-1}$), there is considerable uncertainty regarding partitioning between individual natural and anthropogenic source types and locations (IPCC, 2007). Major sources of anthropogenic CH₄ emissions include the energy, industrial, agricultural, and waste management sectors. In the United States, 50 % of anthropogenic CH₄ emissions are from the energy sector, including natural gas and oil systems, coal mining, and stationary/mobile combustion (EPA, 2011). Global fugitive CH₄ emissions from natural gas and oil systems are of increasing concern, estimated at $1354.42 \text{ MMT CO}_2 \text{ E yr}^{-1}$ ($64.50 \text{ Tg CH}_4 \text{ yr}^{-1}$) and expected to increase 35 % by 2020 (EPA, 2006). Recent studies also suggest official inventories are underestimated, for example, top-down estimates indicate fugitive CH₄ emissions are between 2.3 and 7 % of CH₄ produced annually for the Denver–Julesburg Basin, Colorado (Petron et al., 2012). In the Los Angeles Basin, CH₄ emissions appear underestimated (Wunch et al., 2009) and unaccounted sources appear to be fugitive and natural CH₄ emissions (Wennberg et al., 2012).

Significant natural CH₄ sources include wetlands, termites, and geological seeps (IPCC, 2007). Globally, geological seeps are highly uncertain but estimated to contribute between 20 to 40 Tg CH₄ yr⁻¹ for terrestrial environments (Etiope et al., 2009) and about 40 Tg CH₄ yr⁻¹ for marine seepage (Kvenvolden and Rogers, 2005). In addition, increased surface and ocean temperatures associated with global warming may increase CH₄ emissions from melting permafrost (Woodwell et al., 1998) and CH₄ hydrate destabilization (Kvenvolden, 1988).

2 Airborne measurements of CH₄

Aircraft measurements of gas concentrations are useful because they offer the potential to measure local/regional variations in gas concentrations and complement ongoing efforts at coarser spatial resolutions, such as spaceborne sensors. These airborne measurements can improve greenhouse gas emissions inventories and offer the potential for detection and monitoring of emissions (NRC, 2010).

Research and commercial aircraft equipped with in situ gas measurement provides some sense of CH₄ variability at local and regional scales (ARCTAS, 2010; Schuck et al., 2012). The nadir-viewing Fourier Transform Spectrometer (FTS) included as part of the Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) (Miller and Dinaro, 2012) and spectrometers like MAMAP (Methane Airborne MAPper) (Gerilowski et al., 2011) also offer the potential to measure local emissions. For example, MAMAP detected elevated CH₄ concentrations from coal mine ventilation shafts near Ibbenbüren, Germany (Krings et al., 2013). However, these non-imaging spectrometers have a small field of view (FOV) and are limited to flying transects across local gas plumes rather than mapping plumes in their entirety.

By combining large image footprints and fine spatial resolution, airborne imaging spectrometers are well suited for mapping local CH₄ plumes. However, increased spatial resolution requires reduced spectral resolution, thereby decreasing detection sensitivity. The Airborne Visible/Infrared Imaging Spectrometer (AVIRIS) has a 34° FOV and measures reflected solar radiance at the nadir viewing geometry across 224 channels between 350 and 2500 nm (Green et al., 1998). Strong CH₄ absorption features present between 2000 and 2500 nm can be observed at a 10 nm spectral sampling and Full Width Half Maximum (FWHM). These absorptions are clearly shown in Fig. 1 by transmittance spectra calculated for CH₄ using Modtran 5.3 (Berk et al., 1989), parameterized for a mid-latitude summer model atmosphere and nadir-looking sensor at 8.9 km altitude. High resolution transmittance is shown in red for Fig. 1a and convolved

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to AVIRIS wavelengths in Fig. 1b, while water vapour (H₂O) transmittance has been included in blue to indicate spectral overlap with CH₄.

These shortwave infrared (SWIR) absorptions have permitted mapping of concentrated gas plumes in both marine and terrestrial environments using AVIRIS. For bright sun-glint scenes at the Coal Oil Point (COP) marine seep field in the Santa Barbara Channel, California, Roberts et al. (2010) developed a spectral residual approach between 2000 and 2500 nm and Bradley et al. (2011) a band ratio technique using the 2298 nm CH₄ absorption band and 2058 nm carbon dioxide (CO₂) absorption band. However, these techniques are not suited for terrestrial locations that have lower albedos and have spectral structure in the SWIR. A Cluster-Tuned Matched Filter (CTMF) technique is capable of mapping CH₄ plumes from marine and terrestrial sources (Thorpe et al., 2013) as well as CO₂ from power plants (Dennison et al., 2013), however, this method does not directly quantify gas concentrations.

The logical next step is to focus on quantification and uncertainty estimation using techniques originally developed for satellite sensors such as Differential Optical Absorption Spectroscopy (DOAS) (Platt, 1994). In this study, an Iterative Maximum a Posteriori Differential Optical Absorption Spectroscopy (IMAP-DOAS) (Frankenberg et al., 2005c) algorithm was adapted for gas detection in AVIRIS imagery. In addition, a hybrid approach using Singular Value Decomposition (SVD) and IMAP-DOAS was also developed as a complementary method of quantifying gas concentrations within complex AVIRIS scenes.

3 Basic principles of IMAP-DOAS

Classical DOAS (Platt, 1994) is based on the Lambert–Beer law and describes the relationship between incident intensity for the vertical column ($I_0(\nu)$) and measured intensity ($I(\nu)$) after passing through a light path (ds) containing multiple absorbers:

$$I(\nu) = I_0(\nu) \cdot \exp\left(-\int \sigma(\nu, p, T) c(s) ds\right). \quad (1)$$

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Each absorber has an associated absorption cross section (σ) and number concentration of the absorber ($c(s)$, molecules m^{-3}). Equation (1) is wavelength dependent and the absorption cross section varies with temperature (T) and pressure (p). For ideal instruments (or weak absorbers), Eq. (1) can be linearized with respect to slant column density S :

$$\tau = \ln \left(\frac{I_0(\nu)}{I(\nu)} \right) \approx \sigma(\nu, \bar{p}, \bar{T}) \cdot \int c(s) ds = \sigma(\nu, p, T) \cdot S, \quad (2)$$

where measured differential optical density (τ) is proportional to the product of the absorption cross section and the retrieved S , the path integral of the concentration of the absorber along the lightpath. S is related to the vertical column density (V), the integral of the concentration along the vertical from the surface to the top of atmosphere, by way of the airmass factor (A), where $A = S/V$. In the SWIR, scattering in the atmosphere is generally low and for our applications, the impact of scattering is far lower than the retrieval precision error. Thus, it can be neglected and $A = 1/\cos(\text{SZA}) + 1/\cos(\text{LZA})$, where SZA is the solar zenith angle and LZA is the line of sight zenith angle.

For a single absorber measured with a moderate spectral resolution and ignoring scattering, a theoretical slant optical density ($\tau_\lambda^{\text{meas}}$) can be calculated as follows

$$\tau_\lambda^{\text{meas}}(x) = -\ln \left(\left\langle \exp \left(-x \cdot A \cdot \tau_\lambda^{\text{ref}} \right) \right\rangle \right), \quad (3)$$

where the reference vertical optical density ($\tau_\lambda^{\text{ref}}$) is scaled by both the airmass factor (A) as well as a retrieved scaling factor (x) and $\langle \cdot \rangle$ denotes convolution with the instrument function. In addition to scaling $\tau_\lambda^{\text{meas}}$, x can be used to estimate gas concentrations relative to those concentrations present within the reference atmosphere.

However, moderate spectral resolution spectrometers cannot fully resolve individual absorption lines and must convolve light using an instrument lineshape (ILS) function wider than individual absorption lines. This results in a non-linear relationship between the measured differential optical density (τ) and the retrieved slant column density of

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the absorber (S) shown in Eq. (2) (Frankenberg et al., 2005c). Further, optical densities can be large in the SWIR, especially in the 2300 nm region with its strong H_2O and CH_4 line strengths. These factors invalidate Eq. (2) and cause classical DOAS algorithms to fail for moderate spectral resolution spectrometers and strong absorbers.

To address the strong sensitivity of the shape of spectral absorption lines to temperature and pressure as well as unresolved absorption lines (Platt and Stutz, 2008), the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) retrieval algorithm was developed (Buchwitz et al., 2000). WFM-DOAS introduced weighting functions to linearize the problem about a linearization point in the expected slant column density using vertical profiles of all absorbers as well as pressure and temperature profiles. It has been used to estimate column amounts of CO (carbon monoxide), CO_2 , and CH_4 using Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) data, which has a spectral sampling interval between 0.2 and 1.5 nm (Buchwitz et al., 2005). A modified WFM-DOAS algorithm is used with the airborne MAMAP, which has a SWIR grating spectrometer for measuring CH_4 and CO_2 absorptions between 1590 and 1690 nm with a 0.82 nm FWHM (Krings et al., 2011). In addition to detecting elevated CH_4 concentrations from coal mines (Krings et al., 2013), MAMAP has been used to measure both CH_4 and CO_2 emissions from power plants (Gerilowski et al., 2011).

