Response to anonymous referee #1

We would like to thanks the referees for their thorough comments.

General comments:

It is known that the short-term regional scale variability in total column CH4 is dominated by local sources is well as the dynamics of weather systems. The model system that has been described takes the first into account, but not the second. I mean, it represents the weather but not the corresponding regional-scale patterns in XCH4. I was looking for ways in which the boundary conditions of CH4 where taken into account, but didn't find any. As a result, the fluxes that are derived may well account for unaccounted variations at the domain boundary.

In the inversion, we have represented the boundary conditions by selecting the observations of X_{CH4} dry air mole fractions unaffected by the local sources in our domain. These observations correspond to ideal wind conditions for each day assuming that over such a small domain, no spatial gradient in X_{CH4} mole fractions are expected and therefore one observation location is sufficient. The wind direction is changing but the minimum value is consistent over the two days at about 1.83-1.832ppm, except for a brief decrease to 1.825ppm on January 16th (for about 20 minutes). On January 15th, different instruments measure this value depending on the location relative to the wind direction. Two main reasons motivated our approach. First, we considered that the relative magnitude of the feedlot emissions is several orders of magnitude larger than any other sources in the area. The Chino feedlot is one of the largest sources of CH_4 in the Los Angeles basin, which greatly simplifies the problem of the background conditions (i.e. the relative contributions from other sources are reduced). Nearby sources could still be significant if they are located at a very short distance from the sensors. But drive-around data and upwind EM27/SUN observations did not suggest any prevalent sources around the feedlot, as illustrated by the various EM27/SUN sensors measuring the same background at different times on January 15th. Second, if we were to consider a more regional approach to the problem, we would need a very accurate representation of CH_4 mixing ratios (to few ppb) and therefore an accurate mapping of CH_4 sources within several kilometers around the feedlot in order to simulate the signals from other contributors. This approach is likely to produce larger errors due to missing sources and incorrect magnitudes in the inventories as well as errors in the atmospheric transport, which could all introduce significant biases in the background mixing ratios (much larger than 2ppb). We would also need to define the inflow of CH_4 from outside Los Angeles which would require another set of data outside the city limits. For these reasons, we decided to use an observation-based approach to avoid the complications due to model and inventory errors. We have modified the text to discuss the problem and explain more precisely our approach.

In Chen et al. an upwind-downwind approach was taken, which might indeed work under ideal conditions, with a well defined wind direction. As can be seen in Figure 4, this logic falls apart for low wind speeds. It would have been instructive to plot the zero line in this figure.

We agree and zero lines have been added to each plot.

Unlike the 24th of January, the gradient with respect to Harvard 2 is going everywhere, most notably on the 15th. Therefore I don't understand the role of Harvard 2 is a background on days with a low wind speed and doubt that it can be used that way. The authors may argue that variations in the background may be less important on days with lower wind speed, but this could en should then be demonstrated.

We needed to have the same background instrument for all 3 days of measurements to test the mass balance approach. The Harvard2 X_{CH4} mean and standard deviation are the lowest of all the observations (Figure 3); which is why we have chosen this instrument as a measure of background conditions. We added a sentence on this limitation in the text.

Before the WRF-LEF model is used to fit the FTS data it should be demonstrated that it has a reasonable skill in simulating the observed variability. The fit in S2 doesn't look great (what are the R2 values?), which makes me wonder about the comparison with the prior model. In this context it would be very useful to compare the use of WRF-LES with the use of WRF alone. What do we gain using LES? The results may not look great, but we'd learn about where the remaining problems are.

We have added a paragraph and a reference to a paper that will be published in the coming weeks (Gaudet et al., 2017). This study presents the ability of the Large Eddy Simulation mode to represent plume structures over short distances (less than 10km) compared to the mesoscale mode. Here, we have no simulation of the CH_4 concentrations from coarse-resolution grids. Only 111-m concentrations were simulated.

In line 370 it is mentioned that the modelled wind speed has an error of 'only' 1 m/s increasing further in the PBL. However, on the low wind-speed days this is a very substantial fraction of the total wind speed (according to Figure 1). What is the sensitivity of the emission estimation to errors in the modelling of wind speed and direction? I'd like again to stress that it is important that we learn about what is critical in this approach. The size of the errors in wind speed and direction warrant closer inspection and a test of the potential impact.

The impact of transport errors on inverse estimates is an important topic that has been discussed in previous studies at lower resolutions (e.g. Lauvaux and Davis, 2014). In our study, it remains difficult to address this problem without the use of an ensemble of simulations. Instead, we decided to improve the transport using the WRF-FDDA system. We refer to another study (Deng et al., 2017) that will be published in the coming weeks. In this paper, we performed multiple simulations using WRF-FDDA and propagated the impact of the assimilation into the inverse fluxes. We showed that transport errors were significantly reduced when assimilating vertical profiles of meteorological observations. Here, our estimates of model errors, based on the chi2 normalized distance, suggest that transport errors are relatively small (less than 3ppb) allowing us to produce an inverse estimate for the area. We added a paragraph about model errors.

It is clear to me why the mass balance approach is not used on the 15th and 16th of January. However, it is not clear why the WRF-LES method is not used on the 24th. This would provide a good opportunity to make a direct comparison between the two methods. It may not be the best case to demonstrate the added value of WRF LES, but as a general consistency check to include this comparison is nevertheless necessary.

We decided to avoid January 24th as the wind direction remains constant during the entire day. The inversion uses primarily the information from various wind conditions to constrain the distribution of the CH_4 sources, similar to a triangulation approach. The results for January 15th and 16th revealed that sources are highly variables across the domain. Therefore, the lack of constraints on the spatial distribution (i.e. no change in wind direction) would have been limiting on January 24th. We explain our choice in the section 4.3.1.

Figures 4 and 5 confirm my worry about the emission pattern that is shown in Figure 12 and I wonder why it doesn't receive more attention. The pattern correlates with the configuration of the measurement 'network'. For an inversion the easiest way to fit the data is to modify the emissions in the same grid box as where the measurements are. It may either be that the measurements are very locally influenced, in which case they are not really representative of the domain that is being optimized. Otherwise the inversion may be trying to fit uncertain variables that are not part of the state vector, such as boundary conditions (see my earlier comment) or the emission distribution within 2x2km2 regions. With in situ measurements at LANL as high as 30 ppm CH4, this may well be an important factor. To me this sounds certainly like a more relevant factor to mention than the emission of a 1 cow per year power plant.

Figures S4 and S5 provide additional details to understand the attribution of signals in the state vector. Overall, the inversion is using the triangulation of information from various wind directions, which is why the two-day inversion is likely to produce the best estimate over the period. In general, with 2 sites, the flux corrections are applied to the nearby pixels. But this result is not valid when using 3 or 4 sensors. For example, on January 15th, two sensors (i.e. LANL and Caltech) show no correction in their vicinity whereas major changes are located near the other two sensors. The optimization is consistent across configurations of 3 or 4 sensors, detecting a major source in the North-East and the South-West, but not near the other two sensors. On January 16th, the fluxes remain unchanged near the Caltech instrument which demonstrates that the system is doing better than simply attributing the corrections to the nearly pixels. We provide additional comments based on Figure S4.

It was not clear to me why the simulated annealing approach was chosen. Since the inversion problem is linear, I don't see why its solution should be any different from the Bayesian method. If I understand well, the method was introduced to deal with the difficulty to define the B matrix. But how does simulated annealing solve this problem? Is it just efficiency at which different options for the B matrix can be tried out? If the greens functions are available, then I wonder how simulated annealing could be faster than a Bayesian inversion. This should be explained better.

We have described the Simulated Annealing (SA) in a separate paragraph. The main reason to perform the inversion in two steps is that SA has no prior information which means that if a pixel is not observed, the system will produce an infinite number of solutions. In addition, SA is producing scores (i.e. mismatches between observations and model concentrations) but no optimal solution is guaranteed. SA simply scans the space of solution (i.e. state space) without converging, whereas the Bayesian inversion will provide the optimal analytical solution considering our prescribed prior errors. In theory, if the optimal solution was found by the SA, then the two methods should agree, but there is no guarantee of such agreement.

Specific comments:

Line 98: 'on a high-wind data'. What does this mean? Why is Chen 2016 not included in Table 1?

This has be changed to 'recorded on favorable meteorological conditions (e. g. on a high-wind with constant direction)'. Chen et al. 2016 estimation has been added to Table 1.

Line 117: In this sub section I am missing numbers for the estimated accuracy of the mobile column measurements. How realistic are the fits to the data derived later on in the light of these uncertainties?
We have added a line about accuracy of these mobile column measurements: "Using Allan analysis, it has been found out that the precision of the differential column measurements ranges between 0.1-0.2 ppb with 10 min averaging time (Chen et al., 2016)".

Line 196: ’one way nested grids’: S1 should provide further information about nesting. For CH4 the logical on-way nesting is from the small to the large domain. For meteorology, however, the reverse seems true. Some further explanation is needed.
"one-way" refers to the meteorology only. CH_4 concentrations are simulated only in the 111-m grid. Considering the low risk that CH_4 molecules would re-circulate over the area after having been advected out of the small domain, we ignored the coupling between the grids for CH4. This problem is more significant at larger scales when circulation of air masses is more circular around pressure centers or due to the terrain/surfaces. We provide additional details in S1.

Line 207: To avoid confusion about optimizing methane fluxes and optimizing meteorology it should be stated more clearly that ’data assimilation’ is referring to the latter here.
This has been clarified. We have added "to optimize meteorological fields" after "Data assimilation".

Line 255: What does the random draw refer to: the starting point of simulated annealing, the first guess, a random modification of the prior uncertainty? Does this modify the actual cost function that is optimized?
Simulated Annealing is a random walk which implies that random draws of the prior emissions are made at every iteration. There is no optimization of a cost function in Simulated Annealing, only a score calculated for every possible solution. We clarified the sentence.

Line 257: ’Mean absolute error’ This approach favors the setup that gives the largest freedom to the prior fluxes. This need not be the best solution, or maybe I do not understand what is meant here (see the previous point).
The impact of using the Mean Absolute Error is minor. We could have used the squared distances (like in a typical least-square regression analysis) or the Mean Error which would account for source compensation.

Line 290: ’C’ io ’SC’?
This has been changed

Line 373: ’More relevant to this study’ It is unclear why this should be the case. The difference between the two metrics represents a cancellation of wind speeds in calculating the mean. Such errors could still affect the estimated emissions, e.g. when winds with cancelling errors come from different directions.
The sentence refers to the fact that the first error is computed for the Free Atmosphere (Free Troposphere) whereas our local enhancements are located in the PBL. We clarified the sentence.

Line 458: It is clear that the measurements in Figure 11 represent ruminant emissions. However, these samples are not representative of the air masses that are sampled with the FTS instruments. Therefore this result does not refer to the origin of the enhancements in total column CH4 discussed earlier. Confusion should be avoided on this point.
We have added ‘measured by the Picarro instrument’ to clarify this point.

Line 579: ’The main advantage ...’ This statement should be supported by evidence or a reference to other work. We have added a reference (Wunch et al. 2011).

Figure 1: The wind roses do not add up to 100%. For clarity, we have not shown in this Figure wind data corresponding to null wind speeds, which is why the roses do not add up to 100%.

Figure 5: Please repeat that anomalies are relative to Harvard 2. We have added this in the caption.

Figure 6: It should be made clear that these errors refer to errors in WRF-LES simulated wind speed and direction. As indicated within the panels of Figure 6, the model used to compute these statistics is the 3-km domain, not the 111-m WRF-LES domain. Verification against multiple WMO radiosondes would not be possible for the 111-m WRF-LES domain because it is too small.

Figure 7: Mention the minimum WMO threshold value. We added the value in the caption.

S1: How about the top boundary in the LES part of the domain? The molecules of CH_4 are not well-mixed over the PBL. For this reason, we avoided the use of PBL height as a relevant criterion in our calculation. In the model configuration the top of the LES is the same as the top of the other domains.

Technical corrections:

All of them have been changed

Line 288: ’V is’ io ’Vis’

Line 337: area of Chino in m2

Line 353: ’has’ io ’have weaknesses’

Figure 8: Information on axes parameters and units should be given in the figures themselves instead of the caption. Please also add labels to the rows (as is done for columns)

Figure 9: Figure axes and legend without labels.

Title S1: ’models’ io ’modes’

Figures S4 and S5: Text is too small to read.

Response to anonymous referee #2

We would like to thanks the referees for their insightful comments.

Specific comments:

1. How does the width of the FTS instruments' measurement swath compare to the width of the plume? One reason the "golden day" might not work for the basic flux method described here is because the three downwind FTS instruments are only measuring a small portion of the total plume. Please add some discussion of how this may affect the uncertainty of the flux method.

We have added in the text: 'For that day, the high wind speed causes a reduction of the methane plume width across the feedlot, which may increase uncertainties on the mass-balance approach since the FTS' measurements may only detect a small portion of the total plume.'

2. Line 455, Are these the results from a Keeling plot? Or are you just looking at the variability of d13C? If you look at the enhancements and a Keeling plot, do the d13C values make sense with the notion that the methane enhancements are consistent with dairy emissions?

The values written at line 455 represent the variability of d13C measures in the feedlot. Looking at the Keeling plot for all the measurements, we obtained values around -40 ‰ (mixture of biogenic and fossil sources). However, when driving thought the feedlot with the Picarro instrument, methane enhancements associated with low d13C were recorded, especially when measuring right next to the cows. Therefore, methane emissions are found to be consistent with dairy emissions.

3. How does the length of the measurement period couple into the uncertainties of the inversion? Are three days of measurements for this size and strength of emission sufficient? Could you have gotten away with fewer, or would more have helped?

