



1           **LA Megacity: a High-Resolution Land-Atmosphere**  
2           **Modelling System for Urban CO<sub>2</sub> Emissions**

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26



## 1 **Abstract**

2 Megacities are major sources of anthropogenic fossil fuel CO<sub>2</sub> emissions. The spatial  
3 extents of these large urban systems cover areas of 10,000 km<sup>2</sup> or more with complex  
4 topography and changing landscapes. We present a high-resolution land-atmosphere  
5 modelling system for urban CO<sub>2</sub> emissions over the Los Angeles (LA) megacity area.  
6 The Weather Research and Forecasting (WRF)-Chem model was coupled to a very high-  
7 resolution FFCO<sub>2</sub> emission product, Hestia-LA, to simulate atmospheric CO<sub>2</sub>  
8 concentrations across the LA megacity at spatial resolutions as fine as ~1 km. We  
9 evaluated multiple WRF configurations, selecting one that minimized errors in wind  
10 speed, wind direction, and boundary layer height as validated by its performance against  
11 meteorological data collected during the CalNex-LA campaign (May-June 2010). Our  
12 results show no significant difference between moderate- (4-km) and high- (1.3-km)  
13 resolution simulations when evaluated against surface meteorological data, but the high-  
14 resolution configurations better resolved PBL heights and vertical gradients in the  
15 horizontal mean winds. We coupled our WRF configuration with the Vulcan 2.2 (10 km  
16 resolution) and Hestia-LA (1.3-km resolution) fossil fuel CO<sub>2</sub> emission products to  
17 evaluate the impact of the spatial resolution of the CO<sub>2</sub> emission products and the  
18 meteorological transport model on the representation of spatiotemporal variability in  
19 simulated atmospheric CO<sub>2</sub> concentrations. We find that high spatial resolution in the  
20 fossil fuel CO<sub>2</sub> emissions is more important than in the atmospheric model to capture CO<sub>2</sub>  
21 concentration variability across the LA megacity. Finally, we present a novel approach  
22 that employs simultaneous correlations of the simulated atmospheric CO<sub>2</sub> fields to  
23 qualitatively evaluate greenhouse gas measurements over the LA megacity. Spatial  
24 correlations in the atmospheric CO<sub>2</sub> fields reflect the coverage of individual measurement  
25 sites when a statistically significant number of sites observe emissions from a specific  
26 source or location. We conclude that elevated atmospheric CO<sub>2</sub> concentrations over the  
27 LA megacity are composed of multiple fine-scale plumes rather than a single  
28 homogenous urban dome. Furthermore, we conclude that FFCO<sub>2</sub> emissions monitoring in  
29 the LA megacity requires FFCO<sub>2</sub> emissions modelling with ~1 km resolution since  
30 coarser resolution emissions modelling tends to overestimate the observational  
31 constraints on the emissions estimates.



## 1 1 Introduction

2 Carbon dioxide (CO<sub>2</sub>) is a major anthropogenic contributor to climate change. It has  
3 increased from its preindustrial (1750) level of 278 ± 2 ppm (Etheridge et al., 1996) to  
4 over 400 ppm in recent years, as reported by the National Oceanic and Atmospheric  
5 Administration (NOAA) and Scripps Institution of Oceanography [<http://co2now.org/>].  
6 Clear evidence has shown that the continued increase of the atmospheric CO<sub>2</sub>  
7 concentration is dominated by global fossil fuel consumption during the same period  
8 (IPCC, 2013) and land use change (Houghton, 1999).

9 Urban areas are significant sources of fossil fuel CO<sub>2</sub> (FFCO<sub>2</sub>), representing more than  
10 50% of the world's population and more than 70% of FFCO<sub>2</sub> (UN, 2006). In particular,  
11 megacities (cities with urban populations greater than 10 million people) are major  
12 sources of anthropogenic emissions, with the world's 35 megacities emitting more than  
13 20% of the global anthropogenic FFCO<sub>2</sub>, even though they only represent about 3% of  
14 the Earth's land surface (IPCC, 2013). The proportion of emissions from megacities  
15 increases monotonically with the world population and urbanization (UN, 2006, 2010).  
16 Developed and developing megacities around the world are working together to pursue  
17 strategies to limit CO<sub>2</sub> and other greenhouse gas (GHG) emissions (C40, 2012).

18 Carbon fluxes can be estimated using “bottom-up” and “top-down” methods. Typically,  
19 FFCO<sub>2</sub> emissions are determined using “bottom-up” methods, by which fossil fuel usage  
20 from each source sector is convolved with the estimated carbon content of each fuel type  
21 to obtain FFCO<sub>2</sub> emission estimates. Space-time resolved FFCO<sub>2</sub> data sets using “bottom-  
22 up” methods clearly reveal the fingerprint of human activity with the most intense  
23 emissions being clustered around urban centres and associated power plants (e.g., Gurney  
24 et al., 2009; Gurney et al., 2012). At the global and annual scale, FFCO<sub>2</sub> emission  
25 estimates remain uncertain at ±5%, varying widely by country and reporting method (Le  
26 Quéré et al., 2014). At the urban scale, the uncertainties of FFCO<sub>2</sub> emission estimates are  
27 often 50-200 % (Turnbull et al., 2011; Asefi-Najafabady et al., 2014). “Top-down”  
28 methods could potentially estimate biases in bottom-up emissions, and could also detect  
29 trends that cities can use for decision-making, due to changing economic activity or  
30 implementation of new emission regulations.



1 “Top-down” methods involve atmospheric measurements and usually include an  
2 atmospheric inversion of CO<sub>2</sub> concentrations, using atmospheric transport models to  
3 estimate carbon fluxes (i.e., posterior fluxes) by adjusting the fluxes (i.e., prior fluxes) to  
4 be consistent with observed CO<sub>2</sub> concentrations (e.g., Lauvaux et al., 2012; Lauvaux et  
5 al., 2015; Tarantola, 2005; Enting et al., 1994; Gurney et al., 2002; Baker et al., 2006;  
6 Law et al., 2003). In general, a prior flux is required for estimating the fluxes using an  
7 atmospheric inversion. The uncertainties in “top-down” methods therefore can be  
8 attributed to errors in the observations (e.g., Tarantola, 2005), emission aggregation  
9 errors from the prior fluxes (e.g., Gurney et al., 2012; Engelen et al., 2002), and physical  
10 representation errors in the atmospheric transport model (e.g., Díaz Isaac et al., 2014;  
11 Gerbig et al., 2008; Kretschmer et al., 2012; Lauvaux et al., 2009; Sarrat et al., 2007).  
12 Previous studies showed that regional high-resolution models can capture the measured  
13 CO<sub>2</sub> signal much better than the global models with lower resolution and simulate the  
14 diurnal variability of the atmospheric CO<sub>2</sub> field caused by recirculation of nighttime  
15 respired CO<sub>2</sub> well (Ahmadov et al., 2009). Pillai et al. (2011 and 2012) and Rödenbeck et  
16 al. (2009) have discussed about the advantages of high resolution CO<sub>2</sub> modelling on  
17 different domains and applications. Recent efforts to study FFCO<sub>2</sub> emissions on urban  
18 scales have benefited from strategies that apply in-situ observations concentrated within  
19 cities and mesoscale transport models (e.g., Wu et al., 2011; Lauvaux et al., 2015; Strong  
20 et al., 2011; Lac et al., 2013; Bréon et al., 2015).