Frankenberg et al. (2005c) developed the IMAP-DOAS algorithm, which uses optimal estimation theory to adjust the slant column densities of multiple gasses until total optical density fits the observed measurement. IMAP-DOAS considers the shape of the spectral absorption lines as they vary with temperature and pressure in multiple atmospheric layers and convolves absorption lines using the instrument lineshape function. This technique is based on a simple non-scattering radiative transfer scheme, which allows very fast retrievals. For the 2300 nm range, where Rayleigh scattering can be ignored and aerosol optical depths are low, this assumption is valid given errors induced by neglected scattering in AVIRIS scene are typically much lower (0 to 2 %) than preci-

sion errors in retrieved column estimates ($> 3\%$). Additional details of the IMAP-DOAS algorithm and retrieval method are presented in Sect. 5.

While IMAP-DOAS has been used with SCIAMACHY data to estimate global column-averaged mixing ratios for CH_4 (Frankenberg et al., 2005a, 2011) and CO (Frankenberg et al., 2005b), this study is the first to use aircraft measurements. Moderate resolution spectrometers like AVIRIS require large fitting windows and disentangling surface spectral features from atmospheric absorptions becomes more complicated using fitting routines such as WFM-DOAS and IMAP-DOAS. High resolution spectrometers can circumvent this problem since atmospheric absorption lines are narrow and surface properties, which vary on a scale greater than 5 to 10 nm, can be fitted using polynomial functions. For the 10 nm spectral sampling and FWHM of AVIRIS, distinguishing surface features from atmospheric absorptions will be more difficult. Therefore, we developed an alternative hybrid approach using both IMAP-DOAS and SVD of surface reflectance properties at background CH_4 concentrations.

4 Study sites and AVIRIS data

Two AVIRIS scenes were used in this study, both acquired in California in 2008. The first scene was acquired over the COP marine seep field near Santa Barbara from an 8.9 km altitude, resulting in an image swath of ~ 5.4 km and a ground instantaneous field of view (IFOV) of ~ 7.5 m. The scene was acquired on 19 June 2008 at approximately 19:55 UTC with a 11.4° solar zenith resulting in high sun-glint. COP is one of the largest natural seeps with total atmospheric CH_4 emissions estimated at $100\,000\text{ m}^3\text{ day}^{-1}$ ($0.024\text{ Tg CH}_4\text{ yr}^{-1}$) (Hornafius et al., 1999). A 308 by 191 pixel image subset was used for the IMAP-DOAS and SVD algorithms, covering 3.31 km^2 centered on the COP seep field ($34^\circ 23' 46.59''\text{ N}$, $119^\circ 52' 4.47''\text{ W}$).

The second scene covered the Inglewood Oil Field, located in Los Angeles in an area that has active oil and gas extraction (DOGGR, 2010). The AVIRIS scene was acquired at approximately 20:12 UTC on 18 September 2008 at 4.0 km altitude, result-

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ing in a swath width of ~ 2.7 km, ground IFOV of ~ 3 m, and a 38.1° solar zenith. For this scene, a 161 by 172 pixel image subset (0.25 km^2 centered at $33^\circ 59' 28.68''$ N, $118^\circ 21' 34.59''$ W) was selected because it contains a CH_4 plume detected using a CTMF technique, with hydrocarbon storage tanks as a probable emission source (Thorpe et al., 2013).

5 IMAP-DOAS retrieval method

The IMAP-DOAS retrieval relies on layer optical properties of absorbing species calculated for a realistic temperature/pressure and trace gas concentration profile for a given location. In addition, instrument lineshape and flight parameters are used with geometric radiative transfer calculations to simulate at-sensor radiances and Jacobians with respect to trace gas abundances for each atmospheric layer. In the following, we describe input parameters and additional details of the IMAP-DOAS retrieval.

5.1 IMAP-DOAS input parameters

For the two 2008 AVIRIS scenes, temperature, pressure, and H_2O volume mixing ratio (VMR) profiles acquired from the National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis project were extracted for the appropriate date and time for either location (Kalnay et al., 1996). Prior profiles of CH_4 and N_2O are based on the US standard atmosphere obtained from the radiative transfer models LOWTRAN/MODTRAN (Kneizys et al., 1996). These profiles were scaled to reflect the VMR for CH_4 and N_2O using the 2008 mean VMR provided from the NOAA Mauna Loa station, United States (NOAA, 2013). For both gasses, the percent increase of the 2008 mean VMR compared to the US standard atmosphere at 0 km altitude was calculated and used to update the VMR up to 25 km altitude. Finally, we computed vertical optical depths for 10 atmospheric layers at 100 mb intervals between 0 and 1000 mb.

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For AVIRIS, the strongest CH₄ absorptions occur between 2200 to 2400 nm (Fig. 1). Spectral parameters for CH₄, H₂O, and N₂O were used from the HITRAN database (Rothman et al., 2009). We used a classical Voigt spectral line-shape to calculate CH₄, H₂O, and N₂O vertical optical densities for each of the 10 atmospheric layers.

Given that the two AVIRIS scenes were acquired at different flight altitudes and SZA, geometric air mass factors (AMF) had to be calculated for each of the 10 layers to account for either one (above sensor) or two (below sensor) way transmission through each layer. For example, the COP flight was at 8.9 km altitude with a solar zenith angle of 11.4°, placing the aircraft approximately at the boundary between atmospheric layer 3 and 4 (Fig. 2). In this simplified setup, the AMF for layers 1 to 3 (above the aircraft) is calculated as $1/\cos(11.4^\circ) = 1.02$, while for layers 4 to 10, an AMF of 2.02 ($1/\cos(11.4^\circ) + 1/\cos(0.0^\circ)$) accounts for two way transmission. Similar calculations were performed for the Los Angeles scene, which was acquired with a SZA of 38.1° at 4.0 km altitude placing the aircraft approximately at the boundary between layer 5 and 6.

Additional input parameters for the IMAP-DOAS algorithm are shown in Fig. 3, including the AVIRIS radiance data, spectral sampling of the sensor, signal-to-noise ratio (SNR) estimate, and the full width at half maximum of the instrument line-shape (FWHM = 10.42 nm, assuming a Gaussian line-shape). An average FWHM and SNR was calculated for bands included within the fitting window, while the high resolution solar transmission spectrum was generated using a solar linelist (G. Toon, personal communication, 2013).

The optimal choice of a fitting window for the IMAP-DOAS CH₄ retrievals was determined iteratively. We began using all spectral bands between 2100 and 2500 nm corresponding to strong CH₄ absorptions, but observed strong correlations with surface features. This is likely related to spectrally smooth convolved transmissions from 2200 to 2300 nm and above 2370 nm (Fig. 1b). As we decreased the size of the fitting window to focus on the more high-frequency CH₄ features, the spectral variability associated with AVIRIS bands at either end of the fitting window was reduced and re-

sults improved. The fitting window selected for this study used 9 bands between 2278 and 2358 nm, including three prominent absorption features visible in CH₄ Jacobians shown in Fig. 4a.

5.2 Forward model and optimal estimation

Using 10 atmospheric layers and the gasses CH₄, H₂O, and N₂O results in a state vector with 30 rows (\mathbf{x}_n). In principle, N₂O could be neglected at this spectral resolution but we included it for the sake of completeness. A forward radiative transfer model at high spectral resolution was used to calculate modeled radiance at each wavelength using the equation below

$$F^{\text{hr}}(\mathbf{x}_i) = I_0^{\text{hr}} \cdot \exp\left(-\sum_{n=1}^{30} \mathbf{A}_n \cdot \boldsymbol{\tau}_n^{\text{ref}} \cdot \mathbf{x}_{n,i}\right), \quad (4)$$

where $F^{\text{hr}}(\mathbf{x}_i)$ is the forward modeled radiance at the i th iteration of the state vector, I_0^{hr} is the incident intensity (solar transmission spectrum), \mathbf{A}_n is the AMF for each n layer of each gas (30 rows, specified for each of the 10 layers and repeated for each gas), $\boldsymbol{\tau}_n^{\text{ref}}$ is the reference total optical density for each n layer (the sum of optical densities of CH₄, H₂O, and N₂O), $\mathbf{x}_{n,i}$ is the state vector at the i th iteration, which scales the prior optical densities of CH₄, H₂O, and N₂O in each n layer (30 rows).

The high resolution modeled radiance is then convolved with the ILS and sampled to the center wavelengths of each AVIRIS spectral band. This results in a low resolution modeled radiance at the i th iteration of the state vector ($F^{\text{lr}}(\mathbf{x}_i)$), calculated using a known $\boldsymbol{\tau}_n^{\text{ref}}$ scaled by $\mathbf{x}_{n,i}$.

In addition to the priors for CH₄, H₂O, and N₂O in each n layer (\mathbf{x}_n), the state vector (\mathbf{x}_a) contains the spectral shift as well as a low order polynomial function to account for the broad-band variability in surface albedo (see Frankenberg et al., 2005c).

At each iteration i , a Jacobian Matrix is calculated where each column represents the derivate vector of the sensor radiance with respect to each element of the state

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vector (\mathbf{x}_i).

$$\mathbf{K}_i = \left. \frac{\partial F^{\text{lr}}(\mathbf{x})}{\partial \mathbf{x}} \right|_{\mathbf{x}_i}. \quad (5)$$

The forward model and the Jacobian Matrix can be used to optimize the state vector at the i th iteration as follows (Rodgers, 2000)

$$\mathbf{x}_{i+1} = \mathbf{x}_a + \left(\mathbf{K}_i^T \mathbf{S}_\varepsilon^{-1} \mathbf{K}_i + \mathbf{S}_a^{-1} \right)^{-1} \mathbf{K}_i^T \mathbf{S}_\varepsilon^{-1} \cdot \left[\mathbf{y} - F^{\text{lr}}(\mathbf{x}_i) + \mathbf{K}_i(\mathbf{x}_i - \mathbf{x}_a) \right], \quad (6)$$

where \mathbf{x}_a is the a priori state vector (30 rows), \mathbf{x}_i is the state vector at the i th iteration (30 rows), \mathbf{S}_ε is the error covariance matrix, \mathbf{S}_a is the a priori covariance matrix, \mathbf{y} is the measured AVIRIS radiance, $F^{\text{lr}}(\mathbf{x}_i)$ is the forward model evaluated at \mathbf{x}_i , \mathbf{K}_i is the Jacobian of the forward model at \mathbf{x}_i .

