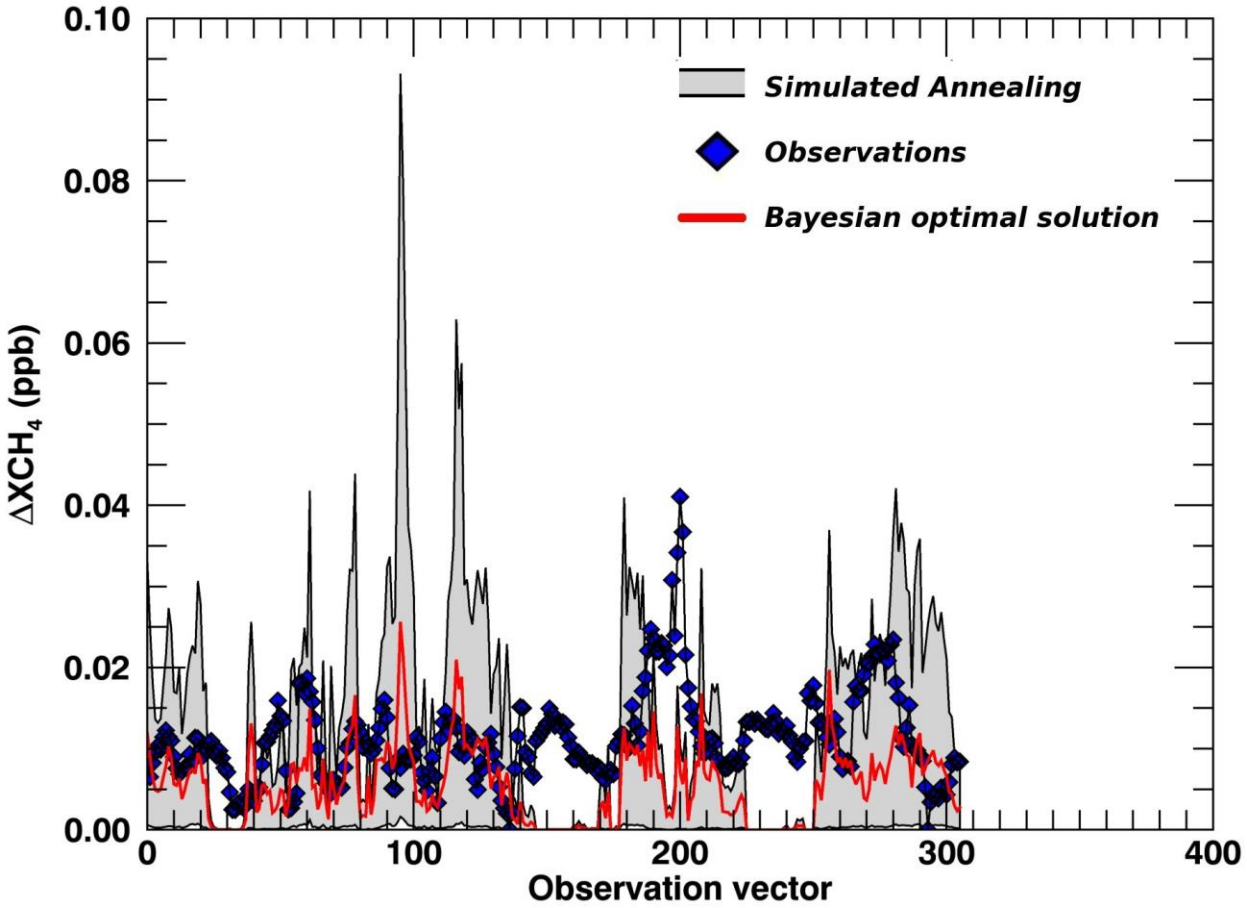
Supplement information:

S1: Coupling between mesoscale and LES model

The GFS analyses are far too coarse-scale to contain turbulent eddy structures, but since no PBL eddies on scales larger than 111 m are expected during the pre-dawn PBL at initialization anyway, there should be no inconsistency with using these fields to initialize all model grids. As the day progresses and the PBL is heated from below, either random or topographically generated perturbations will gradually develop into realistic PBL eddies in the LES, while the PBL parameterizations on the coarser domains should at least predict realistic evolution of the temperature and wind fields. These coarser-domain fields drive the lateral boundary conditions of the LES and help ensure the LES has consistent large-scale meteorology, while within the LES PBL turbulence develops in the flow as it enters the domain. Only the meteorology has been nested while CH_4 tracers are simulated at 111-m resolution only. The LES domain is made large enough such that realistic turbulent eddies have a chance to develop in the inflow before they reach the region of interest. 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.



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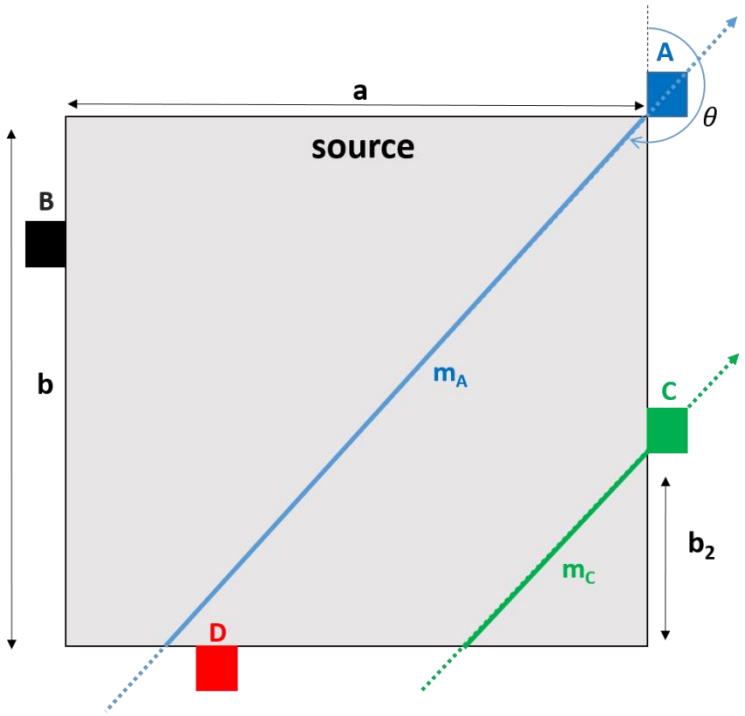
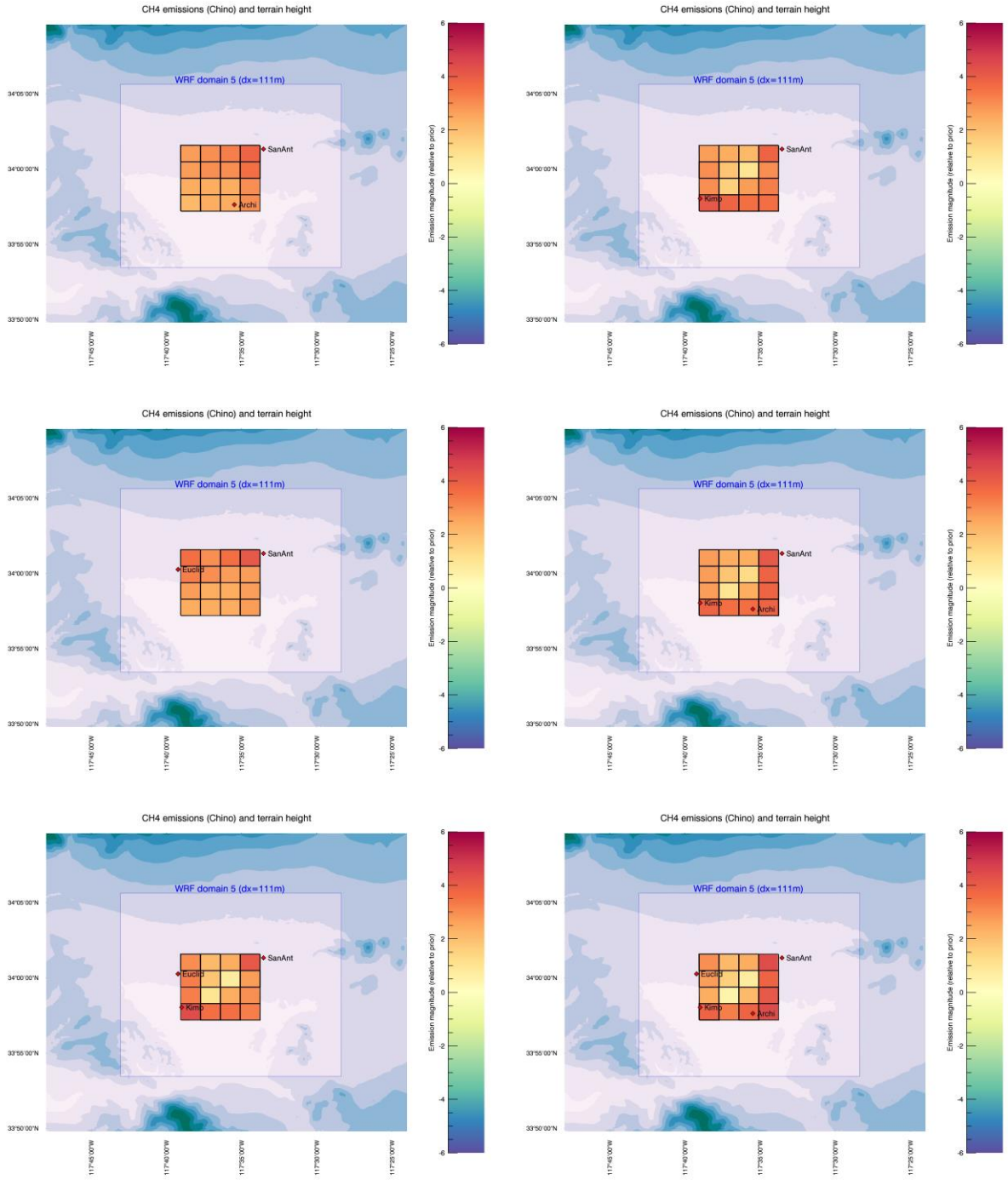
22 S2: Comparison of the observed CH_4 column enhancements (blue diamond), the Simulated
23 Annealing accepted solutions (in grey), and the optimal solution from the Bayesian inversion
24 (red line) in ppm.