The assessment of uncertainties remains undetermined at this point and would require additional measurements to perform an independent evaluation. From the two days of data, we were able to retrieve a value that corresponds to previously published estimates. But the errors remain significant. Continuous measurements over several weeks would have provided a better estimate on the inverse fluxes by constraining the model with concentrations and meteorological fields for many different atmospheric conditions. However, results show (on Figure 10) that flux inversion obtained on two different days (the 15th and the 16th of January) lead to relatively similar results. Increasing the number of days in the inversions would reduce the confidence interval but the use of independent data (such as eddy-covariance flux tower measurements) would be needed to evaluate the performance of the system. The authors aim to demonstrate that FTS measurement network coupled with WRF-LES inversion is a powerful methodology to derive local fluxes, even using relatively small number of data.

4. Are there methane sources upwind that may contribute to the model placing methane emissions in the southeast portion of the study area? For example, how well does the LANL

16th site provide a background for the H1 16th site? If the H1 16th site sees an enhancement in methane that is not measured at the LANL 16th site, must the inverse model place those emissions in the southeastern most section of the grid?

The source attribution problem is solved by combining various wind conditions over the one- or two-day time window (depending on the inversion case). As suggested in the question, information from observed enhancements will be attributed to specific areas depending on the downwind measurements site and the wind direction/speed. We selected two days with varying wind conditions in order to sample the entire domain at different times and with different combinations of sites (upwind/downwind). Whereas our sample size remains small, we have observed most of the wind directions (cf. wind roses in Figure 1) which suggest that our retrieved sources have been measured by more than one combination of sites. We agree though that some of our signals may be more constrained by a specific time (and therefore a specific set of sites) than others. A longer time window of observations with repeated enhancements from different wind directions would help support our findings. Here, we show that major sources can be identified across the feedlots. As an analogy, the inversion performs a triangulation of the sources assuming that our spatial coverage was sufficient. This coverage is a direct function of the wind direction variability as our sites are static.

5. Is there a way to quantify the reduction of dairies between 2010 and 2015? If so, I think this would be a good addition to help strengthen the conclusions based on comparisons with past emissions estimates.

Inventories of dairy and cow's numbers in this area are inexistent. Only previous studies within the area (Peischl et al. 2013, Wennberg et al. 2012) could be used to quantify the reduction of dairies between 2010 and 2015.

Methane emissions from dairies in the Los Angeles Basin

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Abstract

We estimate the amount of methane (CH_4) emitted by the largest dairies in the southern California region by combining measurements from four mobile solar-viewing ground-based spectrometers (EM27/SUN), in situ isotopic $^{13/12}CH_4$ measurements from a CRDS analyzer (Picarro), and a high-resolution atmospheric transport simulation with Weather Research and Forecasting model in Large-Eddy Simulation mode (WRF-LES).

The remote sensing spectrometers measure the total column-averaged dry-air mole fractions of CH_4 and CO_2 (X_{CH4} and X_{CO2}) in the near infrared region, providing information about total emissions of the dairies at Chino. Differences measured between the four EM27/SUN ranged from 0.2 to 22 ppb (part per billion) and from 0.7 to 3 ppm (part per million) for X_{CH4} and X_{CO2}, respectively. To assess the fluxes of the dairies, these differential measurements are used in conjunction with the local atmospheric dynamics from wind measurements at two local airports and from the WRF-LES simulations at 111 m resolution.

Our top-down CH_4 emissions derived using the Fourier Transform Spectrometers (FTS) observations of 1.4 to 4.8 ppt/s are in the low-end of previous top-down estimates, consistent with reductions of the dairy farms and urbanization in the domain. However, the wide range of inferred fluxes points to the challenges posed by heterogeneity of the sources and meteorology. Inverse modeling from WRF-LES is utilized to resolve the spatial distribution of CH_4 emissions in the domain. Both the model and the measurements indicate heterogeneous emissions, with contributions from anthropogenic and biogenic sources at Chino. A Bayesian inversion and a Monte-Carlo approach are used to provide the CH_4 emissions of 3.2 to 4.7 ppt/s at Chino.

1) Introduction

Atmospheric methane (CH_4) concentration has increased by 150% since the pre-industrial era, contributing to a global average change in radiative forcing of 0.5 $W.m^{-2}$ (Foster et al., 2007; Myhre et al., 2013). Methane is naturally emitted by wetlands, but anthropogenic emissions now contribute more than half of its total budget (Ciais et al., 2013), ranking it the second most important anthropogenic greenhouses gas after carbon dioxide (CO_2).

The United Nations Framework Convention on Climate Change (UNFCCC, http://newsroom.unfccc.int/) aims to reduce CH_4 emissions by reaching global agreements and collective action plans. In the United States (US), the federal government aims to reduce CH_4 emissions by at least 17% below 2005 levels by 2020 by targeting numerous key sources such as (in order of importance): agriculture, energy sectors (including oil, natural gas, and coal mines), and landfills (Climate Action Plan, March 2014). Methane emissions are quantified using “bottom-up” and "top down" estimates. The "bottom-up" estimates are based on scaling individual emissions and process level information statistically (such as the number of cows, population density or emission factor) with inherent approximations. “Top-down” estimates, based on atmospheric CH_4 measurements, often differ from these reported inventories both in the total emissions and the partitioning among the different sectors and sources (e.g. Hiller et al., 2014). In the US, the disagreement in CH_4 emissions estimated can reach a factor of two or more (Miller et al., 2013; Kort et al., 2014), and remains controversial regarding the magnitude of emissions from the agricultural sector (Histov et al. 2014). Thus, there is an acknowledged need for more accurate atmospheric measurements to verify the bottom-up estimates (Nisbet and Weiss, 2010). This is especially true in urban regions, such as the Los Angeles basin, where many different CH_4 sources (from farm lands, landfills, and energy sectors) are confined to a relatively small area of ~87000 km^2 (Wunch et al., 2009; Hsu et al. 2010; Wennberg et al., 2012; Peischl et al., 2013; Guha et al., 2015; Wong et al., 2015). Therefore, improved flux estimations at local scales is needed to resolve discrepancies between bottom-up and top-down approach and improve apportionment among CH_4 sources.

Inventories of CH_4 fluxes suggest that emissions from US agriculture increased by more than 10% between 1990 and 2013 (Environmental Protection Agency, EPA, 2015), and by more than 20% since between 2000 and 2015 in California (California Air Resources Board, CARB, 2015). In addition, these emissions are projected to increase globally in the future due to increased food production (Tilman and Clark, 2014). Livestock in California have been estimated to account for 63% of the total agricultural emissions of greenhouse gases (mainly CH_4 and N_2O); dairy cows represented more than 70% of the total CH_4 emissions from the agricultural sectors in 2013 (CARB, 2015). State-wide actions are now underway to reduce CH_4 emissions from dairies (ARB concept paper, 2015). Measurements at the local-scale with high spatial- and temporal-resolution are needed to assess CH_4 fluxes associated with dairy cows and to evaluate the effectiveness of changing practices to mitigate CH_4 emissions from agriculture.

Space-based measurements provide the dense and continuous datasets needed to constrain CH_4 emissions through inverse modeling (Streets et al., 2013). Recent studies have used the Greenhouse gases Observing SATellite (GOSAT – footprint of ~10 km diameter) observations to quantify mesoscale natural and anthropogenic CH_4 fluxes in Eurasia (Berchet et al., 2015) and in the US (Turner et al., 2015). However, it is challenging to estimate CH_4 fluxes at smaller spatial scales using satellite measurements due to their large observational footprint (Bréon and Ciais, 2010). Nevertheless, recent studies used the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY – footprint of 60 km x 30 km) to assess emissions of a large CH_4 point source in the US (Leifer et al., 2013; Kort et al., 2014).

Small-scale CH_4 fluxes are often derived from in situ measurements performed at the surface and from towers (Zhao et al., 2009), and/or in situ and remote-sensing measurements aboard aircraft (Karion et al.,2013; Peischl et al., 2013; Lavoie et al., 2015; Gordon et al., 2015). A recent study emphasized the relatively large uncertainties of flux estimates from aircraft measurements using the mass balance approach in an urban area (Cambaliza et al., 2014).

Ground-based solar absorption spectrometers are powerful tools that can be used to assess local emissions (McKain et al., 2012). This technique has been used to quantify emissions from regional to urban scales (Wunch et al., 2009; Stremme et al., 2013; Kort et al., 2014; Lindenmaier et al., 2014; Hase et al., 2015; Franco et al., 2015, Wong et al., 2015, Chen et al., 2016; Kille et al., 2017).

In this study, we use four mobile ground-based total column spectrometers (called EM27/SUN, Gisi et al., 2012) to estimate CH_4 fluxes from the largest dairy-farming area in the South Coast Air Basin (SoCAB), located in the city of Chino, in San Bernardino County, California. The Chino area was once home to one of the largest concentrations of dairy farms in the United States (US), however rapid land-use change in this area may have caused CH_4 fluxes from the dairy farms change rapidly in both space and time. Chen et al. (2016) used differential column measurements (downwind minus upwind column gradient ΔX_{CH4} across Chino) recorded on favorable meteorological conditions (e. g. constant wind direction) to verify emissions reported in the literature. In this study, the same column measurement network is employed in conjunction with meteorological data and a high-resolution model to estimate CH_4 emissions at Chino for several different days, including more varying wind conditions. The approach proposed here allows us to describe the spatial distributions of CH4 emissions within and around the feedlot at very high resolution by using an advanced atmospheric modeling system applicable to any convective meteorological conditions (Gaudet et al., 2017).

In section 2 of this paper, the January 2015 field campaign at Chino is described, with details about the mobile column and in situ measurements. In section 3, we describe the new high resolution Weather Research & Forecasting (WRF) model with Large Eddy Simulations (LES) setup. In section 4, results of CH_4 fluxes estimates are examined. Limitations of this approach, as well as suggested future analyses are outlined in section 5.

2) Measurements in the Los Angeles Basin dairy farms

2.1) Location of the farms: Chino, California

Chino (34.02°N, -117.69°W) is located in the eastern part SoCAB, called the Inland Empire, and has historically been a major center for dairy production. With a growing population and expanding housing demand, the agricultural industry has shrunk in this region and grown in the San Joaquin Valley (California Central Valley). The number of dairies decreased from ~400 in the 1980's to 95 in 2013 (red area of panels a, b, and c in Figure 1). Nevertheless, in 2013 ~90 % of the southern California dairy cow population (California Agricultural Statistics, 2013) remained within the Chino area of ~6 x 9 km (Figure 1). These feedlots are a major point source of CH_4 in the Los Angeles basin (Peischl et al., 2013).

2.2) Mobile column measurements: EM27/SUN

Atmospheric column-averaged dry-air mole fractions of CH_4 and CO_2 (denoted X_{CH4} and X_{CO2}, Wunch at al., 2011) have been measured using four ground-based mobile Fourier Transform Spectrometers (FTS). The mobile instruments were developed by Bruker Optics, are all EM27/SUN models. The four FTS (two owned by Harvard University, denoted Harvard 1 and 2, one owned by Los Alamos National Laboratory, denoted LANL, and one owned by the California Institute of Technology, denoted Caltech, were initially gathered at the California Institute of Technology in Pasadena, California in order to compare them against the existing Total Carbon Column Observing Network (TCCON, Wunch et al., 2011) station and to each other, over several full days of observation. The instruments were then deployed to Chino to develop a methodology to estimate greenhouses gas emissions and improve the uncertainties on flux estimates from this major local source. Descriptions of the capacities and limitations of the mobile EM27/SUN instruments have been published in Chen et al. (2016) and Hedelius et al. (2016). Using Allan analysis, it has been found out that the precision of the differential column measurements ranges between 0.1-0.2 ppb with 10 min averaging time (Chen et al., 2016). For this analysis, we need to ensure that all the data from the EM27/SUN instruments are on the same scale. Here, we reference all instruments to the Harvard2 instrument. Standardized approaches (retrieval consistency, calibrations between the instruments) are needed to monitor small atmospheric gradients using total column measurements from the EM27/SUN. Indeed we ensured all retrievals used the same algorithm, calibrated pressure sensors, and scaled retrievals according to observed, small systematic differences to reduce instrumental biases (Hedelius et al., 2016).

These modest resolution (0.5 cm^{-1}) spectrometers are equipped with solar-trackers (Gisi et al., 2011) and measure throughout the day. To retrieve atmospheric total column abundances of CH_4, CO_2, and oxygen (O_2) from these Near InfraRed (NIR) solar absorption spectra, we used the GGG software suite, version GGG2014 (Wunch et al., 2015). Column measurements at Chino were obtained on five days: the 15th, 16th, 22nd and 24th of January, and the 13th of August, 2015. Of these days, January 15th, 16th, and 24th are sufficiently cloud-free for analysis. These days have different meteorological conditions (i.e. various air temperatures, pressures, wind speeds and directions), improving the representativeness of the flux estimates at Chino.

Figure 1 shows measurements made on January 15th, 16th, and 24th. Wind speeds and directions, shown in the bottom panels of Figure 1, are measured at the two local airports inside the domain (the Chino airport indicated on panels d, e, and f and the Ontario airport on panels g, h, and i). Wind measurements from these two airports, located at less than 10 km apart, are made at an altitude of 10 meters above the surface. The exact locations of the four EM27/SUN spectrometers (colored symbols in Figure 1 in the upper panels a, b, and c) were chosen each morning of the field campaign to optimize the chance of measuring upwind and downwind of the plume. On the 15th and 16th of January, the wind speed was low with a maximum of 3 ms^{-1} and highly variable direction all day (Figure 1, panels d, e, g and h), therefore the four EM27/SUN spectrometers were placed at each corner of the source area to ensure that the plume was detected by at least one of the instruments throughout the day. On the contrary, the wind in January 24th had a constant direction from the Northeast and was a relatively strong 8-10 ms^{-1} (Figure 1, panels f and i), so the instruments were located such that one spectrometer (Harvard2) was always upwind (blue symbols in Figure 1) and the others are downwind of the plume and at different distances from the sources (black, green, and red symbols in Figure 1).