The a priori state vector was set to 1 for each gas at each layer, while the a priori covariance matrix was set to constrain the fit to the lowest atmospheric layer (height up to 1.04 km) where high variance is expected. To achieve this, very tight prior covariances were set for all atmospheric layers except the lowermost one, which is basically unconstrained. This assumption is reasonable given that the COP and Inglewood scenes contain CH_4 emission from ground sources that are not expected to extend above this atmospheric layer. CH_4 concentrations were calculated by multiplying the CH_4 state vector at the last iteration (CH_4 scaling factor) by the VMR for the lowest layer of the reference atmosphere (Fig. 2).

6 Basic principles of SVD

SVD transforms a large number of potentially correlated vectors into a smaller set of uncorrelated (orthogonal) vectors, denoted as singular vectors (Press et al., 2007;

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Rodgers, 2000). It is closely related to Principal Component Analysis (PCA) and offers the potential for reduced computation time by efficiently summarizing high dimensional data. It has been used in a number of remote sensing applications, including cloud detection using the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) (Hurley et al., 2009), retrieving aerosol optical densities of mineral dust using the Infrared Atmospheric Sounding Interferometer (IASI), and retrieval of terrestrial chlorophyll fluorescence using the Fourier Transform Spectrometer (FTS) onboard the Greenhouse gases Observing SATellite (GOSAT) platform (Guanter et al., 2012).

For this study, we constructed an $m \times n$ matrix \mathbf{L} , where m is the number of spectral bands (for the CH_4 fit window) and n is the number of radiance spectra in a specific AVIRIS scene. This can be expressed as

$$\mathbf{L} = \mathbf{U}\mathbf{\Lambda}\mathbf{V}^T, \quad (7)$$

where the $m \times m$ matrix \mathbf{U} contains the left singular vectors and the $n \times n$ matrix \mathbf{V} contains the right singular vectors in their respective columns. $\mathbf{\Lambda}$ is an $m \times n$ rectangular diagonal matrix containing the m singular values of \mathbf{L} on its diagonal. These singular values are essentially eigenvalues that correspond to the m columns of \mathbf{U} , which are analogous to eigenvectors. Each of the n columns of \mathbf{V} is essentially a principal component of the scene, with each successive column capturing increasingly less signal variability. Therefore, \mathbf{L} can be recomposed as a linear combination of singular vectors scaled by the singular values (Murtagh and Heck, 1987).

7 SVD retrieval method

For each AVIRIS image subset, the radiance scene was first standardized by fitting a first order polynomial to each radiance spectrum and dividing it by the polynomial fit. Next, a mean radiance spectrum was calculated from the standardized data and the IMAP-DOAS retrieval was performed on the mean spectrum to generate the CH_4 Jacobian for the lowest layer (\mathbf{K}_{CH_4}) (Fig. 5). This standardization was performed to

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ensure that the computed CH_4 Jacobian is representative for all pixels; without it, calculations of Jacobians for each continuum level would be required. As an alternative to standardization, a SVD in log-space could be considered since optical depths are linear with respect to changing concentrations in the vicinity of the linearization point.

Using Eq. (7), the SVD was performed on each image subset using the standardized radiance ($m \times n$ matrix \mathbf{L} , where m is the number of spectral bands and n is the number of radiance spectra). Due to computing limitations, the economy version of the SVD was calculated using MATLAB (Mathworks, Natick, Massachusetts). This resulted in \mathbf{U}_{econ} maintaining a dimension of $m \times m$ (left singular vectors in m columns), but reduced matrix dimensions for \mathbf{V}_{econ} and Λ_{econ} ($n \times m$ and $m \times m$ respectively).

The first c columns of \mathbf{U}_{econ} ($\mathbf{U}_{\text{select}}$, an $m \times c$ matrix) and the CH_4 Jacobian (\mathbf{K}_{CH_4} , an $m \times 1$ matrix) are concatenated to generate a matrix \mathbf{J} (dimensions of $m \times c + 1$). The basic principle is to reflect the general variability in spectral radiances by a linear combination of the first c eigenvectors and the CH_4 Jacobian, which relates to deviations from background concentrations since the background radiance is already modeled using the linear combination of eigenvectors. A similar technique was used to retrieve terrestrial chlorophyll fluorescence using the FTS onboard GOSAT (Guanter et al., 2012). The linear combination of eigenvectors is an empirical way to compute the forward model radiance, which can include many detector and surface albedo features that the IMAP-DOAS approach cannot easily handle.

Using linear least squares, we can now find a vector \mathbf{W} that minimizes the cost function involving the measured radiance spectra \mathbf{y}_i (where i refers to a specific spectrum in the scene):

$$\|\mathbf{y}_i - \mathbf{J}\mathbf{W}_i\|^2. \quad (8)$$

\mathbf{W}_i represents the contribution of each column of \mathbf{J} to the measured radiance. The modeled radiance \mathbf{F}_i can be calculated by multiplying \mathbf{J} by \mathbf{W}_i :

$$\mathbf{F}_i = \mathbf{J}\mathbf{W}_i, \quad (9)$$

resulting in a modeled radiance that can be compared to the measured radiance for each spectrum.

For each image pixel, the value in W_i that corresponds to the CH_4 Jacobian is denoted as RCH_4 . RCH_4 indicates how much of the observed radiance for each spectrum can be associated with the CH_4 Jacobian (i.e. changes in absorptions due to CH_4) and can be used to both estimate CH_4 concentrations as well as its uncertainties. Similar to the IMAP-DOAS approach, RCH_4 for each pixel is multiplied by the VMR for the lowest layer of the reference atmosphere and results in an estimated CH_4 concentration in ppm above/below the average.

The same 9 bands between 2278 and 2358 nm that made up the IMAP-DOAS retrieval window were initially used for the hybrid SVD approach. In an iterative process, additional bands between 2218 and 2457 nm were included to better account for high frequency variation present in the scenes. A portion of the scene was selected for a homogeneous landcover and the standard deviation of the RCH_4 results for different fitting windows was calculated. A 16 band fitting window (2278 to 2428 nm) was selected because it produced the lowest standard deviation in RCH_4 and thereby minimized noise in results.

Using these 16 bands, the hybrid SVD retrieval was performed iteratively by increasing the c columns of \mathbf{U}_{econ} used to generate $\mathbf{U}_{\text{select}}$. This resulted in 16 SVD retrievals, which were assessed by minimizing the standard deviation of the RCH_4 results for the portion of the scene selected to represent homogeneous landcover. This technique was used to determine the optimal number of columns of \mathbf{U}_{econ} to use with the SVD retrieval for the COP and Inglewood scenes.

8 Results for IMAP-DOAS sensitivity study

To investigate the expected IMAP-DOAS retrieval errors for the 9 band fitting window between 2278 and 2358 nm, the covariance \hat{S} was calculated using the following

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equation

$$\hat{\mathbf{S}} = \left(\mathbf{K}^T \mathbf{S}_e^{-1} \mathbf{K} + \mathbf{S}_a^{-1} \right)^{-1}, \quad (10)$$

where the diagonal of $\hat{\mathbf{S}}$ corresponds to the covariance associated with CH₄, H₂O, and N₂O at each of the 10 atmospheric layers. \mathbf{S}_e is the error covariance matrix, a diagonal matrix representing expected errors resulting from shot-noise and dark current that is calculated using the SNR for the AVIRIS sensor.

The precision error of the IMAP-DOAS retrieval algorithm is calculated by multiplying the square root of the corresponding diagonal entry of $\hat{\mathbf{S}}$ (the standard deviation of the CH₄ fit factor) by 1.78 ppm CH₄, the 2008 mean VMR provided from the NOAA Mauna Loa station, United States (NOAA, 2013). These errors were calculated for a number of hypothetical sensors with varying spectral sampling intervals (SSI) and FWHM across a range of SNR (Fig. 6). As expected, the IMAP-DOAS error decreases as SNR increases and as the sensor SSI and FWHM become finer. The black line (10 nm SSI and FWHM) approximates the AVIRIS sensor and the SNR for bands used in the IMAP-DOAS retrieval was conservatively estimated between 100 and 200 using an AVIRIS instrument model for low albedo surfaces (R. Green, personal communication, 2013). Using scene parameters similar to the COP flight (8.9 km altitude, 11.4° solar zenith), this corresponds to an error of between 0.31 to 0.61 ppm CH₄ over the lowest atmospheric layer (up to 1.04 km) shown in Fig. 2a. Given that about 10 % of the total column is within the lowest layer, this roughly corresponds to an error of 30 to 60 ppb in column-averaged CH₄ over the total atmospheric column.