21 The Los Angeles (LA) megacity is one of the top three FFCO<sub>2</sub> emitters in the U.S. The  
22 atmospheric CO<sub>2</sub> concentrations show complex spatial and temporal variability resulting  
23 from a combination of large FFCO<sub>2</sub> emissions, complex topography, and challenging  
24 meteorological variability (e.g., Brioude et al., 2013; Wong et al., 2015; Angevine et al.,  
25 2012; Conil and Hall, 2006; Ulrickson and Mass, 1990; Lu and Turco, 1995; Baker et al.,  
26 2013; Chen et al., 2013; Newman et al., 2013). Past studies of exploring CO<sub>2</sub>  
27 concentrations over the LA megacity used measurement methods ranging from ground-  
28 based to airborne, from in-situ to column. Those studies consistently reported robust  
29 enhancements (e.g., 30-100 ppm in-situ and 2-8 ppm column) and significant variability  
30 of the CO<sub>2</sub> concentrations for the LA megacity (Newman et al., 2013; Wunch et al., 2009;  
31 Wong et al., 2015; Kort et al., 2012; Wennberg et al., 2012; Newman et al., 2015). There



1 have been limited radiocarbon ( $^{14}\text{C}$ ) isotopic tracer studies (Newman et al., 2013;  
2 Newman et al., 2008; Djuricin et al., 2010; Riley et al., 2008; Newman et al., 2015).  
3 Newman et al. (2013) showed that FFCO<sub>2</sub> constituted 10 - 25 ppm of the CO<sub>2</sub> excess  
4 observed in the LA basin by averaging the flask samples at 1400 PST during 15 May –  
5 15 June, 2010. Djuricin et al. (2010) demonstrated that fossil fuel combustion contributed  
6 approximately 50~70 % of CO<sub>2</sub> sources in LA. Recently, using CO<sub>2</sub> mole fractions and  
7  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  values of CO<sub>2</sub> in the LA megacity observed in inland Pasadena (2006–  
8 2013) and coastal Palos Verdes peninsula (autumn 2009–2013), Newman et al. (2015)  
9 demonstrated that fossil fuel combustion is the dominant source of CO<sub>2</sub> for inland  
10 Pasadena. Airborne campaigns over LA (typically days to weeks in duration) included  
11 ARCTAS-CA (Jacob et al., 2010) and CalNex-LA (Brioude et al., 2013). All of these  
12 earlier studies were limited in their ability to investigate the spatial and temporal  
13 characteristics of LA carbon fluxes given relatively sparse observations. To better  
14 understand and quantify the total emissions, trends, and the detailed spatial, temporal, and  
15 source sector patterns of emissions over the LA megacity requires both a denser  
16 measurement network and a land-atmosphere modelling system appropriate for such a  
17 complex urban environment. In this paper, we couple the Weather Research and  
18 Forecasting (WRF) – Chem model to a high-resolution FFCO<sub>2</sub> emission product, Hestia-  
19 LA, to study the spatiotemporal variability of urban CO<sub>2</sub> concentrations over the LA  
20 megacity.

21 The mesoscale circulation over the LA megacity is challenging for atmospheric transport  
22 models due to a variety of phenomena, such as “Catalina” eddies off the coast of southern  
23 California and the coupling between the land-sea breeze and winds induced by the  
24 topography (Angevine et al., 2012; Conil and Hall, 2006; Ulrickson and Mass, 1990;  
25 Kusaka and Kimura, 2004b; Kusaka et al., 2001). In this paper we present a set of  
26 simulations exploring WRF model physics configurations for the LA megacity,  
27 evaluating the model performance against meteorological data from the CalNex-LA  
28 campaign period, 15 May – 15 June 2010. Angevine et al. (2012) also investigated how  
29 WRF model performance varied with spatial resolutions and PBL scheme, etc for the  
30 CalNex-LA campaign period; however, Angevine et al. focused solely on model  
31 meteorological evaluation with spatial resolutions of 12- and 4-km. In the present study



1 we focus on three critical aspects of the WRF model configuration – the planetary  
2 boundary layer (PBL) scheme, the urban surface scheme, and the model spatial resolution  
3 – as well as the effects of the FFCO<sub>2</sub> emissions product spatial resolution. Through these  
4 four aspects, the impacts of physical representation errors and emission aggregation  
5 errors on the modelled CO<sub>2</sub> concentrations across the LA megacity are investigated.

6 Moreover, a novel approach is proposed to evaluate the design of the greenhouse gas  
7 (GHG) measurement network for the LA megacity. The LA measurement network  
8 consists of 15 observation sites designed to provide continuous atmospheric CO<sub>2</sub>  
9 concentrations to assess the anthropogenic carbon emissions distribution and trends. The  
10 goal of the network design exploration is to optimize the atmospheric observational  
11 constraints on the surface fluxes. Kort et al. (2013) found that a minimum of eight  
12 optimally located in-city surface CO<sub>2</sub> observation sites were required for accurate  
13 assessment of CO<sub>2</sub> emissions in LA using the “footprint” method (backward mode) and  
14 based on a national FFCO<sub>2</sub> emission product Vulcan. Here we assess the influence of  
15 each observation site using spatial correlations in terms of the simulated CO<sub>2</sub> (forward  
16 mode) at high-resolution.

17 The remainder of the paper is organized as follows. Section 2 describes the modelling  
18 framework, including initial conditions and boundary conditions for WRF-Chem. In  
19 section 3, we assess the quality of the model results, focusing on accurate representation  
20 of the PBL height, wind speed and wind direction. Section 4 presents the spatial and  
21 temporal patterns of simulated CO<sub>2</sub> concentration fields over the LA megacity using  
22 various FFCO<sub>2</sub> emissions products. Section 5 describes the forward mode approach for  
23 evaluating the spatial sensitivity of the 2015-era surface GHG measurement sites within  
24 the LA megacity. Discussion of model errors, model sampling strategy, and the density of  
25 the LA GHG measurement network from the forward model perspective is given in  
26 section 6. A summary is given in section 7. Section 8 lists the author contributions.