2.3) In situ measurements: Picarro

The EM27/SUN column measurements are supplemented by ground-based in situ measurement using a commercial Picarro instruments during January campaign. The Picarro instruments use a Cavity Ringdown Spectroscopy (CRDS) technique that employs a wavelength monitor and attenuation to characterize species abundance.

In situ $^{12}CH_4$, CO_2, and $^{13}CH_4$ measurements were performed on January 15th, 16th, and 22nd, and August 13th 2015 at roughly 2m away from the LANL EM27/SUN (Figure 1 a, b, and c) with a Picarro G2132-I instrument (Arata et al., 2016, http://www.picarro.com/products_solutions/isotope_analyzers/). This Picarro, owned by LANL, utilize a 1/4" synflex inlet tube placed approximately 3m above ground level to sample air using a small vacuum pump. Precisions on $^{12}CH_4$, CO_2, and $^{13}CH_4$ measurements are 6 ppb, 2 ppm, and 0.6 ‰, respectively.

To locate the major CH_4 sources in the dairy farms area, a second Picarro G2401 instrument (http://www.picarro.com/products_solutions/trace_gas_analyzers/) from the Jet Propulsion Laboratory (JPL, Hopkins et al., 2016) was deployed on January 15th, 2015. Precision on CH_4 measurements is ~1 ppb.

3) Model simulations

3.1) Description of WRF-LES model

The Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) is an atmospheric dynamics model used for both operational weather forecasting, and scientific research throughout the global community. Two key modules that supplement the baseline WRF system are used here. First, the chemistry module WRF-Chem (Grell et al., 2005) adds the capability of simulating atmospheric chemistry among various suites of gaseous and aerosol species. In this study, CH_4 is modeled as a passive tracer because of its long life time relative to the advection time at local scales. The longest travel time from the emission source region to the instrument locations is less than an hour, which is extremely short compared to the lifetime of CH_4 in the troposphere (~9 years). Therefore, no specific chemistry module is required. The version of WRF-Chem used here (Lauvaux et al., 2012) allowed for the offline coupling between the surface emissions, prescribed prior to the simulation, and their associated atmospheric tracers. Second, we make use of the Large Eddy Simulation (LES) version of WRF (Moeng et al., 2007) on a high-resolution model grid with 111-m horizontal grid spacing. A key feature of the simulation is the explicit representation of the largest turbulent eddies of the Planetary Boundary Layer (PBL) in a realistic manner. The more typical configuration of WRF (and other atmospheric models) is to be run at a somewhat coarser resolution that is incapable of resolving PBL eddies. An advantage in this study is that the effect of most important PBL eddies to vertical turbulent transport (i.e., the largest eddies) is not parameterized. By having a configuration with the combination of CH_4 tracers and PBL eddies, we can realistically predict the evolution of released material at scales on the order of the PBL depth or smaller. The WRF-LES mode has been evaluated over Indianapolis, IN and compared to the commonly-used mesoscale mode of WRF (Gaudet et al., 2017). The representation of plume structures in the horizontal and in the vertical is significantly improved at short distances (<8km) compared to mesoscale simulations at 1km resolution, while the meteorological performance of WRF-LES remains similar to coarser domains due to the importance of boundary nudging in the nested-domain configuration. Thus, the representation of the CH_4 plumes in this study should be significantly improved with the LES mode configuration of Gaudet et al. (2017).

In this real case experiment, the model configuration consists of a series of four one-way nested grids, shown in Figure 2 and described further in the supplementary information section (S1). Each domain contains 201 x 201 mass points in the horizontal, with 59 levels from the surface to 50 hPa, and the horizontal grid spacings are 3 km, 1 km, 333 m, and 111 m. All four domains use the WRF-Chem configuration. The model 3-km, 1-km, and 333-m grids are run in the conventional mesoscale configuration with a PBL parameterization, whereas the 111-m grid physics is LES. The initial conditions for the cases are derived from the National Centers for Environmental Prediction (NCEP) 0.25-degree Global Forecasting System (GFS) analysis fields (i.e., 0-hour forecast) at 6-hour intervals. The simulations are performed from 12:00 to 00:00 UTC (= 04:00 to 16:00 LT) only, which corresponds to daylight hours when solar heating of the surface is present and measurements are made.

Data assimilation to optimize meteorological fields is performed using Four Dimensional Data Assimilation (FDDA; Deng et al., 2009) for the 3-km and 1-km domains. The assimilation improves the model performance significantly (Rogers et al., 2013; Deng et al., 2017) without interfering with mass conservation and the continuity of the air flow. Surface wind and temperature measurements, including from the Ontario (KONT) and Chino (KCNO) airport stations, and upper-air measurements were assimilated within the coarser grids using the WRF-FDDA system. However, no observations of any kind were assimilated within the 333-m and 111-m domains; therefore, the influence of observations can only come into these two domains through the boundary between the 333-m and 1-km grids. Wind measurements at fine scale begin to resolve the turbulent perturbations, which would require an additional pre-filtering. These measurements are used to evaluate the WRF model performances at high resolutions.

Based on the terrain elevation in the LES domain (Figure 2), target emissions are located in a triangular-shaped valley with the elevation decreasing gradually towards the South. However, hills nearly surround the valley along the southern perimeter. Meanwhile, the foothills of the San Gabriel Mountains begin just off the 111-m domain boundary to the North. As a result, the wind fields in the valley are strongly modified by local topography, and can be quite different near the surface than at higher levels.

3.2) Atmospheric inversion methodology: Bayesian framework and Simulated Annealing error assessment

3.2.1 Prior emissions errors: Simulated Annealing

The definition of the prior error covariance matrix B is most problematic because little is known about the dairy farms emissions except the presence of cows distributed in lots of small sizes. However, we assume no error correlation as it is known that groups of cows are distributed randomly across Chino. For the definition of the variances in B (i.e. diagonal terms), no reliable error estimate is available because non-agricultural emissions are suspected. The lack of error estimate directly impacts the inverse emissions, and therefore results in the generation of unreliable posterior error estimates. Instead, we develop a Monte-Carlo approach using a Simulated Annealing (SA) technique which will define the range of flux estimates for each grid point according to the observed XCH4 mole fractions. We test the initial errors in the emissions by creating random draws (i.e. random walk perturbing the emissions iteratively) with an error of about 200% compared to the expected emissions (based on the dairy cows' emissions from CARB 2015). We then generated populations of random solutions and iterated 2000 times with the SA algorithm. Overall, the SA approach allows us to explore the entire space of solutions without any prior constraint. However, we assume here that each pixel is independent, possibly causing biased estimates of CH_4 emissions. To avoid this problem, we only used the range of emission values for each pixel to construct our prior emission errors but discarded the total emissions from the SA. Instead, we performed a Bayesian inversion to produce total emissions for the area, using the diagnosed emissions from the SA as our prior emission errors.

3.2.2 Bayesian optimization using WRF-LES

Due to the absence of an adjoint model in Large Eddy Simulation mode, the inverse problem is approached with Green's functions, which correspond to the convolution of the Chino dairies emissions and the WRF-LES model response. For the two independent simulations (January 15th and 16th), 16 rectangular areas of 2 x 2 km^2 (Figure 2) are defined across the feedlots to represent the state vector (x) and therefore the spatial resolution of the inverse emissions, which correspond to the entire dairy farms area of about 8 x 8 km^2 once combined together. The 16 emitting areas continuously release a known number of CH_4 molecules (prior estimate) during the entire simulations, along with 16 individual tracers representing the 16 areas of the dairies area. The final relationship between each emitting grid-cell and each individual measurement location is the solution to the differential equation representing the sensitivity of each column measurement to the different 2 x 2 km^2 areas. The WRF-LES results are sampled every 10 minutes at each sampling location to match the exact measurement times and locations of the EM27/SUN instruments.

The inversion of the emissions over Chino is performed using a Bayesian analytical framework, described by the following equation:

$$x = x_0 + BH^T(HBH^T + R)^{-1}(y - Hx_0) \qquad (1)$$

with x the inverse emissions, x_0 the prior emissions, B the prior emission error covariance, R the observation error covariance, H the Green's functions, and y the observed column dry air mole fractions. The dimension of the state vector is 16, and we assume constant CH_4 emissions for each individual day. The column observations (here the vector *y*) correspond to the local enhancements (i.e. the contributions of local sources), the background conditions being subtracted beforehand. Here, we defined the background as the daily minimum for both days, measured by multiple sensors depending on the wind direction and the relative position of the sensor. Figure 3 shows that CH_4 background values vary between 1.83ppm to 1.832ppm, with a minimal value of 1.825ppm on January 16th. We used the averaged of each day as our final CH_4 background mixing ratios. The lack of CH_4 inventory for the LA basin and the impact of transport errors on simulated CH_4 mixing ratios are likely to produce larger uncertainties on the background conditions. For these reasons, upwind observations were used to define the background, assuming that spatial gradients across our simulation domain are small compared to atmospheric signals from Chino. The CH_4 observations used here, after subtracting the background value, correspond to local signals of about 10ppb (with a peak at 25ppb), compared to an uncertainty of about 2ppb on the background values. Two maps of 16 emission estimates are produced corresponding to the 2 x 2 km^2 areas for the two days (January 15th and 16th). A combined inversion provides a third estimate of the emissions using 10-minute average column data from both days. The metric used to select the best solutions is the Mean Absolute Error (or absolute differences) between the simulated and observed column fractions. We store the solutions exhibiting a final mismatch of less than 0.01 ppm to minimize the mismatch between observed and simulated column fractions. The optimal solution and the range of accepted emission scenarios are shown in Figure S2. The space of solutions provide a range of accepted emissions for each 2 x 2 km^2 area that can be used as a confidence interval in the inversion results. The posterior emissions from the Bayesian inversion are then compared to the confidence interval from the Simulated Annealing to evaluate our final inverse emissions estimates and the posterior uncertainties. The results are presented in Section 4.3.

Transport errors in the WRF-LES simulation can impact the accuracy of the inversion and need to be addressed in the optimization. Deng et al. (2017) studied the sensitivity of inverse emissions due to different transport scenarios. To quantify the impact of transport errors on the inverse fluxes, an ensemble approach would be necessary to propagate transport errors in the inverse solution (e.g. Evensen, 1994). Ensemble-based techniques remain computationally expensive, especially for LES simulations. Instead, we aimed at reducing the transport errors using the WRF-FDDA system to limit the errors in wind direction, wind speed, and PBL height. The improvement in model performance is significant, as demonstrated in Deng et al. (2017) reducing by half the wind speed and wind direction random errors, while removing biases in the three variables. Remaining uncertainties are described in the observation error covariance matrix ***R*** by balancing the normalized Chi-squared distance (Lauvaux et al., 2013) varying between 0.5ppb to 3ppb among all the 10-min column measurements.

4) Results

4.1) Observations of X_{CH4} and X_{CO2} in the dairy farms

Figure 3 shows the 1-minute average time series of X_{CH4} (upper panels a, b, and c) and X_{CO2} (d, e, and f) derived from the four EM27/SUN. For days with slow wind (~3 m s^{-1}), i.e. on January 15th and 16th (Figure 1, panels d, e, g and h), the maximum gradients observed between the instruments are 17 and 22 ppb (parts per billion), and 2 and 3 ppm (parts per million), for X_{CH4} and X_{CO2}, respectively. Assuming that the observed Xgas changes are confined to the PBL, gradients in this layer are about ten times larger. Gradients observed on January 15th and 16th are higher than those of X_{CH4} and X_{CO2} of 2 ppb and 0.7 ppm observed on a windy day, the 24th. The X_{CH4} and X_{CO2} variabilities captured by the instruments are due to changes in wind speed and direction, i.e., with high X_{CH4} signals when the wind blows from the dairies to the instruments. Thus, the EM27/SUN are clearly able to detect variability of greenhouses gases at local scales (temporal: less than 5 minutes, and spatial: less than 10 km) indicating that these mobile column measurements have the potential to provide estimates of local source emissions.

4.2) Estimation of fluxes with EM27/SUN column measurements

Total column measurements are directly linked to total emissions (McKain et al., 2012) and are sensitive to surface fluxes (Keppel-Aleks et al., 2012). To derive the total emissions of trace gases released in the atmosphere from a source region, the "mass balance" approach is often used. In its simplest form, the X_{CH4} fluxes can be written as in Equation 2, but this requires making assumptions about the homogeneity of the sources and wind shear in the PBL.

$$F_{X_{CH4}} = \Delta_{X_{CH4}} \frac{V(z)}{m(\theta)} SC_{air}(z) \qquad (2)$$

where $F_{X_{CH4}}$ is the flux (molecules/s.m^2), $\Delta_{X_{CH4}}$ is the X_{CH4} enhancement between the upwind and the downwind region (ppb), V is the average wind speed (ms^{-1}) from both airports, m is the distance in meter that air crosses over the dairies calculated as a function of the wind direction θ, and $SC_{air}(z)$ is the vertical column density of air (molecules/m^2). The distances that airmasses cross over the dairies (m) before reaching a receptor (EM27/SUN) are computed for each day, each wind direction, and each instrument (see complementary information section S3).

Equation 2 can be reformulated as:

$$\Delta_{X_{CH4}} = \Delta_t . \frac{F_{X_{CH4}}}{SC_{air}(z)} \qquad (3)$$

where $\Delta t = \frac{m(\theta)}{V(z)}$ is the residence time of air over the dairies (in seconds).

A modified version of this mass balance approach has been used by Chen et al. (2016) to verify that the X_{CH4} gradients measured by the EM27/SUN are comparable to the expected values measured at Chino during the CalNex aircraft campaign (Peischl et al., 2013). In Chen et al., X_{CH4} enhancements measured between upwind and two of the downwind sites on January 24th (day of constant wind direction, Figure 1 panels f and i) are compared to the expected value derived from Peischl's emission numbers, which were determined using the bottom-up method and aircraft measurements. They found that the measured X_{CH4} gradient of ~2 ppb, agrees within the low range of the 2010 value. However, this differential approach, using upwind and downwind measurements, reduces the flux estimates to only one day (January 24th), since the wind speed and direction were not constant during the other days of field measurements.