9 Results for IMAP-DOAS

9.1 COP

For the COP subset shown in Fig. 7a, measured radiance for the first band of the IMAP-DOAS retrieval window at 2278 nm had a maximum of 6.436 (sensor saturation



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tion), minimum of 0.1158, and mean of $2.0516 \text{ uWcm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$. Sonar return contours of subsurface CH_4 bubble plumes are overlain and correspond to known seep locations (Leifer et al., 2010). In Fig. 7b, the CH_4 scaling factor is shown for the lowest atmospheric layer (height up to 1.04 km) and a CH_4 enhancement is clearly visible consistent with emission from seep locations and the 2.3 ms^{-1} southwesterly wind measured at the nearby West Campus Station. The standard deviation of the residual (the difference between measured and modeled radiance) was also calculated to evaluate the ability of IMAP-DOAS to model radiance. This result is shown in Fig. 7c and has a similar visual appearance to Fig. 7a, indicating a strong albedo influence.

CH_4 concentrations were calculated by multiplying the retrieved CH_4 scaling factor by the VMR for the lowest atmospheric layer (1.78 ppm CH_4). In Fig. 7d, ppm CH_4 for the lowest layer is shown (subcolumn XCH_4), excluding 740 bright pixels (greater than $5 \text{ uWcm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$ in the fitting window) associated with high standard deviation of the residuals. These results indicate enhancements in the lowest layer up to 2.5 times concentrations present in the reference atmosphere, equivalent to 4.46 ppm CH_4 averaged across the distance from the ocean surface to 1.04 km. However, there appears to be a positive bias in these results given concentrations for locations upwind of the plume appear higher than the expected background concentration of 1.78 ppm. Therefore, the subcolumn XCH_4 results appear overestimated. This observed bias will be further addressed in Sect. 11.

In Fig. 7, location L1 and L2 correspond to the measured and modeled radiance plotted in Fig. 8. At location L1 (Fig. 8a), the measured radiance (black) is nearly horizontal for wavelengths between 2278 and 2328 nm, indicating radiance sensor saturation due to high sun-glint. This causes considerable disagreement with the modeled radiance (red) as indicated by the residual radiance shown in the bottom plot; this pixel was excluded from the results shown in Fig. 7d. For Fig. 8b (location L2), the radiance is considerably lower and there is better agreement between measured and modeled radiance, resulting in a retrieved concentration of 2.18 ppm CH_4 for this pixel. This radiance was

detrended in Fig. 8c and the CH₄ Jacobian for the lowest layer is overlain to indicate the location of CH₄ absorptions at 2298, 2318, and 2348 nm.

9.2 Inglewood

The Inglewood subset (Fig. 9a) is highly heterogeneous, with a maximum measured radiance of 0.8033, minimum of 0.0192, and mean of 0.2800 uWcm⁻²sr⁻¹nm⁻¹ at 2278 nm. A road crosses the scene from north to south, separating the Inglewood Oil Field on the left from a residential neighborhood on the right. In this complex urban environment, the low order polynomial in the IMAP-DOAS algorithm is unable to account for some of the high frequency spectral variability that interferes with CH₄ absorptions. Therefore, the CH₄ scaling factor results for the lowest atmospheric layer are heavily influenced by the land surface type (Fig. 9b). For example, the road appears clearly visible and high CH₄ scaling factors occur for individual structures within the neighborhood. Dark spectra also appear to have erroneously high CH₄ scaling factors, including heavily vegetated areas in the northwest and southeast of the scene.

For the lowest atmospheric layer, subcolumn XCH₄ results are shown in Fig. 9d, excluding dark pixels less than 0.1 uWcm⁻²sr⁻¹nm⁻¹ in the fitting window. While background concentrations are expected around 1.78 ppm CH₄, observed background concentrations appear biased upward, between 2 and 3 ppm. Despite the noisy results, a feature of elevated CH₄ is visible in the center of the image with maximum concentrations in excess of 5.5 ppm. This CH₄ plume is consistent with a 2.2 ms⁻¹ southwesterly wind measured nearby at the time of image acquisition (weatherunderground.com, 2012). Using higher resolution Google Earth imagery acquired one year after the AVIRIS flight, two hydrocarbon storage tanks were identified immediately upwind and are the probable emission source (Fig. 9e).

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10 Results for SVD

10.1 COP

While the IMAP-DOAS technique permitted CH₄ retrievals for the more homogeneous marine location, high frequency variation present in the terrestrial example interferes with CH₄ absorptions and makes mapping more challenging. To permit retrievals for terrestrial locations, a hybrid approach using SVD and IMAP-DOAS was used to first account for high frequency variation present in the scene and determine what variance of the standardized radiance resulted from changes in CH₄.

In Fig. 10, all 16 columns of \mathbf{U}_{econ} are shown in addition to the CH₄ Jacobian (\mathbf{K}_{CH_4}). Following the iterative method described in Sect. 7, 4 of the total 16 columns of \mathbf{U}_{econ} were used to generate $\mathbf{U}_{\text{select}}$ and account for over 99.99 % of the variance. Next, $\mathbf{U}_{\text{select}}$ and \mathbf{K}_{CH_4} were concatenated to generate the \mathbf{J} matrix, which is used for modelling radiance (see Eq. 9).

In Fig. 11b the weights (R_{CH_4}) associated with the column of \mathbf{J} that corresponds to the CH₄ Jacobian are shown (see Eq. 9). Within the scene, expected background values are 0 and the distinctive CH₄ plume is similar to the IMAP-DOAS results (Fig. 7b). In Fig. 12d, ppm CH₄ relative to background is shown excluding 323 pixels (0.55 % of total scene) associated with standard deviation of the residuals greater than 0.0075 (Fig. 11c). CH₄ concentrations exceed 3 ppm above background within the plume, gradually decrease downwind, and approach expected background concentrations.

10.2 Inglewood

Using the iterative method described in Sect. 7, 9 columns of \mathbf{U}_{econ} were selected to generate $\mathbf{U}_{\text{select}}$ for the Inglewood scene. The R_{CH_4} results (Fig. 12b) more clearly distinguish the CH₄ plume compared to the IMAP-DOAS results (Fig. 9b), however, the SVD standard deviation of the residuals indicates higher errors for vegetated surfaces (Fig. 12c). Excluding pixels with greater than 0.0075 standard deviation of the resid-

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ual, retrieved concentrations relative to background are shown in Fig. 12d. Expected background concentrations are observed throughout much of the scene and CH₄ concentrations are highest for the western portion of the plume (in excess of 4 ppm above background).

In Fig. 12, location L3 and L4 correspond to the measured and modeled radiance plotted in Fig. 13. At location L3 (Fig. 13a), there is considerable disagreement between the measured (black) and modeled radiance (red) as indicated by the residual. L3 is located in a vegetated region and because the standard deviation of the residual exceeds 0.0075, this pixel was excluded from the results shown in Fig. 12d. In contrast, there is good agreement for L4, which is made up of bare soil with an estimated concentration of 0.38 ppm CH₄ above background (Fig. 13b).

11 Discussion

11.1 Comparison of retrieval results

The IMAP-DOAS and hybrid SVD approach were capable of quantifying CH₄ concentrations from plumes over marine and terrestrial environments. For both techniques, agreement between measured and modeled radiance was poorest at albedo extremes, for example saturated pixels at COP and dark, vegetated surfaces at Inglewood. SVD results indicate near surface enhancements relative to background; absorptions resulting from background CH₄ concentrations in the scene are contained in $\mathbf{U}_{\text{select}}$ and the retrieval used the CH₄ Jacobian from the lowest layer of the atmospheric model. Similarly, the IMAP-DOAS retrieval also provides ppm CH₄ enhancements averaged over the lowest atmospheric layer (up to 1.04 km).

For the IMAP-DOAS results from COP and Inglewood, an average background ppm CH₄ concentration was calculated for the portion of the scene selected to represent homogeneous landcover (see Sect. 7). To account for the observed positive bias in subcolumn XCH₄ (see Sect. 9), this average concentration was subtracted from sub-

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column XCH_4 , resulting in ppm CH_4 relative to background. However, different portions of each scene were excluded from IMAP-DOAS and SVD results due to observed biases. For example, pixels were excluded from IMAP-DOAS results at Inglewood using an albedo threshold (Fig. 9d), while a standard deviation of the residual threshold was applied to SVD results (Fig. 12d). To permit comparison between results, only those pixels not excluded from either the IMAP-DOAS or SVD results are shown in Figs. 14 and 15.

These results were also validated against an independent technique, the Cluster-Tuned Matched Filter (CTMF) that was applied to both scenes (Figs. 14c and 15c). The CTMF uses a gas transmittance spectrum as a target to calculate CTMF scores for each image pixel where scores greater than one indicate significant evidence of the gas signature (Thorpe et al., 2013; Funk et al., 2001). The CTMF does not provide an estimate of gas concentrations, rather it provides an image of gas anomalies that can be evaluated for consistency with probable emissions sources and local wind direction. In contrast, IMAP-DOAS and the hybrid SVD approach provide CH_4 concentrations as well as uncertainty estimates.