27

## 28 **2 Modelling Framework**

29 Sensitivity experiments were conducted using WRF-Chem version 3.6.1 with various  
30 PBL schemes, urban surface schemes, and model resolutions to define an optimized



1 configuration for simulating atmospheric CO<sub>2</sub> concentration fields over the LA megacity.  
2 The impact of the resolution of FFCO<sub>2</sub> emission products is investigated as well.

### 3 **2.1 WRF model setup**

4 All of the model runs used one-way triple-nested domains with resolutions of 12-, 4-, and  
5 1.3-km. The coarse domain (d01) covers most of the western US; the intermediate  
6 domain (d02) covers California and part of Mexico (Figure 1a); the innermost domain  
7 (d03) covers the majority of the South Coast Air Basin, a portion of the southern San  
8 Joaquin Valley and extends into the Pacific Ocean to include Santa Catalina and San  
9 Clemente Islands (Figure 1b). The Los Angeles basin is surrounded to the north and east  
10 by mountain ranges with summits of 2-3 km, with the ocean to the west and the desert to  
11 the north. The basin consists of the West Coast Basin, Central Basin, and Orange County  
12 Coastal Plain. The boundaries of these three regions are Newport Inglewood Fault and  
13 the boundary between Los Angeles County and Orange County. In this study, our  
14 analysis is limited to the innermost domain (d03), referred to hereafter as the LA  
15 megacity. All three of the model domains use 51 terrain following vertical levels from  
16 surface to 100 hPa, of which 29 layers are below 2 km above ground level (AGL).

17 The meteorological fields and surface parameters, such as soil moisture, were initialized  
18 by the three-hourly North American Regional Reanalysis (NARR) data set with a  
19 horizontal resolution of 32 km (Mesinger et al., 2006) and the six-hourly NCEP sea  
20 surface temperature data set with a horizontal resolution of 12 km  
21 (<ftp://polar.ncep.noaa.gov/pub/history/sst/ophi>). A summary of WRF configurations  
22 common to all sensitivity runs is shown in Table 1. The impact of varying the PBL  
23 parameterization, urban surface, and model resolution was investigated by conducting  
24 sensitivity runs summarized in Table 2.

25 PBL schemes are used to parameterize the unresolved turbulent vertical fluxes of heat,  
26 momentum, and constituents within the planetary boundary layer. There are tens of  
27 mesoscale PBL schemes available in the WRF package. We selected the three most  
28 commonly used turbulent kinetic energy (TKE)-driven PBL schemes for the sensitivity  
29 runs: the Mellor-Yamada-Janjic technique (MYJ, Janjić, 1994), Mellor-Yamada



1 Nakanishi and Niino Level 2.5 (MYNN, Nakanishi and Niino, 2006), and Bougeault-  
2 Lacarrère (BouLac, Bougeault and Lacarrere, 1989). The TKE-driven PBL schemes  
3 explicitly estimate the turbulent fluxes from mean atmospheric states and/or their  
4 gradients and can be used to drive a Lagrangian particle dispersion models in subsequent  
5 atmospheric inversions (e.g., Lauvaux et al., 2008).

6 For an accurate representation of the LA CO<sub>2</sub> simulation, the necessity of incorporating  
7 the urban surface scheme was tested by alternatively including an urban canopy model  
8 (UCM, Kusaka and Kimura, 2004a), a building environment parameterization (BEP,  
9 Martilli et al., 2009), and no urban surface scheme.

10 We chose to test and evaluate our WRF-Chem configuration during the May-June 2010  
11 time period of the CalNex-LA campaign (Ryerson et al., 2013) to take advantage of the  
12 extra meteorological measurements recorded during the campaign. Hourly simulations  
13 were conducted for 36-h periods starting with a 12-h meteorological spin-up at 12:00  
14 UTC of the previous day. Hence, when concatenating the model output, each new run is  
15 introduced at 0000 UTC. All of the analyses in the following sections are limited to the  
16 region of the LA megacity.

## 17 **2.2 Configuration for the CO<sub>2</sub> simulation**

18 WRF-Chem version 3.6.1 was modified to allow for online CO<sub>2</sub> tracer transport coupled  
19 with the Vegetation Photosynthesis and Respiration Model (VPRM) (Ahmadov et al.,  
20 2007; Xiao et al., 2004). VPRM calculates hourly net ecosystem exchange based on  
21 MOIDS satellite estimates of the land surface water index and enhance vegetation index,  
22 short wave radiance and surface temperature. A detailed description of VPRM can be  
23 found in Mahadevan et al. (2008).

24 Anthropogenic FFCO<sub>2</sub> fluxes were alternatively prescribed from the Vulcan 2.2 and  
25 Hestia-LA 1.0 FFCO<sub>2</sub> emission products developed at Arizona State University (Gurney  
26 et al., 2009; Gurney et al., 2012; Gurney et al., 2015; Rao et al., 2015). Both emission  
27 products were developed using “bottom-up” methods. Vulcan quantifies FFCO<sub>2</sub>  
28 emissions for the entire contiguous United States (CONUS) hourly at approximately 10  
29 km spatial resolution for the year of 2002, combining data sources such as local pollution





1 reporting, traffic data, and point source monitoring (Gurney et al., 2009). Hestia-LA, by  
2 contrast, is a fossil fuel CO<sub>2</sub> emissions data product specific in space and time to the  
3 individual building, road segments, and point sources of the Los Angeles megacity (Rao  
4 et al., 2015; Gurney et al., 2015; Gurney et al., 2012; Zhou and Gurney, 2010).  
5 Leveraging from the Vulcan constraint at the county level, Hestia-LA quantifies FFCO<sub>2</sub>  
6 emissions for Los Angeles County, Orange County, San Bernardino County, Ventura  
7 County, and Riverside County, at approximately 1.3 km x 1.3 km every hour of the years  
8 of 2011 and 2012. More details about Hestia-LA see Rao et al. (2015).