In this study, we extend the analysis of the Chino dataset using the mass balance approach on steady-wind day (on January 24th) for all the FTS instruments (i.e three downwind sites), as well as employing the other two days of measurements (January 15th, and 16th) in conjunction the WRF-LES model to derive a flux of X_{CH4} from the dairy farms. We exclude measurements from January 22nd and August 13th because of the presence of cirrus clouds during those days, which greatly reduce the precision of the column measurements. Our X_{CH4} signal measured by the FTS can be decomposed as the sum of the background concentration and the enhancements due to the local sources:

$$X_{CH4,measured} = X_{CH4,background} + \Delta_{X_{CH4}} \quad (4)$$

Gradients of X_{CH4} ($\Delta_{X_{CH4}}$) are calculated relative to one instrument for the three days. The X_{CH4} means (and standard deviations) over the three days of measurements at Chino are 1.824 (±0.003) ppm, 1.833 (±0.007) ppm, 1.823 (±0.003) ppm, and 1.835 (±0.010) ppm for the Caltech, Harvard1, Harvard2, and LANL instruments, respectively. The Harvard2 X_{CH4} mean and standard deviation are the lowest of all the observations, therefore these measurements are used as 'background'. This background site is consistent with wind directions for almost all observations, except for small periods of time on January 16th, which highlights the limitation of our method. Gradients of X_{CH4} ($\Delta_{X_{CH4}}$) for an instrument i (i.e. Caltech, Harvard1, or LANL) are the differences between each 10-minute average X_{CH4} measured by i and the simultaneous 10-minute average X_{CH4} measured by the Harvard2 instrument. Details about the residence time calculation can be found in the supplementary information section (S3). Time series of anomalies for individual measurements days are presented in Figure 4.

Assuming the background levels X_{CH4} are similar at all the instrument sites within 10 km distance and steady state wind fields, equation 3 can be written as:

$$(X_{CH4,i} - X_{CH4,Harvard2}) \propto (t_i - t_{Harvard2}).F_{X_{CH4}} \quad (5)$$

Graphical representation of equation 5 is shown in Figure 5 in which $\Delta_{X_{CH4}}$, the measured gradients by the four FTS during January 24th, are plotted as a function of Δ_t, so that the slope corresponds to a flux in ppb/s or ppt/s (parts per trillion). In this figure the slope of the blue lines (dark and light ones) represents the flux measured at Chino in previous studies (Peischl et al., 2013). These studies estimating CH_4 fluxes at Chino in 2010 reported a bottom-up value of 28 Gg/yr with a range of top-down measurements from 24 to 74 Gg/yr (Table 1). To compare these values (in Gg/yr) to the fluxes derived from column average (in ppt/s), we used Equation 6:

$$F_{col} = \frac{F\,.10^9}{a.Y.SC_{air}(z).\frac{m_g}{Na}}\,.\,10^{12} \quad (6)$$

where F_{col} is the column average flux in ppt/s, F the flux in Gg/yr, a the area of Chino (m^2), Y the number of seconds in a year, $SC_{air}(z)$ the vertical column density of air ($molecules/m^2$), m_g the molar mass of CH_4 (g/mol), and Na the Avogadro constant (mol^{-1}).

On January 24th, when the wind speed is higher than the other days (Figure 1, panels f, and i), the residence time over the dairies (Δ_t) is reduced by a factor of 30. The mean Δ_t from the closest to the furthest instruments to the upwind site are 4 minutes for Caltech (black square, Figure 5), 13 minutes for Harvard2 (green square, Figure 5), and 16 minutes for LANL (red square, Figure 5). The X_{CH4} fluxes estimated using the mean states (mass balance approach) are 4.8, 1.6, and 1.4 ppt/s for the Caltech, LANL, and Harvard2 downwind instruments. For that day, the high wind speed causes a reduction of the methane plume width across the feedlot, which may increase uncertainties on the mass-balance approach since the FTS' measurements may only detect a small portion of the total plume. Overall, the FTS network infers X_{CH4} emissions at Chino that are in the low-end of previous top-down estimates reported by Peischl et al. (2013), which is consistent with the decrease in cows and farms in the Chino area over several past years.

However, the flux estimated using the closest instrument/shortest residence time (i.e. Caltech) exceeds the value from previous studies by almost a factor of two. The other values from LANL and Harvard2, on the other hand, are lower than previous published values. This analysis demonstrates that, even with the steady-state winds day, and the simple geometry, the mass balance still has weaknesses, since it does not properly explain the differences seen among the three downwind sites. The close-in site exhibits the highest apparent emission rate possibly due to the proximity of a large CH_4 source. This exhibits delusive approximations implied by this method (i.e., spatial inhomogeneity of X_{CH4} sources completely averaged out and conservative transport in the domain) even on the "golden day" of strong steady-state wind pattern. Therefore, when investigating emissions at local scales these assumptions can be dubious and lead to errors in the flux estimates.

4.3) Spatial study of the CH_4 fluxes using WRF-LES data

Analysis of the spatial sources at Chino is developed in this section using the WRF-LES model and in section 4.4 with in situ Picarro measurements.

To map the sources of CH_4 at Chino with the model, we focus on the two days of measurements during which the wind changed direction regularly (i.e. January 15th and 16th, Figure 1 panels d, e, g and h). This provides the model information about the spatial distribution of CH_4 emissions.

4.3.1) WRF-LES model evaluation

The two WRF-Chem simulations were evaluated for both days (January 15th and 16th) using meteorological observations (Figures 6 and 7). EM27 XCH4 measurements from January 24th correspond to a constant wind direction and therefore are less suitable for mapping CH_4 emissions. The triangulation of sources requires changes in wind direction when using a static network of sensors. Starting with the larger region on the 3-km grid where WMO sondes are available (Figure 6), model verification for both days indicates that wind speed errors averaged over the domain are about 1 ms^{-1} in the free atmosphere and slightly larger in the PBL (less than

2 ms^{-1}). For wind direction, the Mean Absolute Error (MAE) is less than 20 degrees in the free atmosphere and increases approaching the surface, reaching a maximum of about 50 degrees there. In the PBL where local enhancements are located, the Mean Error (ME) remains small oscillating between 0 and 10 degrees. At higher resolutions, the comparison between observed and WRF-predicted surface wind speed (Figure 7) indicates that WRF is able to reproduce the overall calm wind conditions for both days at both WMO stations, Chino (KCNO) and Ontario (KONT). However, measurements below 1.5 ms^{-1} are not reported following the WMO standards, which limits the ability to evaluate the model over time. On January 15th at KCNO, consistent with the observations, all domains except the 3-km grid predict no surface wind speeds above 2 ms^{-1} from 16:00 – 19:00 UTC, except for one time from the 111-m LES domain. After this period, the 111-m LES domain successfully reproduces the afternoon peak in wind speed of about 3 ms^{-1}, only slightly smaller than the observed values (3.6 ms^{-1} at Chino and 3.9 ms^{-1} at Ontario airports). However, we should not expect perfect correspondence between the observations and the instantaneous LES output unless a low-pass filter is performed on the LES to average out the turbulence. On January 16th 2015, the model wind speed at KONT remained low throughout the day, in good agreement with the (unreported) measurements, and also with available observations.

4.3.2) Dispersion of tracers in LES mode: 15th and 16th January 2015

We use the January 15th 2015 case as an example showing the detail in the local winds that can be provided by the high-resolution LES domain. Prior to approximately 19:00 UTC (= 11:00 LT) a brisk easterly flow is present in the valley up to a height of 2 km; however, near the surface, a cold pool up to several hundred meters thick developed with only a very weak easterly motion. A simulated tracer released from a location near the east edge of the Chino area stays confined to the cold pool for this period (Figure 8, upper row). Solar heating causes the cold pool to break down quite rapidly after 19:00 UTC, causing the low-level wind speed to become more uniform with height (around 3 ms^{-1} from the east), and allowing the tracer to mix up to a height of about 1 km (Figure 8, middle row). Beginning around 22:00 UTC (= 14:00 LT) however, a pulse of easterly flow scours out the valley from the east, while a surge of cooler westerly flow approaches at low levels from the west, undercutting the easterly flow. By 00:00 UTC (=16:00 LT) the tracer seems to be concentrated in the cooler air just beneath the boundary of the two opposing air streams (Figure 8, lower row).

The tracer released (right columns in Figure 8) from an emitting 2 x 2 km^2 pixel shows complex vertical structures and two different regimes over the day. At 18:00 UTC, the tracer is concentrated near the surface, except toward the West with a maximum at 600 m high. At 21:00 UTC, the tracer is well-mixed in the vertical across the entire PBL, from 0 to about ~1 km, corresponding to convective conditions of daytime. At 00:00 UTC, the stability increased again, generating a low vertical plume extent with complex structures and large vertical gradients along the transect. Several updrafts and downdrafts are visible at 18:00 and 00:00 UTC, indicated by the shift in wind vectors and the distribution of the tracer in the vertical (Figure 8). These spatial structures are unique to the LES simulation, as the PBL scheme of the mesoscale model does not reproduce turbulent eddies within the PBL.

In the horizontal, convective rolls and large tracer gradients are present, with visible fine-scale spatial structures driven by the topography (i.e. hills in the South of the domain) and turbulent eddies. Figure 9 (left panel) illustrates the spatial distribution of the mean horizontal wind at the surface over the 111-m simulation domain at 18:00 UTC, just prior to the scouring out of the cold pool near a large Chino feedlot. It can be seen that the near-surface air that fills the triangular valley in the greater Chino area is nearly stagnant, while much stronger winds appear on the ridges to the south. There are some banded structures showing increased wind speed near KONT to the north of the main pool of stagnant air. Figure 9 (right panel) illustrates the wind pattern for the 18:00 UTC January 16th case. The same general patterns can be seen, with the main apparent differences being reduced wind speed along the southern high ridges, and more stagnant air in the vicinity of KONT along with elevated wind speed bands near KCNO. These results emphasize how variable the wind field structures can be from point-to-point in the valley.

4.3.3) Bayesian inversion and error assessment

We present the inverse emissions from the Bayesian analytical framework in Figures 10. The Bayesian analytical solution was computed for both days, assuming a flat prior emission rate of 2150 mol/km^2/hour corresponding to a uniform distribution of 115000 dairy cows over 64 km^2

emitting methane at a constant rate of 150 kg of CH_4 per year (CARB 2015), plus 18 kg annually per cow from dry manure management assumed to be on-site (Peischl et al., 2013). The colored areas in Figure 10 represent the ranges of solutions defined by the Simulated Annealing (SA) analysis, for the two days of the campaign (in blue and green). The Bayesian averages agree well with the SA estimates, with high confidence for half of the pixels (1, 2, 3, 4, 8, 13, 15, and 16), and lower confidence for the other pixels. High values coincide with high confidence, which confirms the fact that large signals constrain the inverse solution better. This would possibly suggest that only the largest emissions could be attributed with sufficient confidence using these tools.

The spatial distribution of the emissions is shown in Figure 13, which directly corresponds to the pixel emissions presented in Figure 10. The largest sources are located in the southern part of the dairy farms area, and in the northeastern corner of the domain. Additional interpretation of these results is presented in the following section. The combination of the results from two dates (January 15th and 16th) is necessary in order to identify the whole southern edge of the feedlots as a large source. Sensitivity results are presented in the discussion and in the supplementary information section (S4 and S5). The triangulation of sources performed by the inversion produced consistent results using different configurations of EM27 sensors for each day. Inversion results cover the entire domain with all wind directions being observed over the two days (cf. Figure 1, panels d, e, g, and h). Additional sensitivity tests were performed to evaluate the impact of instrument errors, introducing a systematic error of 5 ppb in X_{CH4} measured by one of the EM27/SUN. The posterior emissions increased by 3-4 Gg/year for a +5ppb bias almost independent of the location of the biased instrument. This represents ~10% of the total emission at Chino.

4.4) Spatial study of the CH_4 emissions at Chino using Picarro measurements

During the field campaign in January 2015, in situ measurements of CH_4, CO_2, as well as $\delta^{13}C$ are collected simultaneously with a Picarro instrument at the same site as the LANL EM27/SUN. Fossil-related CH_4 sources, such as power plants, traffic, and natural gas, emit CH_4 with an isotopic depletion $\delta^{13}C$ ranging from -30 to -45 ‰, whereas biogenic methane sources, such as those from enteric fermentation and wet and dry manure management in dairies and feedlots emit in the range of −65 to −45 ‰ (Townsend-Small et al., 2012). During the January 2015 campaign, the δ^{13}C at Chino ranged from -35 to -50 ‰, indicating a mixture of fossil and biogenic sources respectively. Most of the air sampled included a mixture of both sources. However, the measurements with the highest CH_4 concentrations had lowest δ^{13}C signatures, suggesting that the major CH_4 enhancements measured by the Picarro instrument can be attributed to the dairy farms and not the surrounding urban sources.

On January 16th and 22nd, the Picarro and the LANL EM27/SUN were installed at the southwest side of the largest dairies in Chino (red pin, Figure 1b), near a wet lagoon that is used for manure management (< 150 m away). For these days, the Picarro measured enhancements of CH_4 up to 20 ppm above background concentrations, demonstrating that the lagoon is a large source of CH_4 emissions in the Chino area. The location of the lagoon was identified and verified by satellite imagery, visual inspection, and also with measurements from the second Picarro instrument deployed in the field on January 15th, 2015. With this instrument, CH_4 spikes up to 23 ppm were observed near the wet manure lagoon. The measurements from both Picarros and the LANL EM27/SUN instrument near the lagoon suggested that this is a significant local source of CH_4 emissions in the Chino area.