At COP, there is good spatial agreement between the observed plumes obtained with the IMAP-DOAS (Fig. 14a), hybrid SVD (Fig. 14b), and CTMF (Fig. 14c) approaches (Thorpe et al., 2013). IMAP-DOAS CH_4 concentrations are generally higher (mean 0.12, standard deviation 0.43 ppm relative to background) than the SVD results (mean -0.01 , standard deviation 0.63 ppm relative to background). The location of an identical transect is shown for the IMAP-DOAS (Fig. 14a, green line), SVD (Fig. 14b, cyan), and CTMF results (Fig. 14c, red). At each point along the transect, an average value was calculated for 21 pixels centered on the transect in the horizontal direction. The average values along the transect are plotted in Fig. 14d and indicate concentrations for IMAP-DOAS (green) are generally higher than for the SVD approach (cyan). Where the transect intersects the plume, there is good agreement in the pronounced peak in values from the three techniques, including CTMF results (red). While the CTMF

technique appears better suited for detecting diffuse portions of the plume (Fig. 14c), it does not provide CH₄ concentrations.

Using the hybrid SVD approach, the maximum observed concentration within the scene was 2.85 ppm CH₄ above background, located at a region of subsurface CH₄ bubble plumes as shown by the sonar return contours (Fig. 11a). Averaged over the lowest atmospheric layer (a distance of 1.04 km), this maximum concentration will increase when scaled for a smaller atmospheric column. For example, concentrations increase to 590 ppm CH₄ above background if all enhancements are within a 5 m atmospheric column. Near surface concentrations are likely much higher; Leifer et al. (2006) measured up to 2×10^4 ppm CH₄ at 5 m height using a flame ion detector.

For Inglewood, the CH₄ plume is clearly visible in IMAP-DOAS (Fig. 15a), hybrid SVD (Fig. 15b), and CTMF (Fig. 15c) results (Thorpe et al., 2013). CH₄ concentrations for IMAP-DOAS are generally higher (mean 0.13 and standard deviation 1.03 ppm relative to background) than the hybrid SVD results (mean -0.04 and standard deviation 1.60 ppm relative to background). Similar to the COP comparison, the location of an identical transect is shown for the IMAP-DOAS, SVD, and CTMF results. An average was calculated at each point along the transect (for 9 pixels centered on the transect in the vertical direction) and plotted in Fig. 15d, indicating good agreement between techniques for the observed CH₄ plume.

For the SVD approach at Inglewood, the maximum within the CH₄ plume was 8.45 ppm above background with concentrations decreasing downwind of the hydrocarbon storage tanks (Fig. 12d). Such enhancements are feasible given tanks represent large emission sources; natural gas storage tanks can emit between 4.3 and 42.0×10^{-4} Gg CH₄ per (10⁶) m³ gas withdrawals per year (IPCC, 2000) and tank venting represented approximately 14.4% (212 Gg CH₄) of the total US CH₄ emissions from petroleum systems in 2009 (EPA, 2011).

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11.2 Potential for AVIRISng and future sensors

While CH₄ retrievals are promising using AVIRIS, the next generation sensor (AVIRISng) will have a 5 nm SSI and FWHM that should significantly improve CH₄ sensitivity. An IMAP-DOAS retrieval error between 0.31 to 0.61 ppm CH₄ over the lowest atmospheric layer (height up to 1.04 km) is expected for an AVIRIS scene acquired at 8.9 km altitude, 11.4° solar zenith, and with a SNR conservatively set between 100 and 200 (Fig. 6, black line). This corresponds to about a 32 to 63 ppm retrieval error for a 10 m thick plume or 322 to 634 ppm for a 1 m thick plume. For a similar AVIRISng scene, the IMAP-DOAS retrieval error would be reduced to between 0.18 to 0.35 ppm over the lowest atmospheric layer for the same range of SNR (Fig. 6, red line). In addition, SNR for AVIRISng should be considerably improved, further reducing retrieval errors.

To further assess this increased sensitivity, CH₄ Jacobians were calculated for AVIRISng and AVIRIS for a 5% CH₄ enhancement over the lowest atmospheric layer. In Fig. 16a, the AVIRIS CH₄ Jacobian (black line) has a $-4.7 \times 10^{-4} \Delta uW cm^{-2} sr^{-1} nm^{-1} / \Delta VMR$ amplitude between a peak at 2310 nm and the CH₄ absorption at 2320 nm. For AVIRISng (red line) this amplitude is $-9.8 \times 10^{-4} \Delta uW cm^{-2} sr^{-1} nm^{-1} / \Delta VMR$, roughly representing a doubling of CH₄ sensitivity compared with AVIRIS. However, additional improvements should result from a greater number of detector pixels and the improved SNR of AVIRISng. Sensors with a finer SSI and FWHM offer the potential for even greater sensitivity, as shown by the grey line in Fig. 16a for a SSI and FWHM of 1 nm and reduced IMAP-DOAS retrieval errors indicated by the grey dashed line in Fig. 6.

12 Conclusions

In this study, two retrieval techniques were used to measure CH₄ enhancements for concentrated plumes over marine and terrestrial locations in AVIRIS data. The IMAP-

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DOAS algorithm performed well for the homogenous ocean scene containing the COP seeps and retrieval errors are estimated between 0.31 to 0.61 ppm CH₄ over the lowest atmospheric layer (height up to 1.04 km). For the Inglewood subset, IMAP-DOAS results became heavily influenced by the underlying landcover, while the hybrid SVD approach was particularly effective given that it could better account for spectrally variable surface reflectance. Using the hybrid SVD approach for the COP and Inglewood plumes, maximum near surface concentrations were 2.85 and 8.45 ppm CH₄ above background respectively. An additional benefit of the hybrid SVD approach is that it requires less than half the computational time of the IMAP-DOAS retrieval.

Given a 5 nm SSI and FWHM, CH₄ sensitivity should be more than doubled for AVIRISng. This might permit CH₄ retrievals for weaker absorption features centered at 1650 nm, as well as CO₂ retrievals for absorptions at 1572, 1602, and 2058 nm. However, both the AVIRIS and AVIRISng sensors were not designed for detecting gas plumes and sensitivity could be dramatically improved using a spectrometer designed exclusively for mapping gas plumes. For example, an imaging spectrometer with 0.05 nm SSI and 0.15 nm FWHM would have an IMAP-DOAS error around 18 times smaller than AVIRIS.

While non-imaging spectrometers such as MAMAP have increased CH₄ sensitivity compared to AVIRIS and AVIRISng, they are limited to flying transects across local gas plumes due to a small field of view. In contrast, airborne imaging spectrometers combine large image footprints and fine spatial resolution necessary to map local CH₄ plumes in their entirety. In this study, the observed COP plume extended more than 1 km, however, the Inglewood plume was much smaller, extending only 0.1 km downwind. Such plumes with a small spatial extent are of increasing concern, including industrial point source emissions, leaking gas pipelines (Murdock et al., 2008), and fugitive CH₄ from the oil and gas industry (Howarth et al., 2011). Therefore, AVIRIS-like sensors offer the potential to better constrain emissions on local and regional scales (NRC, 2010), improve greenhouse gas budgets and partitioning between natural and

anthropogenic sources, as well as complement data provided at coarser spatial resolutions.

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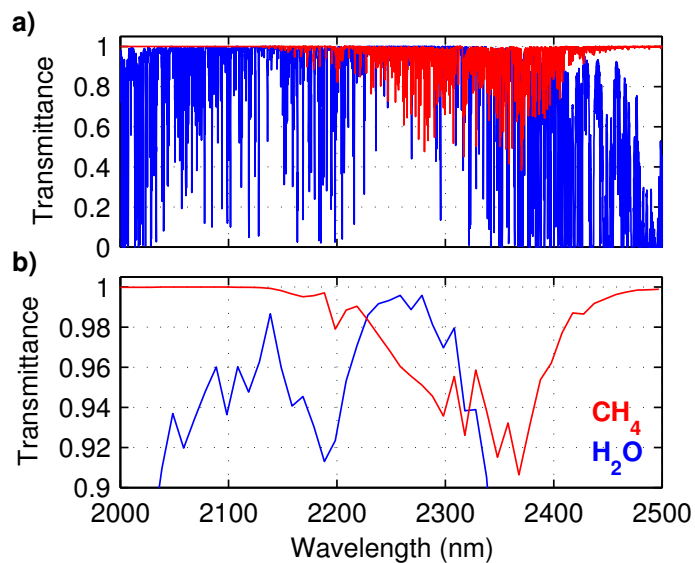


Fig. 1. (a) High resolution CH₄ and H₂O transmittance. (b) Transmittance convolved to the 10 nm AVIRIS spectral sampling interval.

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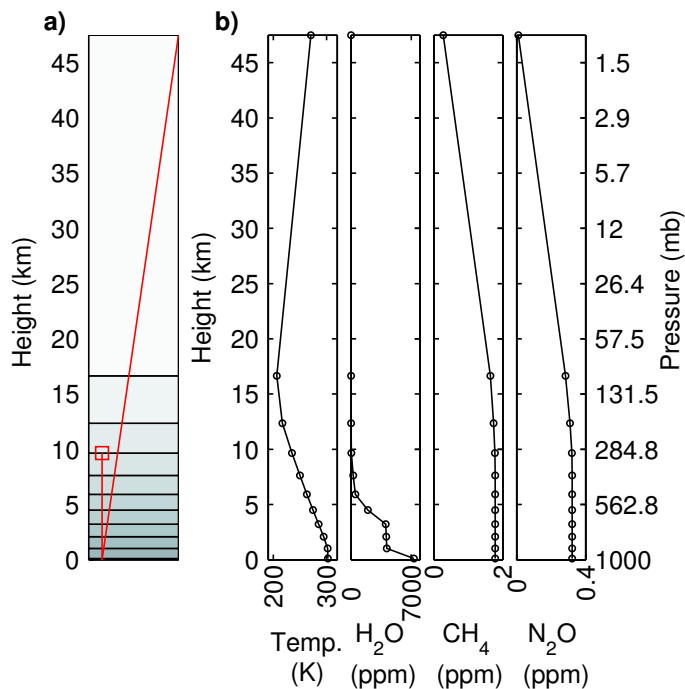


Fig. 2. (a) 10 atmospheric layers were used for retrievals (layer 1 at the top). For the COP scene, the aircraft was placed between layer 3 and 4 (red square). The slant and vertical light paths (red lines) were used to scale optical densities appropriately. (b) Profiles of temperature and VMR of H₂O, CH₄, and N₂O for the boundaries of each layer (black circles).