9 Atmospheric CO<sub>2</sub> concentrations in WRF-Chem were alternatively driven by the Vulcan  
10 and Hestia-LA emissions at the resolutions of 4 km and 1.3 km. Hence, four different  
11 emission datasets were generated – Vulcan 10 km emissions transported at 4-km or 1.3-  
12 km resolution, and Hestia-LA 1.3 km emissions transported at 4-km or 1.3-km resolution.  
13 The Hestia-LA emissions were aggregated from the native building-level resolution to  
14 the 1.3 and 4 km resolutions via direct summation in the specified model grids. Hestia-  
15 LA 2011 is temporally shifted for creating the weekday-weekend cycle for the year of  
16 2010. The Vulcan FFCO<sub>2</sub> emissions were interpolated by using a bilinear operator and by  
17 preserving the value of the integral of data between the source (10-km) and destination  
18 (4- and 1.3-km) grid. Also, the ratio of the total carbon emissions over the state between  
19 the years of 2002 and 2015 from California Air Resource Board (<http://www.arb.ca.gov/>)  
20 was uniformly applied to the Vulcan emissions to temporally scale Vulcan from the 2002  
21 base year to 2010. At regional scales, anthropogenic and biogenic fluxes are much larger  
22 than ocean fluxes. Hence, no CO<sub>2</sub> ocean fluxes were prescribed. This paper analyses the  
23 impact of both physical representation errors and emission aggregation errors on the  
24 modelled CO<sub>2</sub> concentrations across the LA megacity.

25 Lateral boundary conditions and initial conditions for CO<sub>2</sub> concentration fields were  
26 taken from the three-dimensional CO<sub>2</sub> background (often called “NOAA curtain” for  
27 background) estimated from measurements in the Pacific (Jeong et al., 2013).

28



### 1 **3 Model – data comparison**

2 Meteorological observations obtained during the CalNex-LA campaign  
3 (<http://www.esrl.noaa.gov/csd/projects/calnex/>) include PBL height sampled by NOAA  
4 P-3 flights and aerosol backscatter ceilometer (Haman et al., 2012; Scarino et al., 2013), a  
5 radar wind profiler operated by the South Coast Air Quality Management District near  
6 Los Angeles International Airport (LAX), and CO<sub>2</sub> in situ measurements (Newman et al.,  
7 2013). Additionally, the NWS (National Weather Service, [www.weather.gov](http://www.weather.gov)) surface  
8 observations are used.

#### 9 **3.1 Comparison to aircraft PBL height**

10 During CalNex-LA, 17 P-3 research flights sampled the daytime and nighttime PBL,  
11 marine surface layer, and the overlying free troposphere throughout California (Ryerson  
12 et al., 2013). We imposed five criteria for selecting aircraft profiles of potential  
13 temperature for PBL height comparisons:

- 14 1) Aircraft profiles sample within the innermost model domain (d03, Figure 1b);
- 15 2) Profiles sample during daytime (1100 PST – 1700 PST) when the CO<sub>2</sub> concentrations  
16 in PBL is well mixed;
- 17 3) Profiles acquired within ±30 min of the model output;
- 18 4) Profiles with valid sampling below and above 1 km AGL to assure the chance to  
19 determine the PBL height from the potential temperature vertical gradient;
- 20 5) Ability to determine the PBL height from the vertical gradient of potential  
21 temperature.

22 Based on these five criteria, we selected seven aircraft profiles collected between 16 May  
23 and 19 May 2010. Figure 2 shows a profile acquired on 19 May 2010 when the aircraft  
24 was sampling over Pasadena.

25 The model diagnostic PBL height calculated by each PBL scheme can differ from the  
26 others due to the Richardson bulk number ( $R_i$ ) used (e.g., Kretschmer et al., 2014; Hong  
27 et al., 2006; Yver et al., 2013). To avoid this difference, we determined modelled PBL  
28 height based on the vertical virtual potential temperature gradient. For the case (Figure



1 2), the modelled PBL height agrees within 50 meters of the aircraft-determined and  
2 ceilometer-measured PBL height

3 Figure 3 shows the absolute difference between the modelled and aircraft-determined  
4 PBL height for each selected aircraft profile. The differences between the modelled and  
5 aircraft-determined PBL height differ case by case. None of the model physics is  
6 systematically better than others. However, BouLac\_BEP and MYNN have larger biases  
7 than others. The averaged bias of BouLac\_BEP is 289 m for d02, 295 m for d03; MYNN  
8 is 179 m for d02 and 216 m for d03. For other configurations, the averaged biases are  
9 smaller than 160 m. The modelled PBL bias appears somewhat smaller in the 4-km runs  
10 than the 1.3-km runs. This, however, is obtained based on seven selected aircraft profiles  
11 only. To further define the optimal physics for the PBL height simulation, we will present  
12 the all-hours statistics with the ceilometer data in section 3.2.

### 13 **3.2 Comparison to ceilometer PBL height**

14 Accurate simulation of the time evolution of the PBL height is crucial to properly  
15 simulate the vertical mixing and ventilation of CO<sub>2</sub> emitted at the surface. The ceilometer  
16 measurements during CalNex-LA (Haman et al., 2012) allow us to evaluate the time  
17 evolution of the modelled PBL height. Compared with the ceilometer-measured PBL  
18 height, the maximum discrepancies between model and observations occur from around  
19 1100 PST – 1200 PST when the nocturnal PBL is fully collapsed and 1700 PST when it  
20 starts to form again (Figure 4). Among all of the model physics, MYNN\_UCM shows the  
21 best agreement with the observations, while BouLac\_BEP differs from ceilometer the  
22 most. The absolute bias of the MYNN\_UCM modelled PBL height ranges from 5 to 198  
23 m and 0 to 184 m with mean bias of  $-15.3 \pm 66.1$  m and  $-6.9 \pm 82.7$  m for d02 and d03,  
24 respectively, suggesting the 1.3-km model resolution statistically improves the model  
25 performance in the PBL simulation as compared with the ceilometer. The improvement  
26 in the high-resolution model runs can be seen in other configurations as well. However,  
27 the ceilometer measurements were all at Caltech and thus reflect interior conditions.  
28 These are expected to be very different from coastal conditions in terms of the temporal  
29 evolution and eventual height of the mid-day PBL as well as the timing of the nocturnal



1 PBL collapse, etc. The domain is much larger and more varied than captured by a single  
2 location.

3 We also notice that using UCM-coupled simulations agree with the ceilometer better than  
4 other combinations (MYNN\_UCM vs. MYNN, MYJ\_UCM vs. MYJ, BouLac\_UCM vs.  
5 BouLac\_BEP). Using UCM can largely reduce the difference across the model runs and  
6 discrepancy from the observations.