As opposed to column measurements, Picarro measurements are very sensitive to the dilution effect of gases in the PBL. With a low boundary layer, atmospheric constituents are concentrated near the surface, and the atmospheric signal detected by the in situ surface measurements is enhanced relative to the daytime, when the PBL is fully developed. For this reason, additional Picarro measurements were made at night on August 13th 2015, when the PBL height is minimal. Between 04:00 to 07:00 (LT), we performed Picarro measurements at different locations in Chino, to map the different sources of CH_4 and verify that the large sources observed in January, such as the lagoon, are still emitting in summer. Figure 11 shows the scatter plot of one minute-average anomalies of CH_4 (Δ_{CH4}) versus CO_2 (Δ_{CO2}), colored by the δ^{13}C values, measured by the Picarro on the night of August 13th 2015. During that night, the isotopic range of δ^{13}C in sampled methane range from -45 ‰ to -65 ‰. These low δ^{13}C values are consistent with the expectation that the sources of CH_4 in the Chino area are dominated by biogenic emissions from dairy cows.

In the feedlots (side triangles, Figure 11), Δ_{CH4} and Δ_{CO2} are well correlated (r^2 = 0.90), because cows emit both gases (Kinsman et al., 1995). The observed $\Delta_{CH4}/\Delta_{CO2}$ emission ratio, 48 ± 1.5 ppb/ppm, is in good agreement with a previous study measuring this ratio from cow's breath (Lassen et al., 2012). Measurements obtained at less than one meter away from cows (circles, Figure 11), had the lowest the $\delta^{13}C$ observed, ~-65 ‰, and these points scale well with the linear correlation observed during the survey. This confirms that the emission ratio derived surveying the feedlots is representative of biogenic emissions related to enteric fermentation. Measurements obtained next to the lagoon (diamond marks, Figure 11), the $^{12}CH_4$ concentrations enhanced by up to 40 ppm above background levels observed that night, while the relative enhancement of CO_2 was much smaller. This extremely large CH_4 enhancement relative to CO_2 indicates a signature of CH_4 emissions from wet manure management (lagoon), confirming that there is significant heterogeneity in the CH_4 sources within the Chino dairy area.

5) Discussion

The fluxes derived by the FTS observations and the WRF-LES inversions, as well as previous reported values are summarized in Table 1.

The top-down CH_4 estimate using FTS observations in Chino provide a range of fluxes from 1.4 to 4.8 ppt/s during January 2015 (Table 1), which are on the lower-end of previously published estimates. These values of CH_4 flux estimates for January 2015 based on the FTS measurements are consistent with the decrease in cows in Chino over the past several years as urbanization spreads across the region.

Considering the decrease of dairy cows number by ~20% from 2010 to 2015, and using the emission factor of 168 kg/yr per head (CARB 2015 inventory: enteric fermentation + dry manure management), the CH_4 flux associated with dairy cows at Chino decreased from 2.0 to 1.7 ppt/s, which agrees well with our low flux estimates derived from FTS observations. However, fluxes derived using the simple mass balance approach differs from each other, exhibiting the limitations of this method, even on a "golden day" (steady-state wind day on January 24th). The WRF-LES inversions (Figures 10 and 12) and mobile in situ measurements with the Picarro instrument (Figure 11) indicate that the CH_4 sources are not homogeneous within this local area. In addition, wind measurements from the two local airports typically disagree regarding the direction and speed (Figure 1, panels d, e, f, g, h, and i), and the WRF-LES tracer results indicate non-homogeneous advection of tracers (Figure 8, right panels).

Figure 12 shows the map of the *a posteriori* X_{CH4} fluxes (mean of January 15th and 16th runs) from the WRF-LES simulations, superimposed on a Google earth map, with the location of dairy farms represented by the red areas. The domain is decomposed into 16 boxes (Section 3.2), in which the colors correspond to the *a posteriori* emissions derived from the WRF-LES inversions. Red (blue) colors of a box mean more (less) CH_4 emissions compared to the *a priori* emissions, which corresponds to the dairy cow emissions contained in the CARB 2015 inventory (emission factor multiplied by the number of cows). Results of the inversion exhibit more CH_4 emissions at the South and the Northeast parts of the domain, and emissions corresponding to dairy cows in the center of the area.

The higher CH_4 emissions from the southwestern part of the domain can be attributed to the wet manure lagoon (yellow pin, Figure 12) in January 2015. During the night of August 13th 2015, Picarro measurements confirmed that the lagoon was still wet and emitted a considerable amount of CH_4 relative to CO_2 (Figure 12). The second mobile Picarro instrument from JPL was deployed on January 15th 2015 and measured CH_4 spikes up to 23 ppm near the wet manure lagoon. The WRF-LES model also suggests higher methane fluxes in these regions (red boxes, Figure 12). The CARB 2015 inventory estimates that manure management practices under wet (e.g. lagoon) conditions emit more CH_4 than the dairy cows themselves: 187 kg CH_4 cow^{-1} yr^{-1} from wet manure management, 18 kg CH_4 cow^{-1} yr^{-1} from dry management practices, and 150 kg CH_4 cow^{-1} yr^{-1} from enteric fermentation in the stomachs of dairy cows. Therefore, we expect measurements in which the lagoon emissions were detected by our instruments will lead to higher methane fluxes in the local region, compared to measurements detecting emissions from enteric fermentation in cows alone. Bottom-up emission inventory of CH_4 is 2 times higher when considering wet lagoons (Wennberg et al., 2012) instead of dry management practices (Peischl et al., 2013) at Chino (Table1). The location and extent of wet lagoons in the Chino region is not expected to be constant with time and could be altered due to changing land use and future development in the area. Bottom-up estimates of CH_4 emissions from dairies in the Chino region could be further improved if the extent and location of wet manure lagoons were well-known.

The WRF-LES model also suggests higher methane fluxes in the Southeast (red boxes, Figure 13). No dairy farms are located in these areas, but an inter-state pipeline is located nearby, thus these CH_4 enhancements could be attributed to natural gas. The $^{13}CH_4$ Picarro measurements indicate the Chino area is influenced by both fossil- and biogenic- related methane sources. A recent study has suggested the presence of considerable fugitive emissions of methane at Chino (http://www.edf.org/climate/methanemaps/city-snapshots/los-angeles-area), probably due to the advanced age of the pipelines. Natural gas leaks in the Chino area were not specifically targeted during the time of this field campaign and cannot be confirmed using available data. This possibility should thus be confirmed by future studies.

In addition to possible fugitive emissions at Chino, the inversion also predicts higher CH_4 flux in the Northeastern region of the study domain, which is in the vicinity of a power plant that reportedly emits a CH_4 flux roughly equivalent of one cow per year (only including enteric fermentation) (http://www.arb.ca.gov/cc/reporting/ghg-rep/reported_data/ghg-reports.htm). Further analysis and measurements of fossil methane sources in the Chino area would help verify potential contributions from fossil methane sources, including power plants and/or fugitive natural gas pipeline emissions.

Overall, FTS and in situ Picarro measurements, as well as WRF-LES inversions, all demonstrate that the CH_4 sources at Chino are heterogeneous, with a mixture of emissions from enteric fermentation, wet and dry manure management practices, and possible additional fossil methane emissions (from natural gas pipeline and power plants). The detection of CH_4 emissions in the Chino region and discrepancies between top-down estimates could be further improved with more FTS observations and concurrent in situ methane isotopes measurements combined with high-resolution WRF-LES inversions. This would improve the spatial detection of the CH_4 emissions at Chino, in order to ameliorate the inventories among the individual sources in this local area.

6) Summary and conclusions

In January 2015, four mobile low-resolution FTS (EM27/SUN) were deployed in a ~6 x 9 km area in Chino (California), to assess CH_4 emissions related to dairy cows in the SoCAB farms. The network of column measurements captured large spatial and temporal gradients of greenhouses gases emitted from this small-scale area. Temporal variabilities of X_{CH4} and X_{CO2} can reach up to 20 ppb and 2 ppm, respectively, within less than a 10-minute interval with respect to wind direction changes. This study demonstrate that these mobile FTS are therefore capable of detecting local greenhouses gas signals and these measurements can be used to improve the verification of X_{CO2} and X_{CH4} emissions at local scales.

Top-down estimates of CH_4 fluxes using the 2015 FTS observations in conjunction with wind measurements are 1.4-4.8 ppt/s, which are in the low-end of the 2010 estimates (Peischl et al., 2013), consistent with the decrease in cows in the Chino area. During this campaign, FTS measurements were collected in close proximity to the sources (less than a few km) in order to capture large signals from the local area. The main advantage of this type of deployment strategy is to better constrain the emissions, while avoiding vertical mixing issues in the model with the use of column measurements in the inversion (Wunch et al., 2011). Therefore, the model transport errors, which often limit the capacity of the model flux estimates, are considerably reduced. However, the close proximity of the measurements to the sources makes the assumptions about homogeneity of the sources and wind patterns questionable.

The FTS and the Picarro measurements detected various CH_4 signatures over Chino, with extreme CH_4 enhancements measured nearby a wet lagoon (Picarro and FTS measurements enhanced by 40 ppm CH_4 and 60 ppb X_{CH4}, respectively) and possible fugitive fossil-related CH_4 emissions in the area (indicated by higher $\delta^{13}C$ values than expected from biogenic emissions alone).

Wind speed and direction measurements derived from the two local airports (less than 10 km apart), as well as the WRF meteorological simulations at different FTS sites, differ greatly with each other, suggesting that an assumption of steady horizontal wind can be improved upon in the use of the mass balance approach in our study. This may explain some differences between the CH_4 flux estimates from the mass balance approach and the Bayesian inversion.

This study demonstrates the value of using mobile column measurements for detection of local CH_4 enhancements and the estimation of CH_4 emissions when these measurements are combined with modeling. High-resolution (111 m) WRF-LES simulations were performed on two dates, constrained by four column measurements each day, to map the heterogeneous CH_4 sources at Chino. The average a posteriori flux over the domain is 3.2 ppt/s when only considering the boxes in the center of the domain, and 4.7 ppt/s when all the boxes are averaged. A major emitter (a wet manure lagoon) was identified by the inversion results, and is supported by in-situ $^{13}CH_4$ measurements collected during the campaign. The CH_4 flux estimates are within the range of the top-down mass balance emissions derived with the four FTS and estimates reported by Peischl et al. 2013 (i.e., 2.1 to 6.5 ppt/s), showing that column measurements combined with high resolution modeling can detect and be used to estimate CH_4 emissions.

The instrumental synergy (mobile in situ and column observations) coupled with a comprehensive high-resolution model simulations allow estimation of local CH_4 fluxes, and can be useful for improving emission inventories, especially in a complex megacity area, where the different sources are often located within small areas.

This study highlights the complexity of estimating emissions at local scale when sources and wind can exhibit heterogeneous patterns. Long term column observations and/or aircraft eddy covariance measurements could improve estimations.

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study	time of study	sources	CH_4 emission (Gg/year)	CH_4 emission (ppt/s)
Peischl et al., 2013	2010	inventory (dry manure + cows)	28	2.5
Peischl et al., 2013	2010	aircraft measurements	24-74	2.1-6.5
Wennberg et al., 2012	2010	inventory (wet manure + cows)*	66	5.8
CARB 2015	2015	inventory (dry manure + cows)	19	1.7
Chen et al.	2015	FTS measurements only	19-32	2.4-3.3
This study	2015	FTS measurements only	16-55	1.4-4.8
This study	2015	WRF inversions	36-54	3.2-4.7

* Value reported for the SoCAB, apportioned for Chino in this study.

Table1: Emissions of CH_4 at Chino.