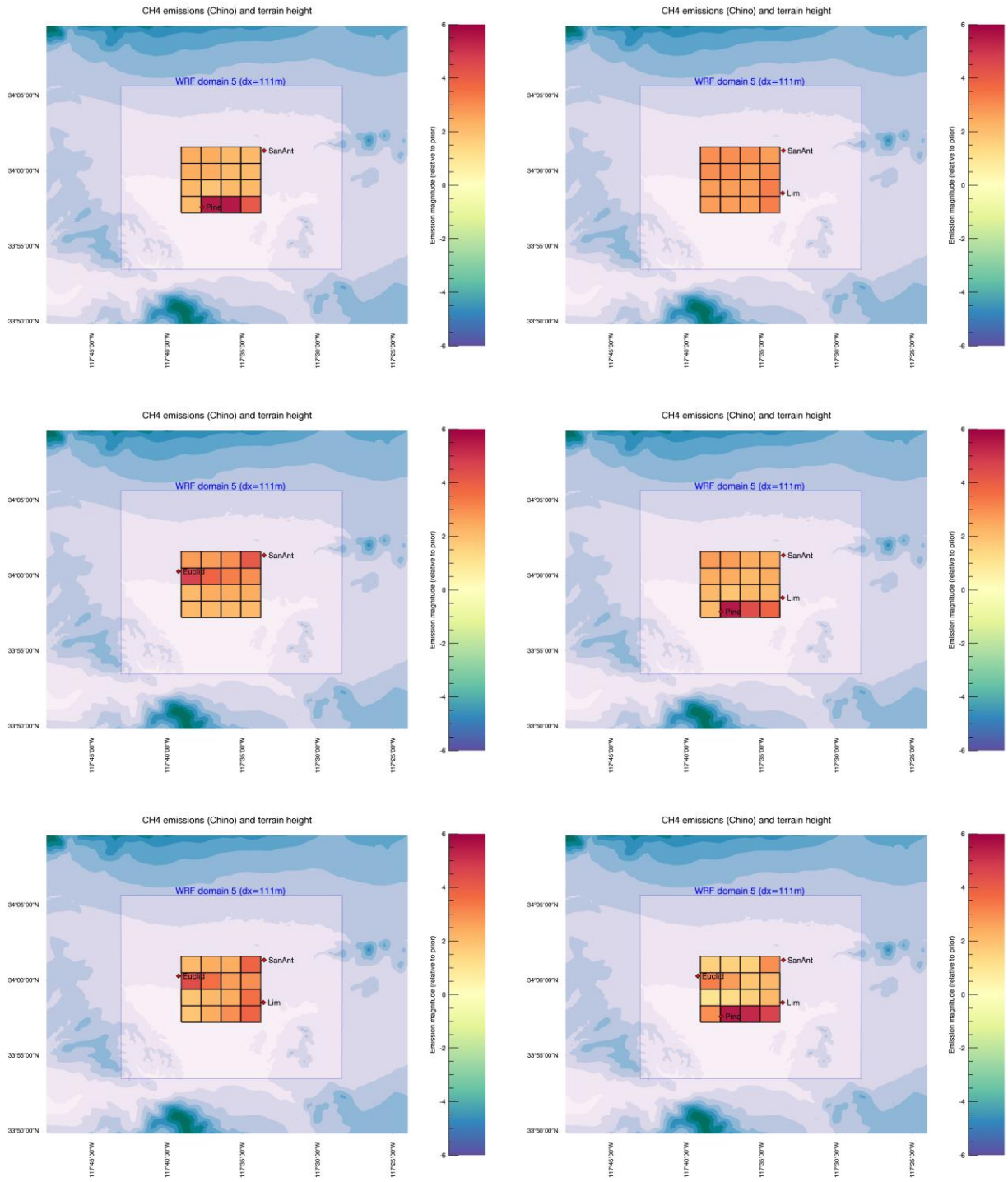
S3: computation of the residence times that airmasses cross over the dairies