### 7 **3.3 Comparison to radar wind profiler**

8 Atmospheric dynamics has a direct influence on the CO<sub>2</sub> transport. Realistically  
9 reproducing the vertical gradient of wind fields is crucial. In Figure 5, we show the  
10 average difference in the wind profiles between the models and the radar wind profiler at  
11 LAX (Angevine et al., 2012). Most of the simulations show relatively larger wind speed  
12 bias near the surface: BouLac\_BEP, MYJ, and MYNN with bias of  $2.4 \pm 2.2$  m/s,  
13 BouLac\_UCM and MYJ\_UCM with bias of  $2.0 \pm 2.3$  m/s. In contrast, it is encouraging  
14 to see that MYNN\_UCM agrees with the radar measurement best with mean bias of  $1.4 \pm$   
15  $2.0$  m/s, a lower mean bias than for the other configurations. Additionally, UCM-coupled  
16 simulations tend to reduce the wind speed bias at this location.

17 For wind direction, likewise, MYNN\_UCM agrees with the observations slightly better  
18 below 800 m (About 1.1 m/s for the averaged error), although the model bias is much less  
19 pronounced across the configurations. However, we notice that MYNN\_UCM shows  
20 larger wind direction bias between 800 – 1400 m than others due to relatively lower PBL  
21 height simulated (not shown).

22 Improvement provided by the 1.3-km model resolution is visible near the PBL height  
23 (800 – 1400 m). A finer model resolution tends to resolve the vertical gradients of the  
24 atmospheric states better. This also can be demonstrated by the PBL comparisons with  
25 ceilometer (Figure 4).

26 Angevine et al. (2012) evaluated a set of model configurations with the highest model  
27 resolution at 4 km for CalNex-LA using the same radar wind profiler data. The optimal  
28 configuration (the total energy–mass flux boundary layer scheme and ECMWF  
29 reanalysis) they found showed  $1.1 \pm 2.7$  m/s bias in wind speed and  $-2.6 \pm 67^\circ$  in wind



1 direction near the surface. Here MYNN\_UCM displays similar performance to the  
2 optimal configuration they concluded. At the 4-km model resolution, the biases of  
3 MYNN\_UCM are  $1.4 \pm 2.0$  m/s in wind speed and  $-1.3 \pm 20.0^\circ$  in wind direction. In  
4 section 3.4, we will examine the performance of MYNN\_UCM across the LA megacity.

### 5 **3.4 Comparison to NWS surface stations**

6 Due to the limited number of observation sites available at this time in this region, the  
7 analysis above can only be done at specific locations. We therefore introduce the NWS  
8 surface network to demonstrate the model performance across the LA megacity. The  
9 objective analysis program OBSGRID is used to remove erroneous data and observations  
10 that are not useful (Deng et al., 2009; Rogers et al., 2013).

11 Figure 6 shows the model bias compared to the NWS surface data across the LA  
12 megacity. The locations of the GHG measurement sites are marked (see details in Table 3  
13 and Figure S1). Overall, there is little difference in the simulated surface atmospheric  
14 state variables between the 4-km and 1.3-km runs; i.e., the 1.3-km run does not show any  
15 significant improvement compared to the 4-km run at the surface (even though it resolves  
16 the vertical gradient of atmospheric states and PBL better, Figure 4 and 5).

17 For temperature (Figure 6a1 and 6b1), the model is colder than the observations by 0.5 -  
18 1.0 K. Larger temperature biases occur in the desert. For relative humidity (Figure 6a2  
19 and 6b2), the model is dry compared to the observations, which is consistent with the  
20 findings of Nehr Korn et al. (2012). The model is 5% dryer over the basin with a  
21 somewhat larger bias of 5% - 10% near Granada Hills and Ontario that have the highest  
22 temperature in the summer – typically 20 °F or more warmer than downtown LA in May-  
23 June. The dryness in the model tends to cause lower PBL heights, which can be seen in  
24 the comparison to the ceilometer-determined PBL height at Caltech in Pasadena,  
25 California (Figure 4): MYNN\_UCM shows a shallower PBL in comparison to the  
26 ceilometer during the 1400 PST – 1800 PST time period.

27 The model overestimates wind speed by  $\sim 1.0$  m/s (Figure 6a3 and 6b3). The tendency of  
28 the model to overestimate wind speed is fully documented in previous studies (e.g.,  
29 Angevine et al., 2012; Brioude et al., 2013; Nehr Korn et al., 2012; Yver et al., 2013). For



1 surface wind direction, model bias is within  $\pm 10^\circ$  for most of the LA megacity. The  
2 larger biases appear near the foothills of Santa Monica Mountains, San Gabriel  
3 Mountains, and University of Southern California (USC) due to the challenging land  
4 surface and terrain.

5 Compared with other model physics (not shown), we notice that USC located in the  
6 downtown LA is a challenging location for mesoscale modelling, in particular for wind  
7 simulations. All of the model physics consistently show a relatively large wind bias at  
8 USC except BouLac\_BEP that fails in the remainder of the domain. We also noticed that  
9 adding UCM to MYNN decreases the modelled temperature, while all of other models'  
10 physics have a warm bias compared to observations.

11 All of the analyses above focused on the meteorology over the LA megacity. The results  
12 indicate little difference horizontally between 4- and 1.3-km runs across the basin, which  
13 is consistent with the Angevine et al. (2012) assumption that a finer grid may not give  
14 better results. However, the 1.3-km run tends to resolve the vertical gradients of  
15 atmospheric state variables and PBL better, which can improve the vertical mixing and  
16 ventilation of modelled atmospheric CO<sub>2</sub> concentrations.

17 Overall, the MYNN\_UCM configuration showed the best agreement with meteorological  
18 observations of all the configurations we evaluated. Therefore, we will use the MYNN-  
19 UCM configuration in our simulations of atmospheric CO<sub>2</sub> concentration fields over the  
20 LA megacity.

### 21 **3.5 Comparisons to in-situ CO<sub>2</sub>**

22 We coupled Hestia and Vulcan FFCO<sub>2</sub> emission products individually with the  
23 MYNN\_UCM WRF configuration to generate four sets of simulated CO<sub>2</sub> concentrations:  
24 WRF-Hestia 1.3-km, WRF-Hestia 4-km, WRF-Vulcan 1.3-km, and WRF-Vulcan 4-km.  
25 The runs with the same model resolution have the same meteorology but differ in  
26 emissions, and vice versa.