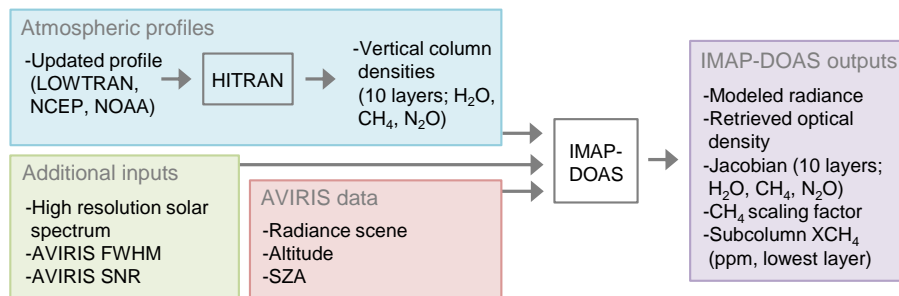


Fig. 3. Processing steps for IMAP-DOAS CH₄ retrieval.

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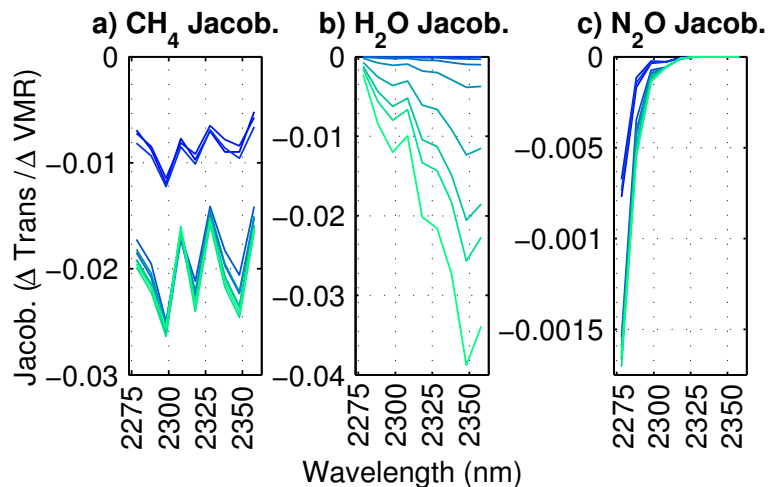


Fig. 4. (a) CH₄ Jacobian for each of the 10 atmospheric layers with colors transitioning from dark blue at the highest layer (layer 1) to light green for the lowest layer (layer 10). The CH₄ Jacobians with smaller magnitudes (dark blue) are for layers above the flight altitude. The same color scheme is used for the H₂O Jacobians (b) and N₂O Jacobians (c).

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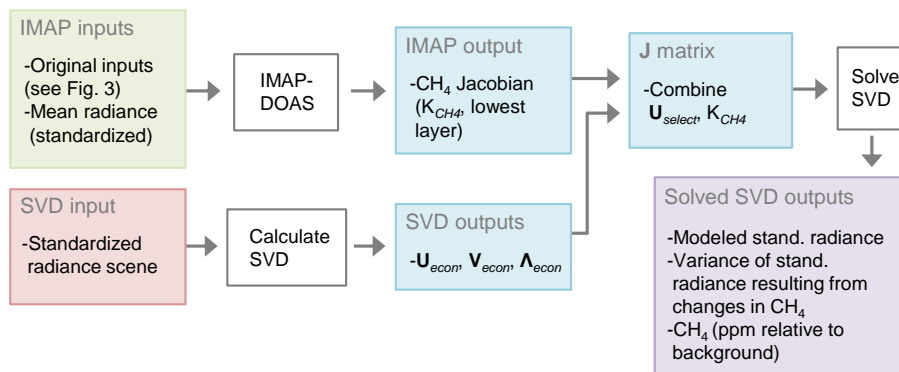


Fig. 5. Processing steps for the SVD retrieval method. The IMAP-DOAS retrieval is performed on a mean radiance for the image subset to generate the CH₄ Jacobian for the lowest layer. The SVD is used to calculate \mathbf{U}_{econ} , \mathbf{V}_{econ} , and Λ_{econ} while \mathbf{U}_{select} is combined with the CH₄ Jacobian to generate the \mathbf{J} matrix. \mathbf{J} is used to determine the portion of each radiance spectra associated with the CH₄ Jacobian (i.e. absorptions due to CH₄) and can be used to estimate CH₄ concentrations.

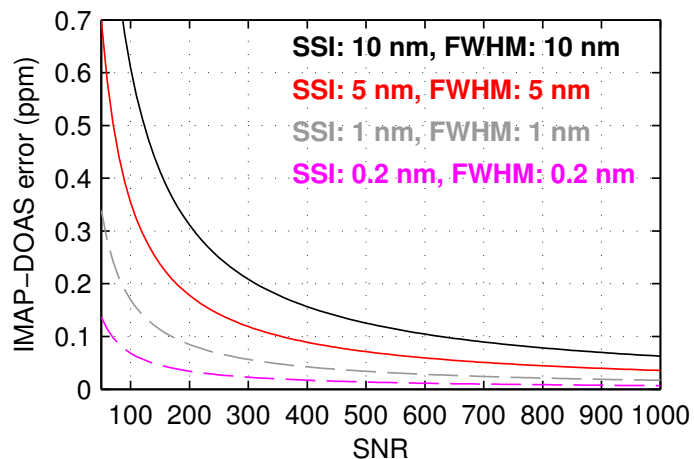


Fig. 6. Estimated IMAP-DOAS retrieval errors (ppm CH₄) for four hypothetical sensors, each with the spectral sampling interval (SSI) equal to the FWHM. Errors are relative to lowest atmospheric layer (height up to 1.04 km) and decline with increased signal to noise ratio (SNR).

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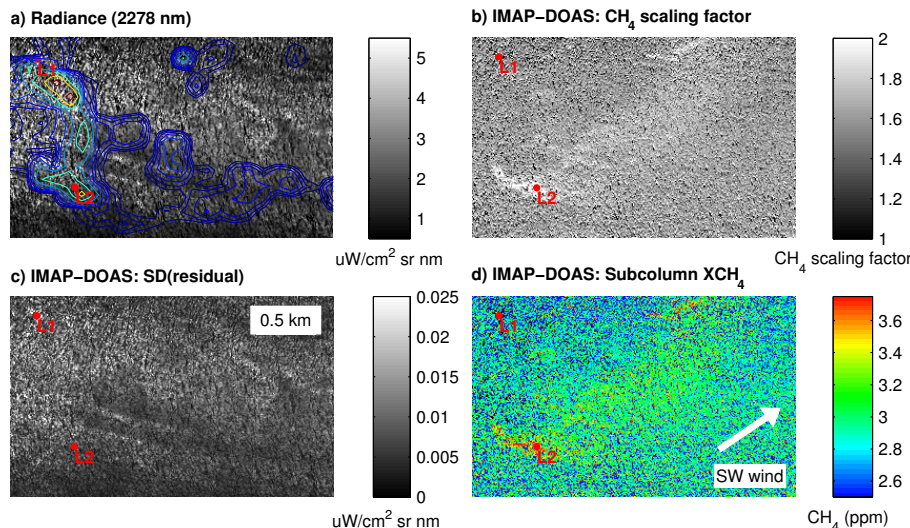


Fig. 7. (a) Measured radiance at 2278 nm showing strong variability in brightness. Sonar return contours (Leifer et al., 2010) are overlain and correspond to known seep locations. (b) For the same image subset, CH_4 scaling factor for the lowest atmospheric layer (layer 10) indicates a CH_4 plume consistent with the local wind direction. (c) The standard deviation of the residuals (measured minus modeled radiance) depends strongly on brightness (a). (d) Subcolumn XCH_4 (ppm CH_4 for the lowest layer) excluding bright pixels (greater than $5 \text{ uW cm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$ in the fitting window) associated with high standard deviation of the residuals. For two spectra (indicated by location L1 and L2), measured and modeled radiance are provided in Fig. 8.