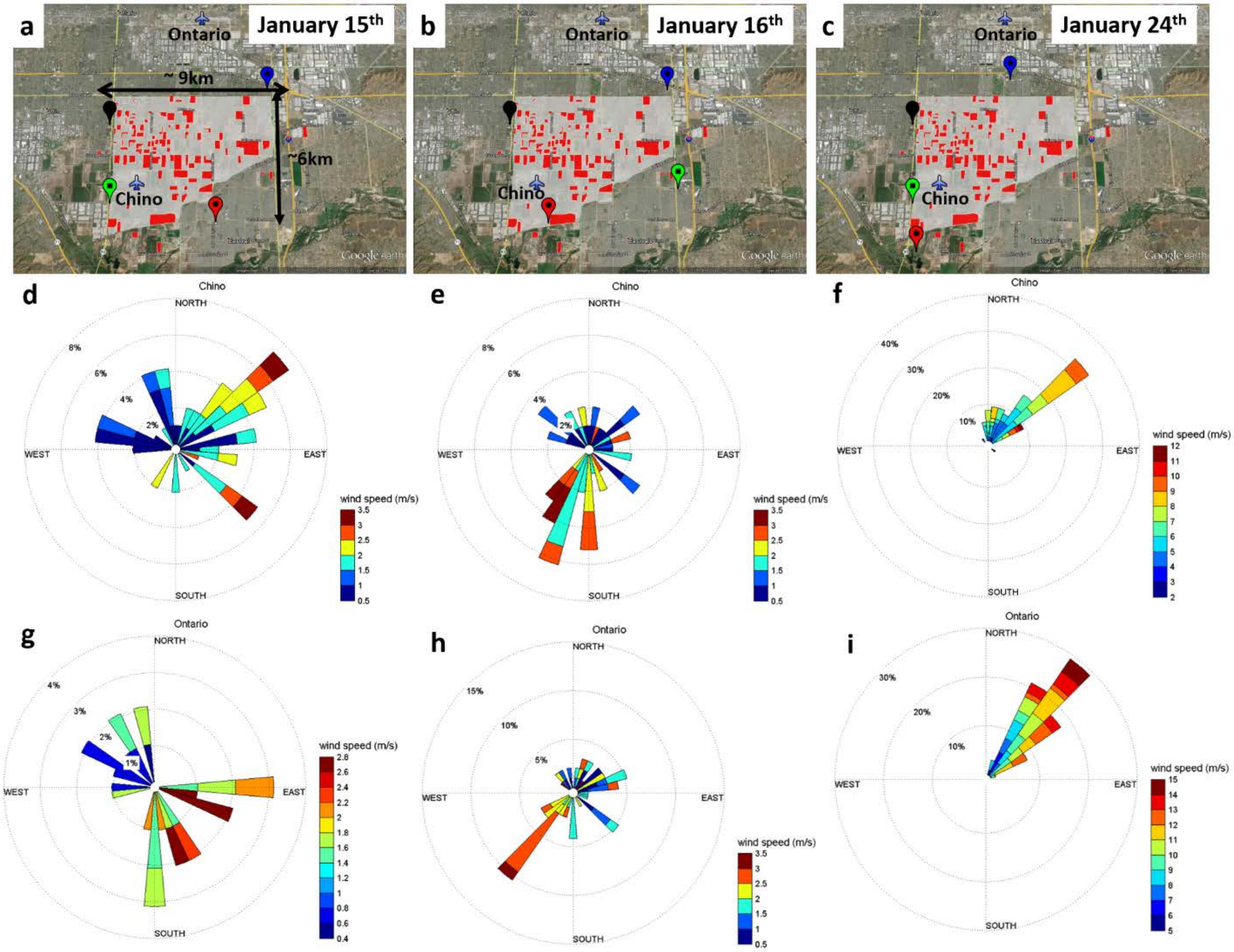

Figure 1: Three different days of measurements during the field campaign at Chino (~9 x 6 km) on the 15th, 16th, and 24th of January 2015. Upper panels (a, b, and c) show the chosen locations of the four EM27/SUN (black, red, green, and blue pins correspond to the Caltech, LANL, Harvard1, and Harvard2 instruments, respectively). The red marks on the map correspond to the dairy farms. Lower panels show wind roses of ten-minute average of wind directions and wind speeds measured at the two local airports (at Chino on panels d, e, and f, and at Ontario on panels g, h, and i). Map provided by GOOGLE EARTH V 7.1.2.2041, US Dept. of State Geographer, Google, 2013, Image Landsat, Data SIO, NOAA, U.S, Navy, NGA, and GEBCO.

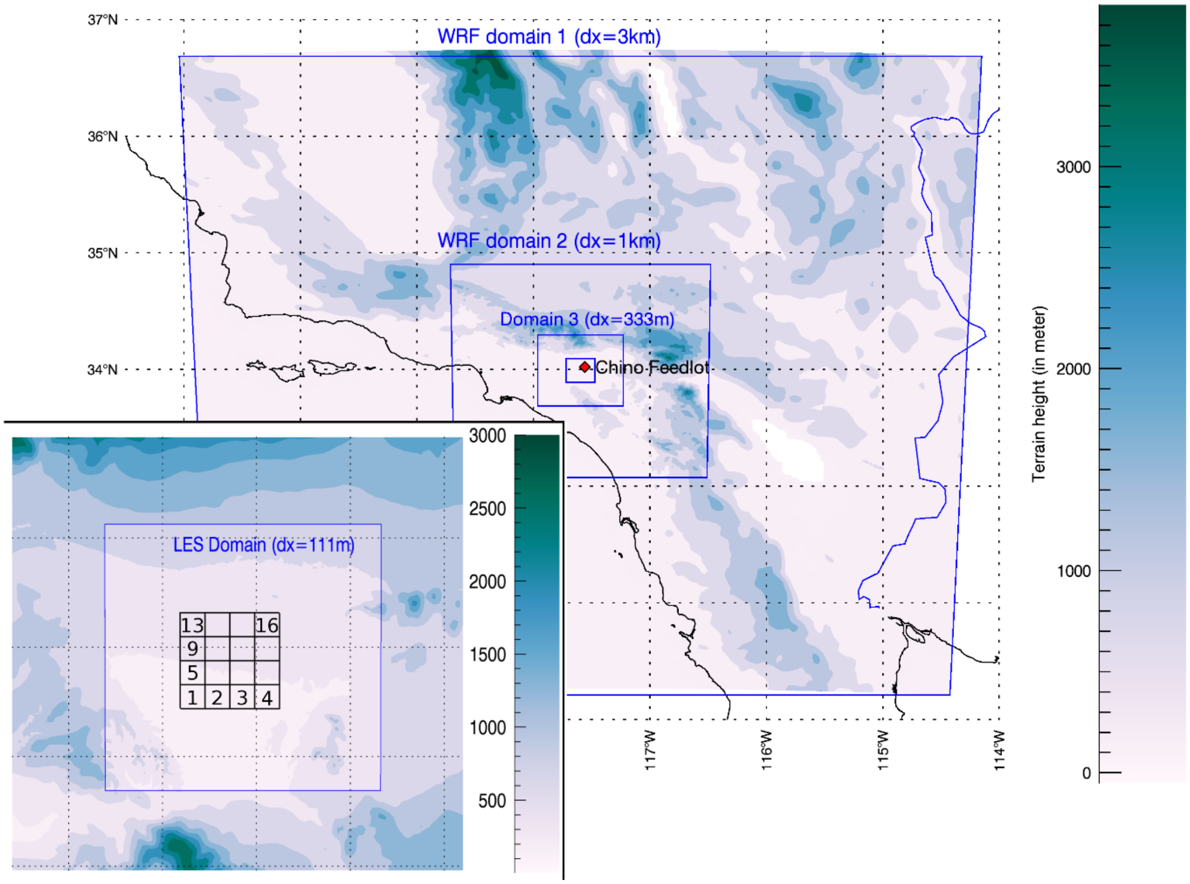

Figure 2: WRF-Chem simulation domains for the 4 grid resolutions (3-km; 1-km; 333-m; 111-m), with the corresponding topography based on the Shuttle Radar Topographic Mission Digital Elevation Model at 90-m resolution). The 16 rectangular areas (2 x 2 km^2) are shown on the LES domain map and numerate by pixel numbers (Figure 10).

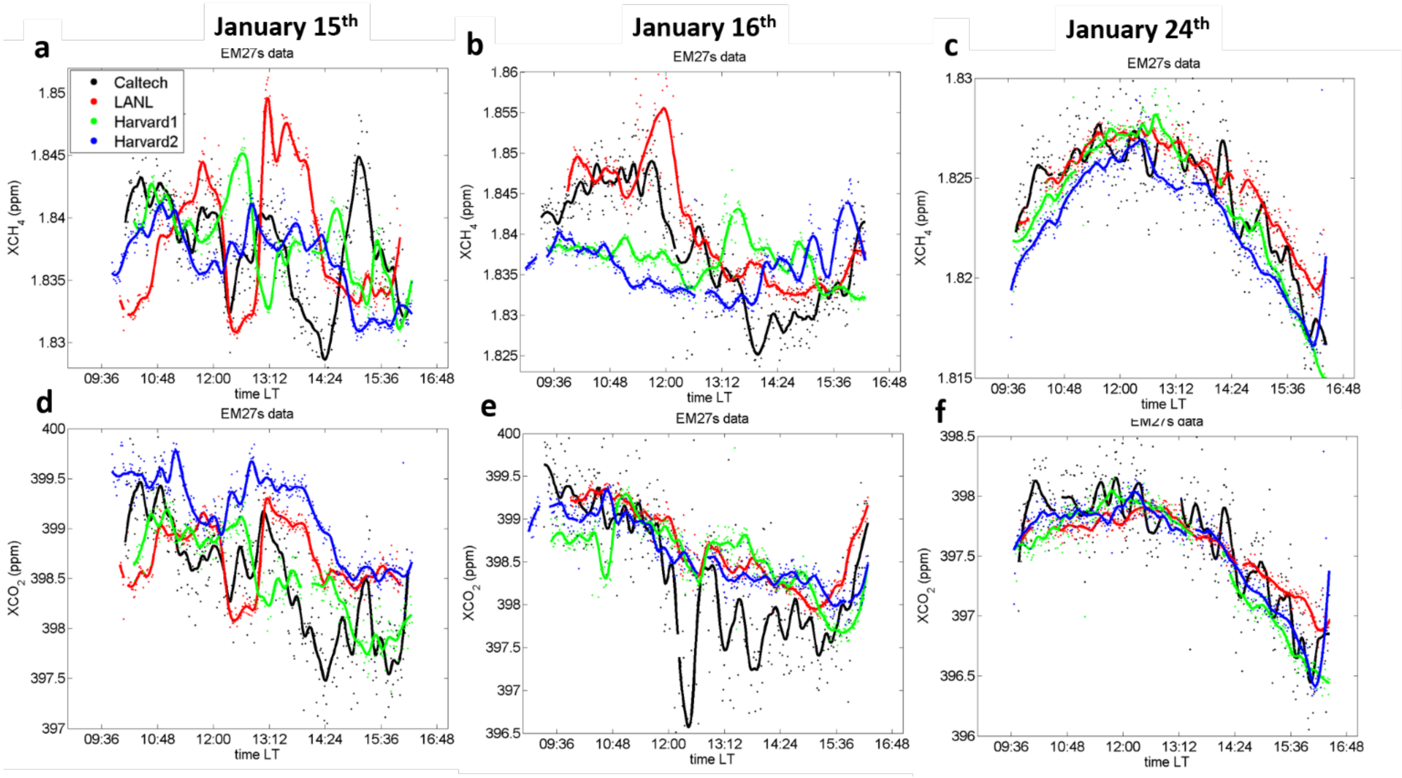

Figure 3: One minute-average time series of X_{CH4} (upper panels a, b, and c) and X_{CO2} (lower panels d, e, and f) measured by the four EM27/SUN (black, red, green, and blue marks correspond to the Caltech, LANL, Harvard1, and Harvard2 spectrometers, respectively).

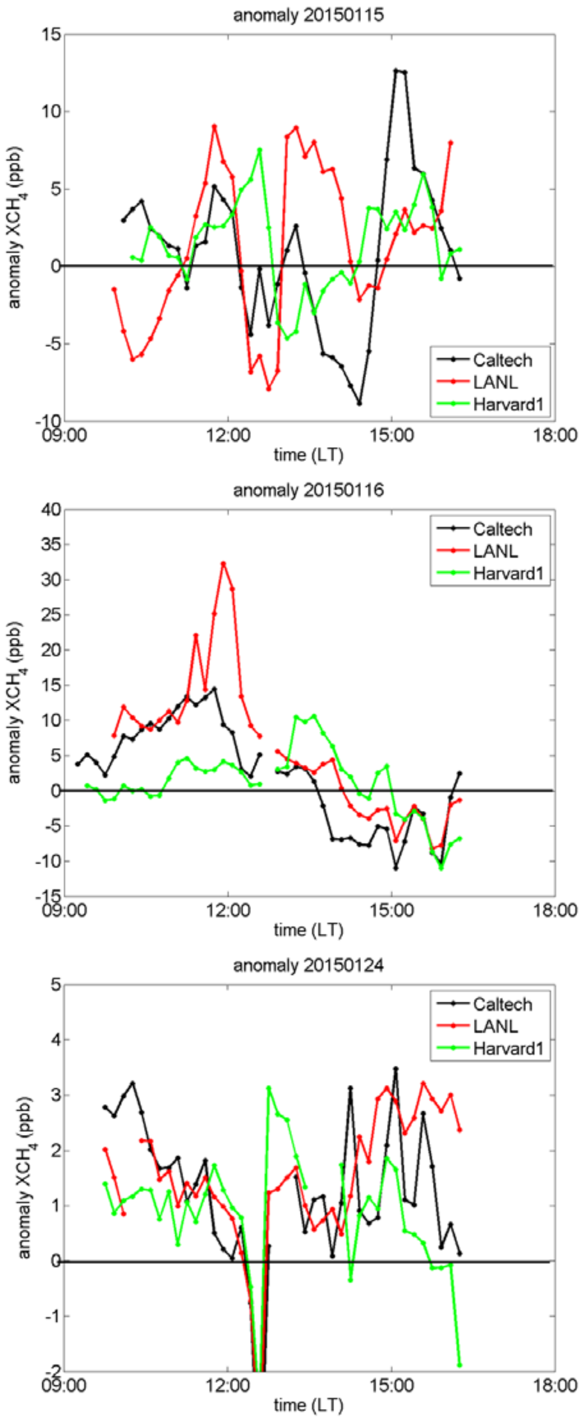

Figure 4: Time series of the 10-minute average X_{CH4} anomaly (Δ_{XCH4}, in ppb) computed relative to the Harvard2 instrument for January 15th (upper panel), January 16th (middle panel), and on January 24th 2015 (lower panel).