27 During CalNex-LA, in-situ observation sites at Pasadena and Palos Verdes continuously  
28 measured surface CO<sub>2</sub> concentrations. Measurements were recorded using a Picarro  
29 (Santa Clara, CA) Isotopic CO<sub>2</sub> Analyser (cavity ring-down spectrometer), model G1101-



1 i, for Pasadena and an infrared gas analyser from PP Systems (Haverford, MA), model  
2 CIRAS-SC for Palos Verdes. In addition, periodic flask samples were collected for  
3 analysis of  $^{14}\text{CO}_2$  for extracting fossil fuel and biogenic signals. See Newman et al.  
4 (2015) for details about the sites and sampling information. Figure 7 shows the  
5 comparison of the time series of hourly (Figure 7a,b) and daily afternoon (Figure 7c,d)  
6 averaged  $\text{CO}_2$  concentrations (1300 PST – 1700 PST) between model and observation.  
7 Overall, the model captures the temporal variability of  $\text{CO}_2$  but overestimates  $\text{CO}_2$  during  
8 nighttime. During afternoons, the model agrees with the observations fairly well (Figure  
9 7c and 7d) except for a few events: all simulations underestimate  $\text{CO}_2$  concentrations by  
10 about 10 ppm around May 28 and June 4-6 for Pasadena and May 21 for Palos Verdes.  
11 These events lasting two – three days are likely related to synoptic scale processes. Using  
12 the averaged Pacific Ocean  $\text{CO}_2$  signal as background may explain the failure to capture  
13 these events. Further investigation of the background air would provide insights related to  
14 synoptic variability but is beyond the scope of this work. We focus here on the diurnal  
15 variability.

16 Clear diurnal variations of the surface  $\text{CO}_2$  concentrations were observed for both sites  
17 (Figure 8). The observed  $\text{CO}_2$  concentrations increase at night and remains high until  
18 sunrise, and quickly drop as the boundary layer grows after sunrise (Figure 8a and 8b).

19 For the Pasadena site, during nighttime, when the PBL is shallow,  $\text{CO}_2$  is trapped locally:  
20 the more fossil fuel is emitted, the higher  $\text{CO}_2$  concentration is simulated. Consequently,  
21 the WRF-Vulcan runs show considerably lower  $\text{CO}_2$  concentration than the WRF-Hestia  
22 runs due to the lower emissions in Vulcan at the Pasadena site (Figure 8c). However,  
23 during daytime, with well-mixed conditions, the discrepancy between the WRF-Hestia  
24 and WRF-Vulcan runs becomes smaller. Among these runs, the 1.3-km WRF-Hestia run  
25 successfully captures the diurnal variation of the surface  $\text{CO}_2$  concentration, although a  
26 peak is not present in the observation around noon. By contrast, the 4-km WRF-Hestia  
27 run underestimates the  $\text{CO}_2$  concentration during 0200 – 0700 PST even though  
28 emissions were comparable between Hestia 4-km and Hestia 1.3-km (Figure 8c). The  
29 underestimation of the simulated  $\text{CO}_2$  concentration must mainly result from the  
30 representation errors in the atmospheric transport due to the coarser model resolution.



1 For Palos Verdes, however, none of the model results match the observations. All of the  
2 runs show a peak in the simulated CO<sub>2</sub> concentration around 0800 PST, which very likely  
3 corresponds to the eastward marine flow as a part of the Catalina eddy (e.g., Bosart,  
4 1983; Davis et al., 2000). This CO<sub>2</sub> concentration peak is incorrectly reproduced by the  
5 model advecting the FFCO<sub>2</sub> emitted from the strong point sources in Long Beach,  
6 California (Figure 1d) and in turn contaminating the air of Palos Verdes.

7

#### 8 **4 Spatial pattern of the surface CO<sub>2</sub>**

9 The spatial pattern of surface CO<sub>2</sub> concentration exhibits diurnal variability over the LA  
10 megacity due to the complexity of the topography and the variability of circulation  
11 patterns, PBL heights, and FFCO<sub>2</sub> emissions. Each plays an important role in sequence or  
12 at the same time. Here, we only focus on the pattern at 1400 PST when the atmospheric  
13 CO<sub>2</sub> concentration is well mixed in the PBL. At 1400 PST, there is a close relationship  
14 between CO<sub>2</sub> concentration and atmospheric transport; the error due to the PBL height  
15 determination is at a minimum. For the same reason, we show that FFCO<sub>2</sub> emissions do  
16 not play a dominant role around 1400 PST unless there are strong local signals from point  
17 sources, such as power plants, refineries, airports etc.

18 In this section, we define the 1.3-km WRF-Hestia run as the reference simulation. For  
19 simplicity, all of the relevant CO<sub>2</sub> spatial patterns we present are selected from the second  
20 model layer (about 24 m AGL). Figure 9a and 9b display the topography and the average  
21 CO<sub>2</sub> concentration at 1400 PST overlaid with the first empirical orthogonal function  
22 (EOF1) of the surface wind pattern, respectively. The locations of the 13 GHG  
23 measurement sites in the LA megacity domain are marked in the figures (see Table 3 and  
24 Figure S1 for details about the observation sites). Note that The 2015-era surface GHG  
25 measurement network includes 14 sites in total, while 13 sites are included in the  
26 innermost model domain. According to the geography mentioned in section 2.1, the  
27 Granada Hills (GH), Compton, USC, and sites are located in the West Coast Basin, the  
28 Pasadena and Mt. Wilson (MWO) sites are in the Central Basin, and California State  
29 University Fullerton (CSUF), Ontario, and San Bernardino (SB) sites are in the Orange  
30 County Coastal Plan. Additionally, the Dryden and Victorville (VV) sites are located in





1 deserts; the Palos Verdes (PV), University of California Irvine (UCI), and San Clemente  
2 Island (SCI) are on the coast. Although the Dryden site is actually a TCCON site, in the  
3 analysis, we assume it is a near-surface point measurement like other sites for simplicity.

4 Blocked by the mountains, the emitted CO<sub>2</sub> is trapped in the basin; the desert is as clean  
5 as the upwind ocean. Specifically, Dryden (not shown on the figure), VV, SCI (not  
6 shown on the figure), Palos Verdes and UCI are much cleaner than other sites (Figure  
7 9b). At 1400 PST, sea breeze prevails over the LA megacity. Affected by the geometry of  
8 Palos Verdes Peninsula, the sea breeze is divided into west and southwest onshore flows  
9 and then converge in the Central Basin. Strong CO<sub>2</sub> signals emitted from electricity  
10 production and industry (with annual emission of 86.9 million kgC, Figure 1d) are  
11 trapped in a limited area. We notice that the south-western flow, which appears stronger  
12 than the western flow, prevents the high CO<sub>2</sub> concentration in the West Coast Basin from  
13 propagating further east and dilutes into the Central Basin. Controlled by the orography,  
14 strong southerly flows occur between the Santa Monica and San Gabriel Mountains,  
15 keeping the contaminated air from propagating to the west. Driven by the same  
16 meteorology, the 1.3-km WRF-Vulcan run shows a more smeared out CO<sub>2</sub> concentration  
17 over the LA megacity (Figure 9c) due to the coarser resolution of the original Vulcan  
18 emissions. High CO<sub>2</sub> plumes seen in the 1.3-km WRF-Hestia run from point sources are  
19 replaced by wide area of the elevated CO<sub>2</sub> concentration in the 1.3-km WRF-Vulcan. The  
20 large differences in the simulated surface CO<sub>2</sub> fields between the 1.3-km WRF-Hestia and  
21 WRF-Vulcan runs are around LAX and north of the Palos Verdes Peninsula where strong  
22 point sources are located (dipole-like pattern in Figure 9d).