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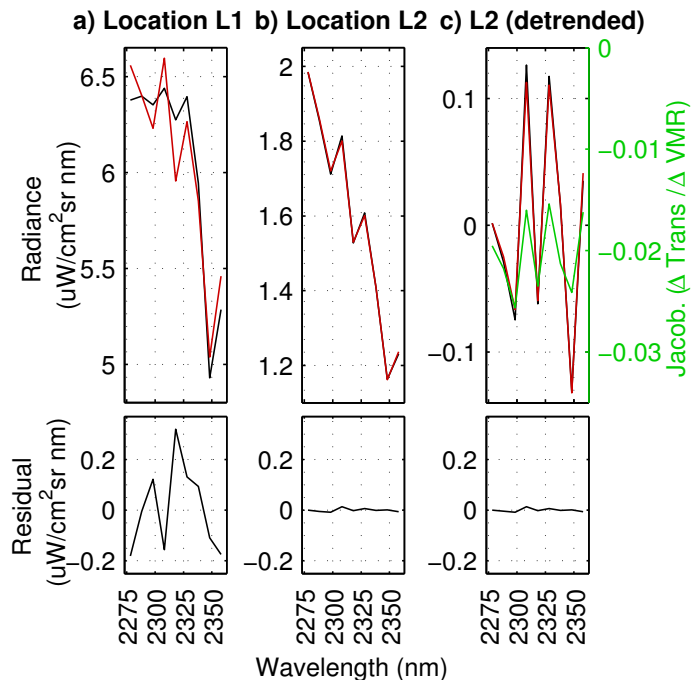


Fig. 8. **(a)** For location L1 (see Fig. 7), the measured radiance (black) indicates sensor saturation due to high sun-glint between 2278 and 2328 nm. This causes considerable disagreement with the modeled radiance (red), as indicated by the residual radiance shown in the bottom plot. **(b)** There is better agreement for location L2. **(c)** The radiance shown in **(b)** was detrended and the CH_4 Jacobian for the lowest layer overlain (green) to indicate the location of CH_4 absorptions at 2298, 2318, and 2348 nm.

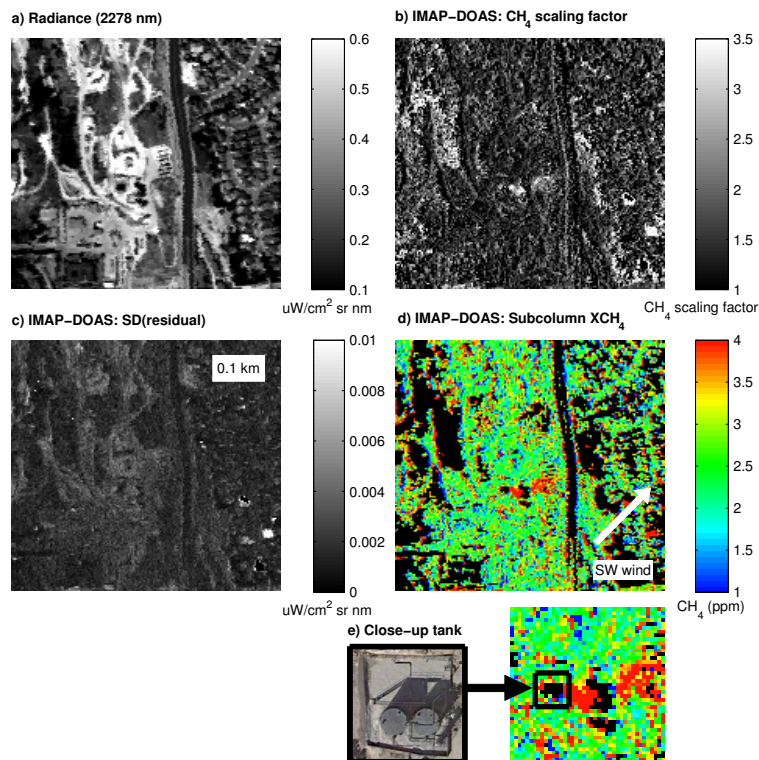


Fig. 9. (a) Radiance at 2278 nm showing a portion of the Inglewood Oil Field. (b) For the same image subset, CH₄ scaling factor for the lowest atmospheric layer (layer 10) appears heavily influenced by land surface type. (c) Standard deviation of the residuals also appears influenced by land cover. (d) Subcolumn XCH₄ (ppm CH₄ for the lowest layer) excluding dark pixels (less than $0.1 \text{ uW cm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$ in the fitting window). (e) Close-up of hydrocarbon storage tanks upwind of observed plume (Google Earth, 2013).

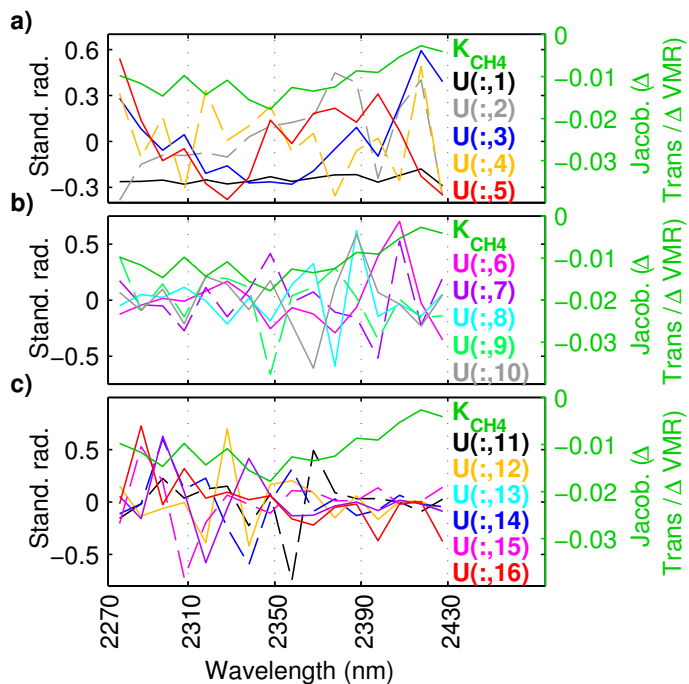


Fig. 10. Singular vectors contained in \mathbf{U}_{econ} for COP scene with CH_4 Jacobian (\mathbf{K}_{CH_4}) plotted for reference.

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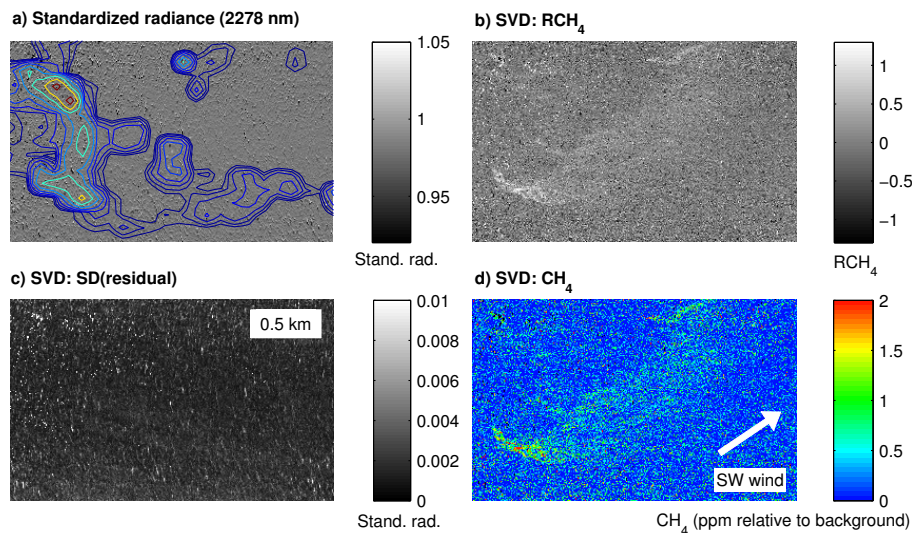


Fig. 11. (a) Standardized radiance used for calculating SVD at COP (showing only 2278 nm). (b) For the same image subset, RCH₄ results clearly indicate CH₄ plume. (c) The standard deviation of the residuals (measured minus modeled radiance). (d) ppm CH₄ relative to background excluding pixels with greater than 0.0075 standard deviation of the residual.

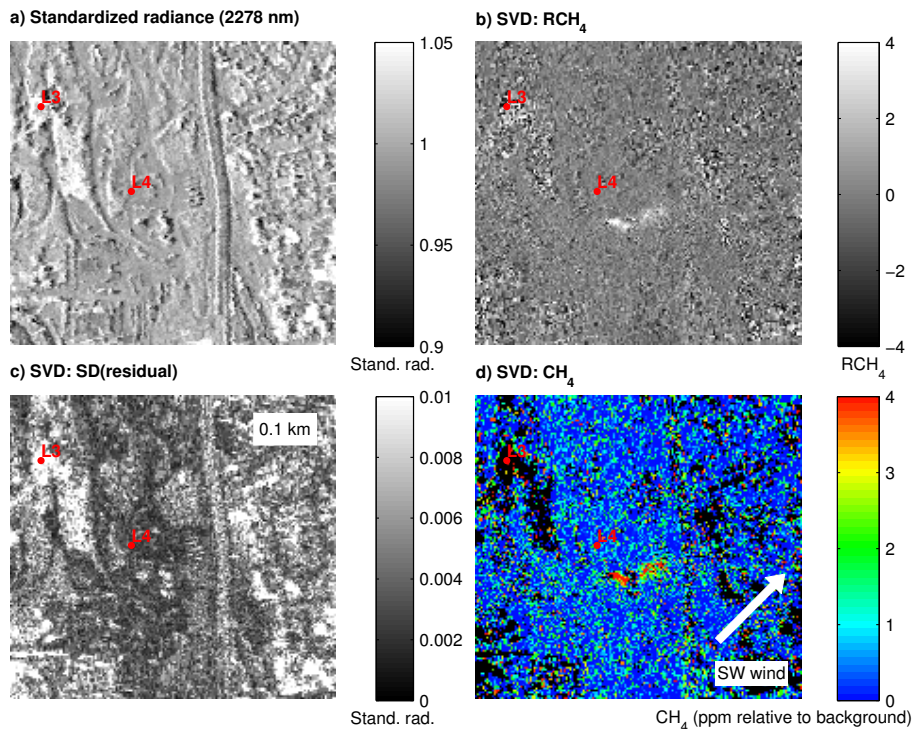


Fig. 12. (a) Standardized radiance used for calculating SVD for Inglewood subset (showing only 2278 nm). (b) For the same image subset, RCH_4 results indicate CH_4 plume at the center of the scene. (c) The standard deviation of the residuals (measured minus modeled radiance). (d) ppm CH_4 relative to background excluding pixels with greater than 0.0075 standard deviation of the residual. For two spectra (indicated by location L3 and L4), measured and modeled radiance are provided in Fig. 13.

