## Supplement information:

## 2 S1: Coupling between mesoscale and LES modes

3 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, 4 5 there should be no inconsistency with using these fields to initialize all model grids. As the day 6 progresses and the PBL is heated from below, either random or topographically generated 7 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 8 9 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 10 11 PBL turbulence develops in the flow as it enters the domain. The LES domain is made large 12 enough such that realistic turbulent eddies have a chance to develop in the inflow before they 13 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 14 towards the South. However, hills nearly surround the valley along the southern perimeter. 15 16 Meanwhile, the foothills of the San Gabriel Mountains begin just off the 111-m domain boundary 17 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. 18



S2: Comparison of the observed CH<sub>4</sub> column enhancements (blue diamond), the Simulated
Annealing accepted solutions (in grey), and the optimal solution from the Bayesian inversion (red
line) in ppm.





To estimate CH<sub>4</sub> fluxes with FTS and wind measurements, the residence time of air over Chino 26 has to be calculated (Equation 5). The distance that airmasses cross over the dairies (m) is first 27 28 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 29  $15^{th}$ ,  $16^{th}$ , and  $24^{th}$ ), and each wind direction ( $\theta$ ). In this case of the scheme above, where 30 180°< $\theta$ <270°,  $m_A = \frac{b}{\cos(\theta)}$ ,  $m_B = 0$ ,  $m_C = \frac{b2}{\cos(\theta)}$ , and  $m_D = 0$ . The residence time t<sub>A</sub>, t<sub>B</sub>, t<sub>C</sub>, and 31 t<sub>D</sub> is then calculated by dividing the distances by the one-minute wind speed measurements 32 (average over the two local airports). 33



S4: Maps of CH<sub>4</sub> emissions (scaling factor relative to prior emissions) for the 15<sup>th</sup> January using different combinations of EM27/SUN locations and numbers. The two major sources located in the northeastern 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 16<sup>th</sup> of January 2015. On the 16th of January, a strong southeastern source was
captured with a second large emitter in the northeastern corner. These two sources require that data

43 from three instruments are assimilated in the inversion, similar to the previous day