23

## 24 **5 Sampling density of the 2015-era GHG measurement network**

25 In this section, we present a forward network design framework, using the modelled CO<sub>2</sub>  
26 concentrations and their relationship with neighbouring grid cells. Compared to previous  
27 studies using tower footprints (i.e. linearized adjoint models) as Kort et al. (2013), we  
28 propose here a forward model assessment of the network using our high-resolution WRF  
29 results. We assume that each observation site can be associated with a specific CO<sub>2</sub> air  
30 mass at any given time. To define this CO<sub>2</sub> air mass, we estimate the spatial coherence in



1 the modelled CO<sub>2</sub> concentration fields. We constrain the coverage of each LA GHG  
2 measurement site by calculating the simultaneous correlation of the site to the rest of the  
3 domain using the simulated CO<sub>2</sub> concentration time series. Figure 10 shows the  
4 correlation map (R) of each site for the 1.3-km WRF-Hestia run. Only areas meeting a  
5 significance level of 0.01 in the t-test ( $|R| \geq 0.46$ ) are coloured. Based on the spatial  
6 patterns of the correlation maps, all of the observation sites can be grouped into (i)  
7 coastal/island sites, i.e., UCI, SCI, and Palos Verdes (right three panels in bottom row of  
8 Figure 10), (ii) western basin sites, i.e., GH, Pasadena, MWO, USC, and Compton (top  
9 row in Figure 10), (iii) eastern basin sites, (i.e., CSUF, Ontario, SB; middle row in Figure  
10 10), and (iv) desert sites, i.e., Dryden and VV (left two panels in bottom row of Figure  
11 10).

12 Not surprisingly, the coastal/island sites are mainly correlated with CO<sub>2</sub> concentration in  
13 upwind areas offshore where there is limited FFCO<sub>2</sub> contamination. The white channel  
14 from Catalina Island to the Huntington Beach area demonstrates the influence of terrain-  
15 induced flows and mountain blocking. The western basin sites are mainly correlated with  
16 CO<sub>2</sub> concentration throughout the western portion of the basin, and the eastern basin sites  
17 are mainly correlated with CO<sub>2</sub> concentration throughout the eastern portion of the basin.  
18 The desert sites are anti-correlated with the basin. CSUF also shows anti-correlation with  
19 the desert. Two reasons can explain this anti-correlation. Firstly, CO<sub>2</sub> is trapped and  
20 accumulates in the basin due to the mountain barrier. Secondly, after CO<sub>2</sub> accumulates in  
21 the basin over a certain amount of time, episodic strong sea breezes may push this basin  
22 CO<sub>2</sub> over the mountains to the desert. As a result, the basin will be relatively clean while  
23 the desert is contaminated.

24 Based on the correlation maps, we can also see how the coverage of each site varies with  
25 the FFCO<sub>2</sub> emissions data products and with the WRF model resolutions. Figure 11  
26 shows the correlation maps across the runs for the Compton, Palos Verdes, and CSUF  
27 stations. All runs use the optimal physics we determined for the LA megacity, i.e.,  
28 MYNN\_UCM. The correlation maps for each site differ with the FFCO<sub>2</sub> emissions data  
29 product used, model resolution, or their combination (Figure 11). Given that the 1.3-km  
30 WRF-Hestia is the reference run, the difference of this to the 1.3-km WRF-Vulcan run  
31 reflects the errors induced by emissions resolution. The difference between the 4-km



1 WRF-Hestia run and the 1.3-km WRF-Hestia run reflects by the model representation  
2 errors. The 4-km WRF-Vulcan run is subject to model representation errors and emission  
3 aggregation errors at the same time. For simplicity, we will not emphasize but show the  
4 comparison of the 4-km WRF-Vulcan to the others.

5 Compton is isolated from the rest of the basin in the 1.3-km WRF-Hestia run but  
6 correlated with most of the basin in the 1.3-km WRF-Vulcan run. A similar discrepancy  
7 is seen for Palos Verdes. Additionally, Palos Verdes appears to be a clean site in the 1.3-  
8 km WRF-Hestia run but dramatically contaminated in the 1.3-km WRF-Vulcan run (even  
9 correlated with the LA downtown area). For CSUF, the anti-correlation between basin  
10 and desert noted above is not visible in the 1.3-km WRF-Vulcan run. Compared to the  
11 1.3-km WRF-Hestia run, the 4-km WRF-Hestia run overall shows a somewhat larger  
12 region with significant correlation for each site.

13 To highlight the discrepancy of the spatial pattern caused by the model representation  
14 errors and emission aggregation errors in the view of the existing GHG measurement  
15 network, a composite map for each run is shown in Figure 12. These maps are  
16 constructed by determining the number of sites for which the absolute value of  $R$  is  
17 greater than 0.46 for each grid cell (i.e., colour-filled area in Figure 10 and 11).  $R=0.46$  is  
18 the critical value for the  $t$ -test at the significance level of 0.01. In the 1.3-km WRF-Hestia  
19 run (reference), the West Coastal Basin and Orange County Coastal Plain are correlated  
20 with up to 6 measurement sites. A gap appears over the Central Basin correlated with up  
21 to 3 sites due to the wind pattern (Figure 9a and 9b). The San Gabriel Mountains and  
22 Peninsular Ranges are rarely correlated to any of the sites due to the elevated terrain. The  
23 4-km WRF-Hestia run shows a similar pattern but with more sites covered over the  
24 Peninsular Ranges and the coast because of the failure to resolve topography by the 4-km  
25 model resolution.