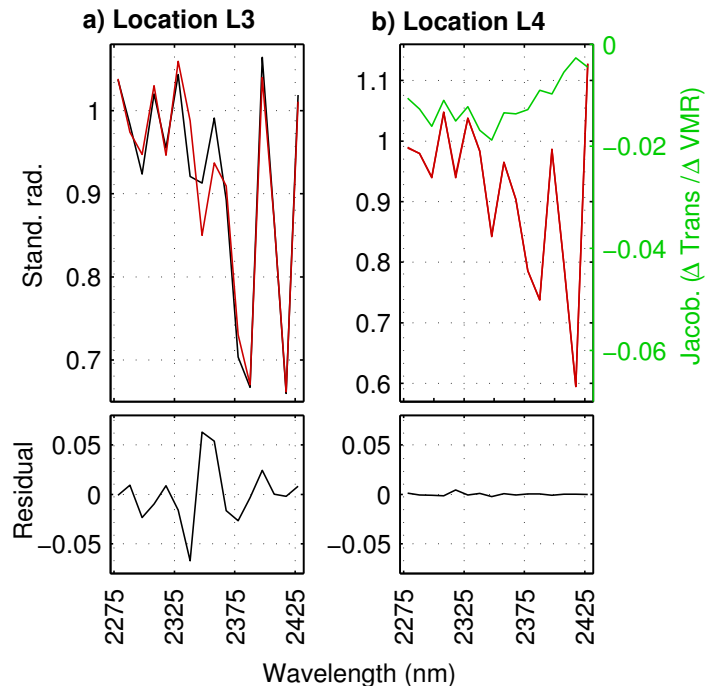


Fig. 13. (a) The modeled (red) and measured standardized radiance (black) for location L3, which corresponds to a dark spectrum with an average radiance of $0.0376 \text{ uW cm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$. L3 is located in a distinct region with high values for the standard deviation of the residuals (see Fig. 12c) and was excluded from the results shown in Fig. 12d. **(b)** For location L4, there is better agreement between modeled and measured radiance (average $0.5187 \text{ uW cm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$). The CH_4 Jacobian for the lowest layer is overlain (green) to indicate the location of CH_4 absorptions.

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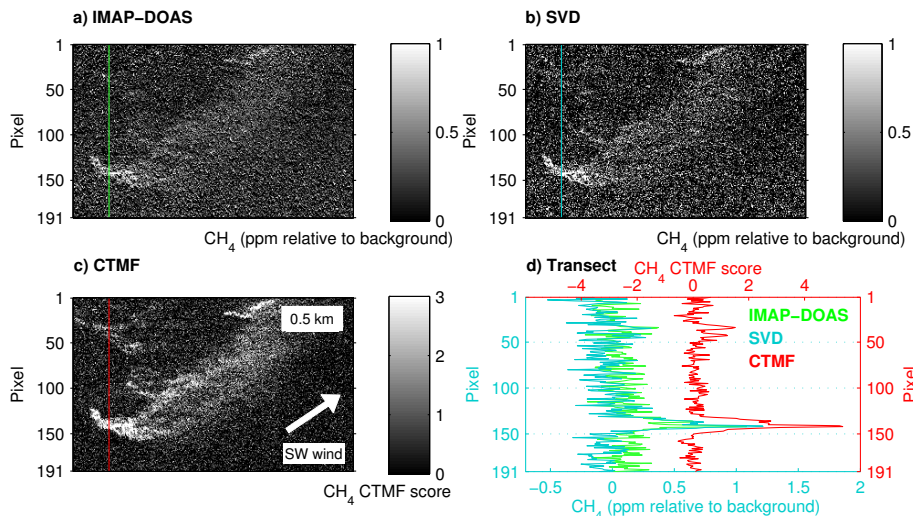


Fig. 14. For the same COP subset, there is good agreement between results obtained using three techniques. **(a)** IMAP-DOAS. **(b)** SVD. **(c)** Cluster-Tuned Matched Filter (CTMF). The location of a vertical transect is shown for the IMAP-DOAS (green line), SVD (cyan), and CTMF results (red). **(d)** Values along the transect are shown for IMAP-DOAS (green), SVD (cyan), and CTMF (red). At each point along the transect, an average value was calculated for 21 pixels centered on the transect in the horizontal direction.

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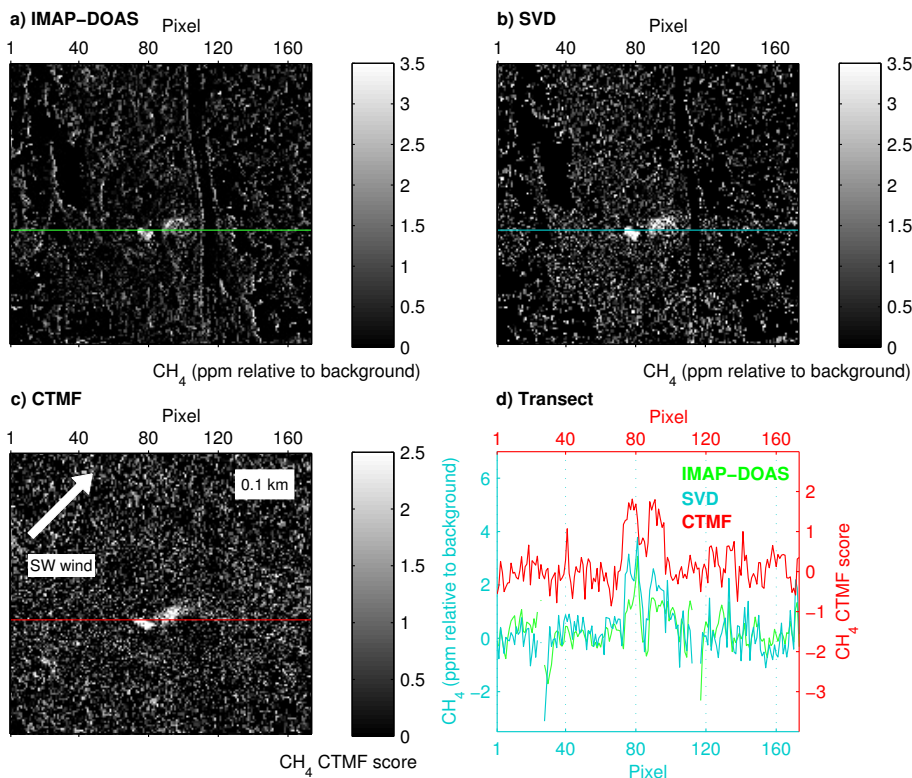


Fig. 15. For the same Inglewood subset, there is good agreement between results obtained using three techniques. **(a)** IMAP-DOAS. **(b)** SVD. **(c)** Cluster-Tuned Matched Filter (CTMF). The location of a horizontal transect is shown for the IMAP-DOAS (green line), SVD (cyan), and CTMF results (red). **(d)** Values along the transect are shown for IMAP-DOAS (green), SVD (cyan), and CTMF (red) approach. At each point along the transect, an average value was calculated for 9 pixels centered on the transect in the vertical direction.

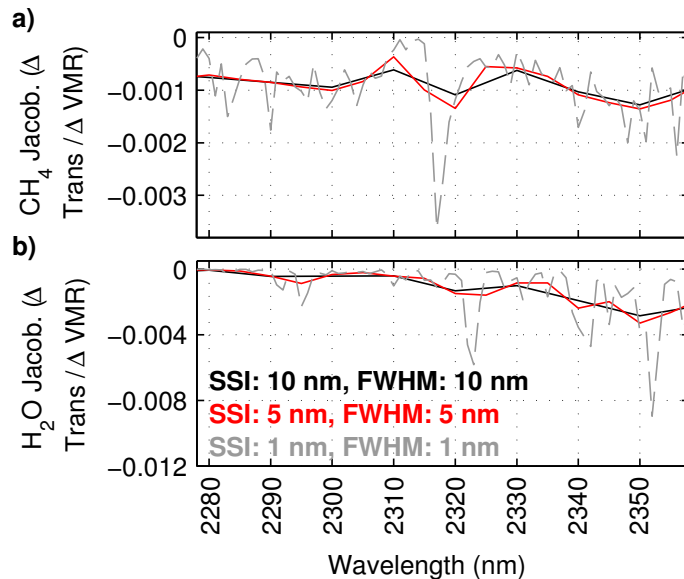


Fig. 16. (a) For the lowest layer of the atmospheric model (height up to 1.04 km), the CH_4 Jacobian calculated for AVIRISng (red) indicates improved sensitivity compared to the CH_4 Jacobian for AVIRIS (black). Even greater sensitivity can be achieved using a finer SSI and FWHM (dashed grey). (b) H_2O Jacobians calculated for the same three sensors.

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