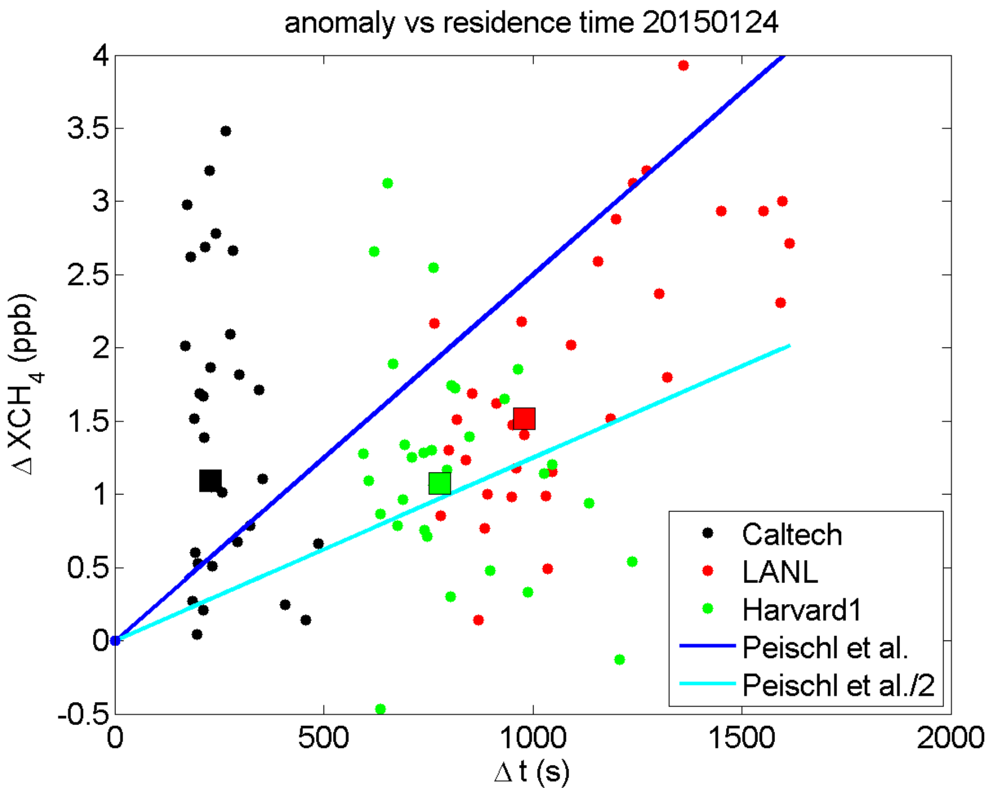

Figure 5: Estimated fluxes using FTS observations on January 24th. The 10-minute anomalies (relative to the Harvard 2 instrument) are plotted against the time that airmass travelled over the dairies, so that the slopes are equivalent to X_{CH4} fluxes (in ppb/s, equation 5). The blue (and cyan) line represents the fluxes (and half of the value) estimated at Chino in 2010 (Peischl et al., 2013). The squares are the medians of the data which correspond to the estimated fluxes using the FTS observations (in black, red and green for the Caltech, LANL, and Harvard2 instruments).

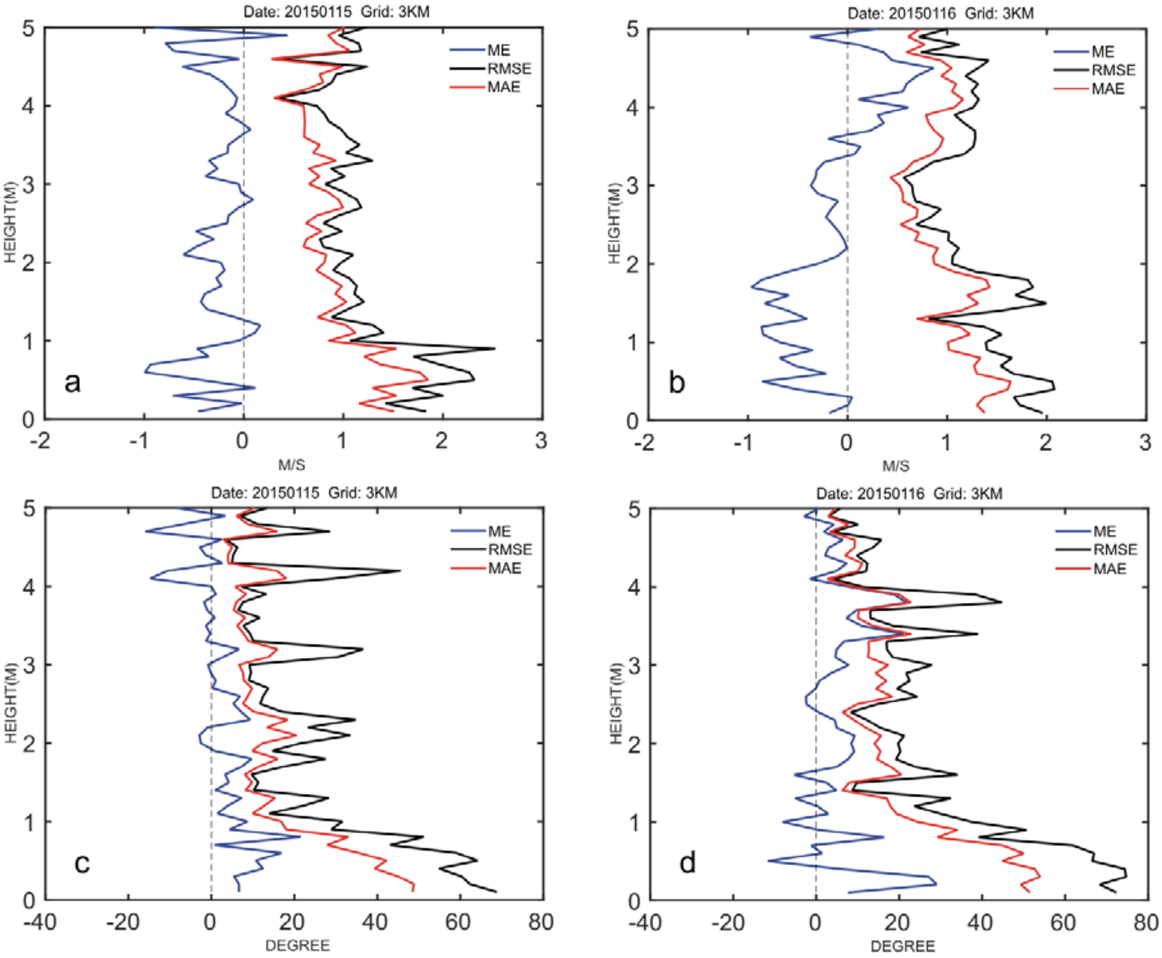

Figure 6: Vertical profiles of mean horizontal wind velocity errors (upper row) and direction (lower row) averaged from the WMO radiosonde sites available across the 3-km domain, with the Mean Absolute Error (in red), the Root Mean Square Error (in black), and the Mean Error (in blue). Only measurements from 00z radiosondes were used in the evaluation.

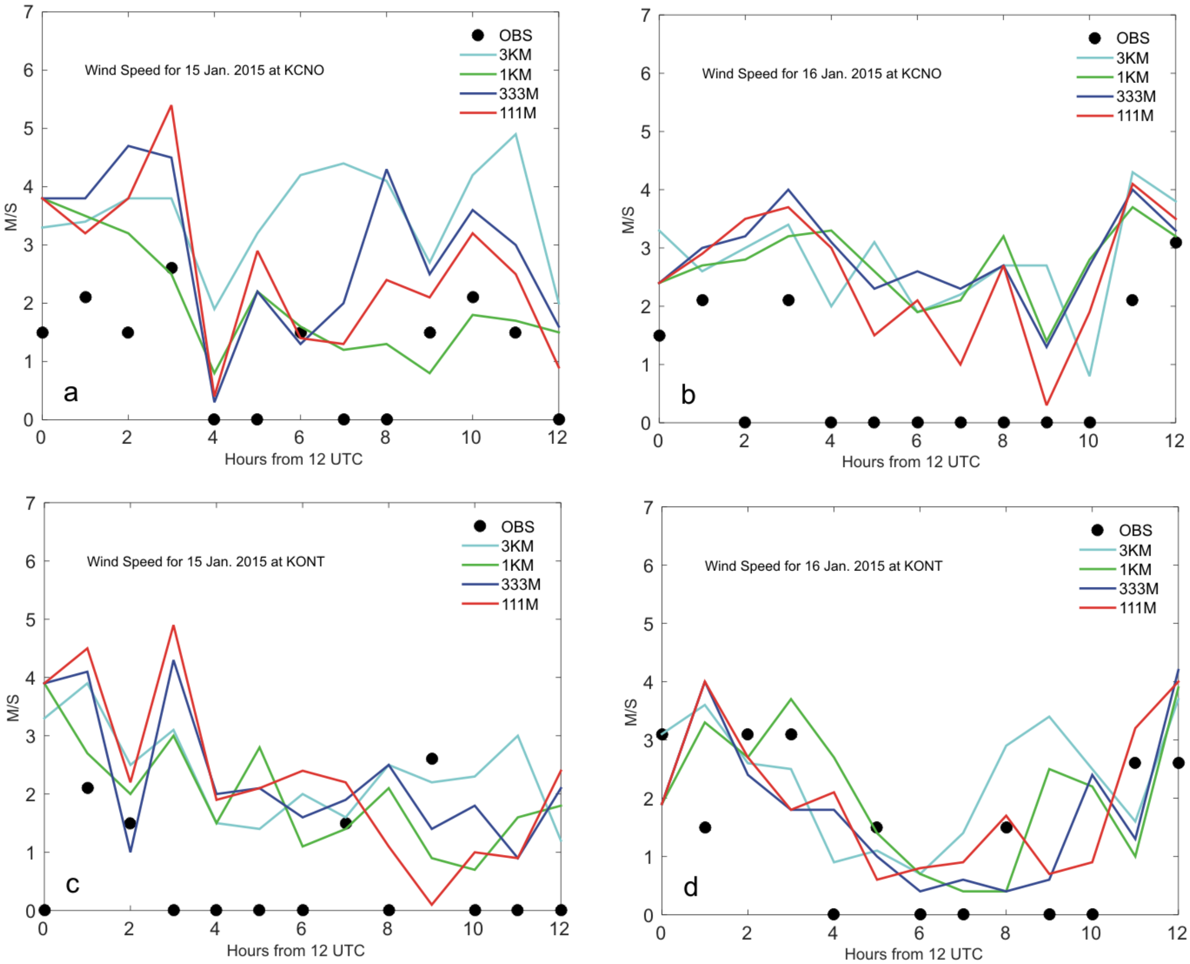

Figure 7: Mean horizontal 10-meter wind velocity in ms^{-1} measured at Chino (KCNO) and Ontario (KONT) airports for January 15th and 16th (black circles) compared to the simulated wind speed for different resolutions using WRF hourly-averaged results. When black circles indicate zero, the wind velocity measurements are below the WMO minimum threshold (i.e. 1.5 m/s).

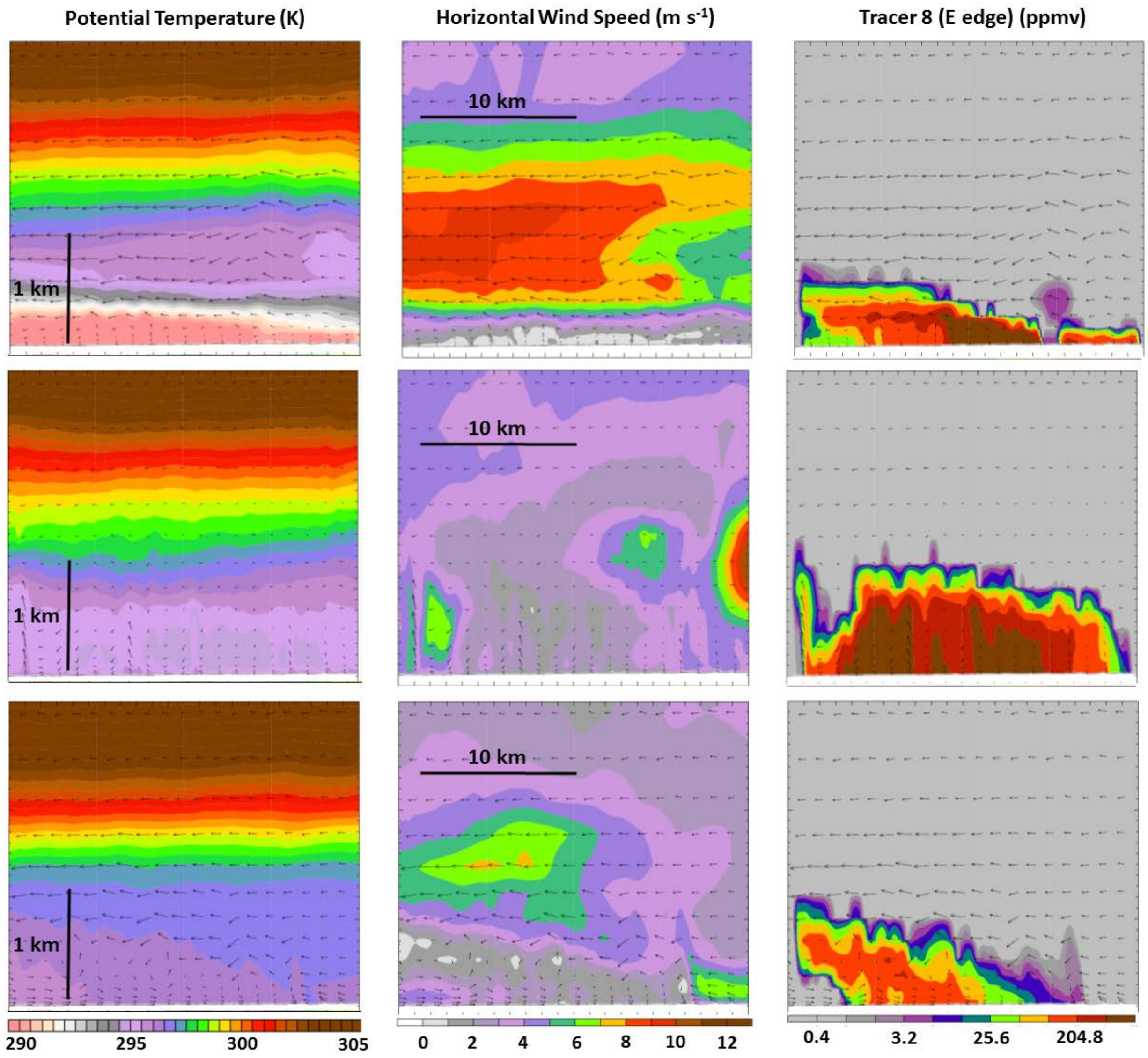

Figure 8: Vertical transects across the 111-m West-East WRF-LES simulation domain (pixels 5, 6, 7, and 8) at 18:00 UTC of January 15th (upper row), 21:00 UTC (middle row), and 00:00 UTC (lower row). From left to right, simulated data are shown for potential temperature (in K, left column), mean horizontal wind speed and direction (in ms^{-1} and degree, middle column), and passive tracer concentration released from an eastern pixel of the emitting area (pixel 5, right column), to illustrate the relationship between the three variables.