To estimate CH_4 fluxes with FTS and wind measurements, the residence time of air over Chino has to be calculated (Equation 5). The distance that airmasses cross over the dairies (m) is first calculated for each one-minute FTS measurement at each site (Harvard2, Caltech, Harvard1, and LANL in blue, black, green, and squares in the figure above), each day of measurement (January 15th, 16th, and 24th), and each wind direction (θ). In this case of the scheme above, where 180°<θ<270°, $m_A = \frac{b}{cos(\theta)}$, $m_B = 0$, $m_C = \frac{b2}{cos(\theta)}$, and $m_D = 0$. The residence time t_A, t_B, t_C, and t_D is then calculated by dividing the distances by the one-minute wind speed measurements (average over the two local airports).



S4: Maps of CH_4 emissions (scaling factor relative to prior emissions) for the 15th January using different combinations of EM27/SUN locations and numbers. The two major sources located in the eastern and southwestern parts of the feedlots are detected with three EM27/SUN instruments, independent of their locations. The atmospheric inversion cases using only two instruments (upper row and left panel of the second row) identify only one source over the domain.



S5: Same as S4 for the 16th of January 2015. On the 16th of January, a strong southwestern source co-located with the manure lagoon was captured with a second large emitter in the northeastern corner. These two sources require that data from three instruments are assimilated in the inversion, similar to the previous day