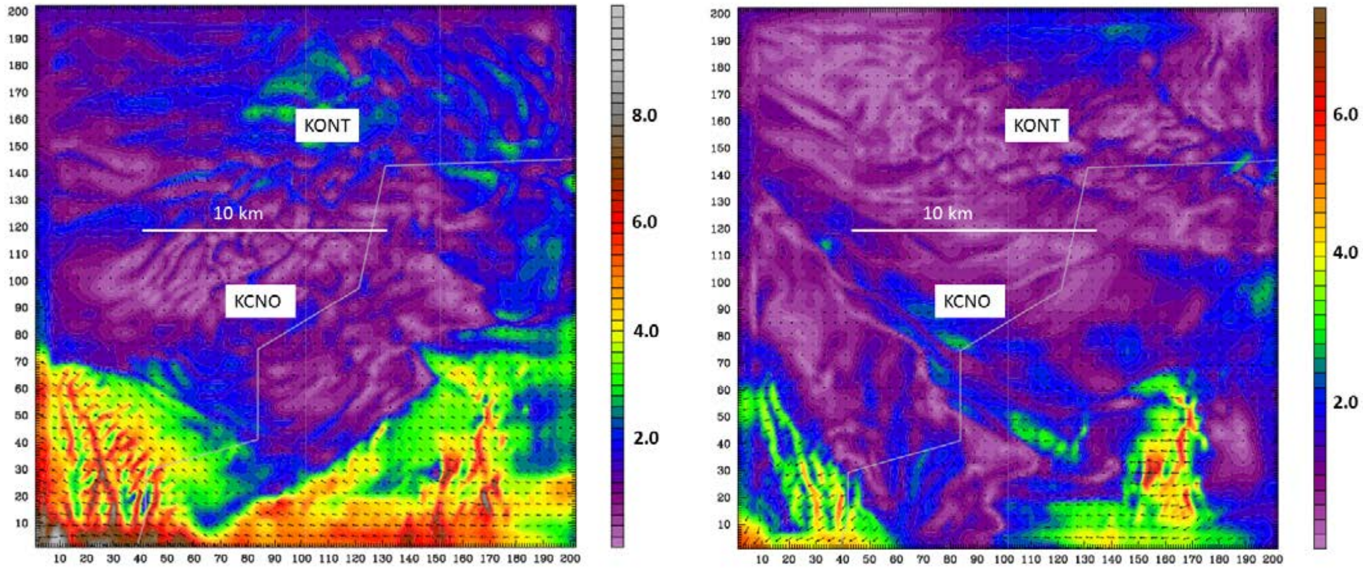

Figure 9: Mean horizontal wind field (in ms^{-1}) in the first level of the domain at 111-m resolution simulated by WRF-LES for January 15th (left panel), and January 16th 2015 (right panel), at 18:00 UTC. High wind speeds were simulated over the hills (southern part of the domain) whereas convective rolls, corresponding to organized turbulent eddies, are visible in the middle of the domain (i.e. over the feedlots of Chino), highlighting the importance of turbulent structures in representing the observed horizontal gradients of CH_4 concentrations. The locations of the Chino (KCNO) and Ontario (KONT) airports and the counties border (white line) are indicated.

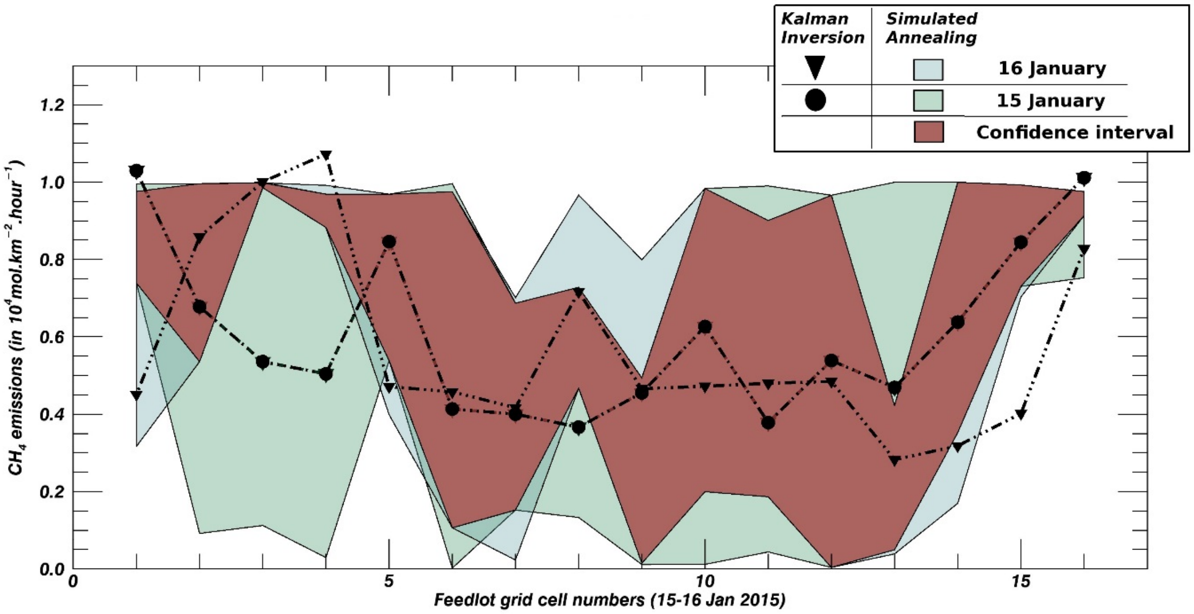

Figure 10: Emissions of CH_4 (in mol/km^2/hour) for the 16 pixels (2 x 2 km^2 shown In Figure 2) describing the dairies for both days, i.e. the 15th and 16th of January 2015. The Bayesian mean emissions are shown in black (triangles and circles) whereas the colored areas represent the accepted range of solutions using the Simulated Annealing technique (see section 3.2).

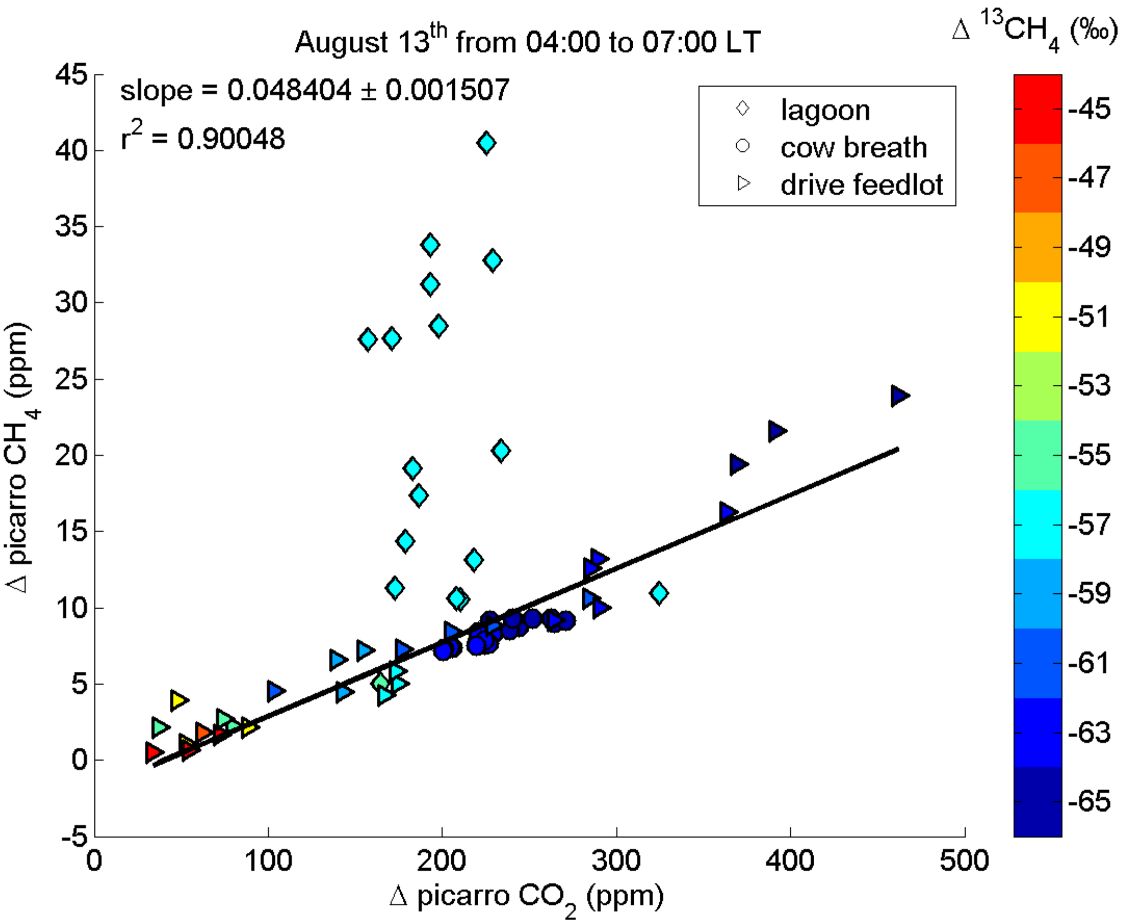

Figure 11: Scatter plot of one minute-average anomalies (from the 5 minutes smoothed) of CH_4 versus CO_2, color coded by the delta CH4 values, measured by the Picarro on August 13th from 04:00 to 07:00 (LT).

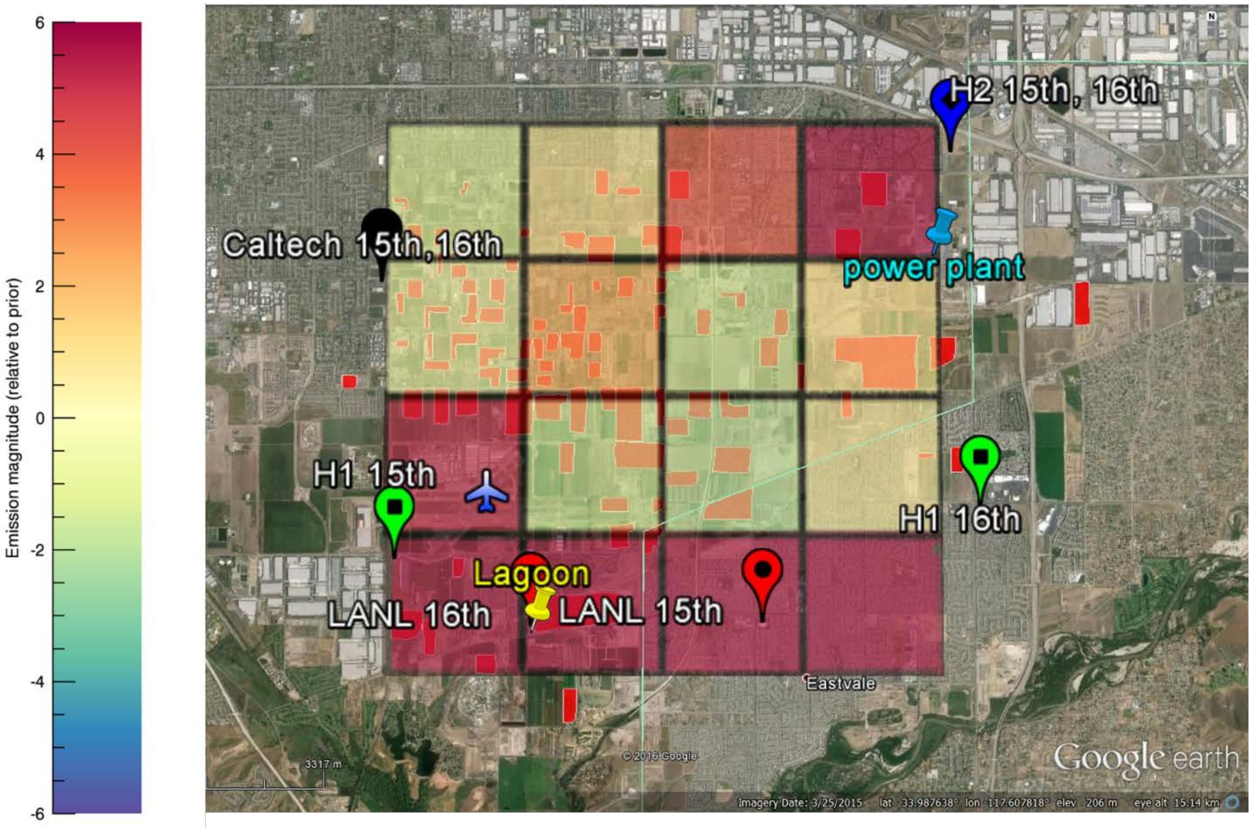

Figure 12: Map of the *a posteriori* X_{CH4} fluxes (mean of January 15th and 16th runs) from the WRF-LES simulations, superimposed on a Google earth map, where the dairy farms are represented by the red areas as shown in Figure 1. The domain is decomposed in 16 boxes (2km x 2km), in which the colors correspond to the *a posteriori* emissions from the WRF-LES inversions. Red (blue) colors mean more (less) CH_4 emissions than dairy cows in that box. The locations of the lagoon (yellow pin) and the power plant (blue pin) are also added on the map. Map provided by GOOGLE EARTH V 7.1.2.2041, US Dept. of State Geographer, Google, 2013, Image Landsat, Data SIO, NOAA, U.S, Navy, NGA, and GEBCO.