26 In the 1.3-km WRF-Vulcan run, by contrast, a large area of the basin is correlated with  
27 most of the sites (9 sites out of 13). The Compton area is even correlated with 11 sites,  
28 which is only correlated with about two sites in the 1.3-km WRF-Hestia run. A similar  
29 contrast can be seen for the GH, USC, and Palos Verdes areas where the multiple strong  
30 point sources nearby in Hestia-LA have been aggregated into one 10 km by 10 km grid



1 cell in Vulcan (Figure 1d vs.1c). Relatively coarser FFCO<sub>2</sub> emissions artificially increase  
2 the coverage of each site, which highlights the importance of using Hestia for the CO<sub>2</sub>  
3 simulation for urban environment to represent the spatial variability in CO<sub>2</sub> and design  
4 the optimal network of surface GHG measurement.

5

## 6 **6 Discussion**

7 Isotopic tracer radiocarbon (<sup>14</sup>C) can be used for distinguishing between fossil fuel and  
8 biogenic sources of CO<sub>2</sub> (Djuricin et al., 2010; Newman et al., 2013; Newman et al.,  
9 2008; Pataki et al., 2006; Pataki et al., 2007; Levin et al., 2003; Miller et al., 2012;  
10 Turnbull et al., 2006; Turnbull et al., 2009). During CalNex-LA, two-weeks' flask  
11 samples were combined to produce two CO<sub>2</sub> samples for extracting anthropogenic and  
12 biogenic signals from the total CO<sub>2</sub> concentration. Note that the two samples for Palos  
13 Verdes were sampled from 1 May to 31 May and from 1 June to 30 June, not exactly  
14 overlapping the CalNex-LA period; the two for Pasadena were sampled from 15 May to  
15 31 May and from 1 June to 15 June, overlapping the CalNex-LA period. See Newman et  
16 al. (2015) for details about the sites and sampling information. Figure 13 presents the  
17 comparisons of the modelled and flask-sampled anthropogenic fossil fuel and biogenic  
18 CO<sub>2</sub>. From both the flask samples and model simulations, the CO<sub>2</sub> signal from the  
19 biosphere is much weaker than FFCO<sub>2</sub> in the LA megacity. The two-week flask sampled  
20 biogenic CO<sub>2</sub> is about 2 ppm on average. We notice that the 1.3-km WRF-Vulcan  
21 overestimates the FFCO<sub>2</sub> concentrations about 20 ppm over the second half of the month  
22 (Figure 13d), implying that low-resolution CO<sub>2</sub> emissions can be very critical for a coast  
23 site (complex terrain) with strong point source nearby.

24 Strong temporal variability of the simulated biogenic and FFCO<sub>2</sub> can be seen for both  
25 sites (Figure 13a,13c,13e,13g). For the Pasadena site, the 1.3-km run shows nearly flat  
26 biogenic CO<sub>2</sub> concentrations during 15 May to 30 May when the 4-km run has more  
27 variability (Figure 13e). We notice that a large botanical garden covering 207 acres (i.e.  
28 The Huntington Library) is about 1.6 km away from the Pasadena site, which may  
29 suggest that higher model resolution (1.3 km vs. 4 km) could be impacted by a change in  
30 land cover. However, there is still up to about 3-ppm discrepancy in the modelled



1 biogenic CO<sub>2</sub> from the flask samples (Figure 13f). Similar discrepancy can be seen for  
2 Palos Verdes as well (Figure 13h). Reasonably determining CO<sub>2</sub> from biogenic sources  
3 remains challenging. Additional measurements are needed to constrain biogenic fluxes.  
4 Here, we focus on FFCO<sub>2</sub> emissions that dominate local CO<sub>2</sub> signals across the basin.

5 The results presented in this paper have shown that the choice of model resolution and  
6 emission products can strongly influence the interpretation of atmospheric CO<sub>2</sub> signals.  
7 Hestia quantifies FFCO<sub>2</sub> emissions down to individual buildings and roadways, in which  
8 strong point sources create large plumes that are extremely sensitive to atmospheric  
9 transport. Reproducing dynamics realistically by the atmospheric transport model is  
10 crucial around strong point sources, such as power plants, refineries, airports, etc. For  
11 instance, a considerable number of point sources are located in Long Beach (harbours,  
12 Figure 1d), about 7 km away from Palos Verdes. In late spring and summer, Palos Verdes  
13 is a clean site, with little evidence of FFCO<sub>2</sub> emissions from the LA megacity most of the  
14 time. However, we can clearly see oftentimes Palos Verdes is simulated to be  
15 contaminated by FFCO<sub>2</sub> in all of the runs, especially during early morning (Figure 8b)  
16 due to incorrectly simulated east marine flows advecting the strong FFCO<sub>2</sub> emissions,  
17 which cannot be seen in the observations. Bias in wind speed and direction becomes  
18 critical for such a location. Palos Verdes may be challenging for the atmospheric model if  
19 used as a background site.

20 For a location like Compton with strong point sources nearby emitting CO<sub>2</sub> at 86.9  
21 million kgC per year (recorded in Hestia-LA version 1.0), a fine resolution emission  
22 product becomes very important due to the strong FFCO<sub>2</sub> gradient. A relatively coarse  
23 emission product likely produces a spurious signal due to aggregating a strong point  
24 source into a large grid cell (Figure 9b and 9c). For instance, dipole-like CO<sub>2</sub> gradients  
25 were created in the difference between the 1.3-km WRF-Vulcan and WRF-Hestia runs  
26 (Figure 9d).

27 In this paper, we focus on the spatial distribution of the CO<sub>2</sub> concentration over the LA  
28 megacity. The choice of model resolution also significantly impacts the vertical gradients  
29 of the CO<sub>2</sub> concentration as a result of the terrain resolved. The 1.3-km model runs  
30 approximates the elevation of MWO as 1129 m, while the 4 km runs is 753 m; the actual



1 elevation is 1600 m. The representation errors in the 4-km model resolution are relatively  
2 large. When there is better topographic resolution, more CO<sub>2</sub> is accumulated in the basin  
3 due to blocking by the mountains. Around noon, the model results show CO<sub>2</sub>  
4 enhancement of 10 ppm over MWO in both of the 1.3-km WRF-Vulcan and WRF-Hestia  
5 runs but only up to 3 ppm in the 4-km model runs. Additionally, because of the reasons  
6 above, reasonable sampling strategy is worth investigating for the mountain sites like  
7 MWO (e.g., Law et al., 2008). Similar problems exist for a site like Palos Verdes, since  
8 the coastline resolved varies with the model resolutions, as does the topography. Model  
9 sampling strategy is therefore recommended even at 1.3-km resolution, as no clear  
10 improvement in the meteorological evaluation was observed in horizontal.

11 Figure 10 presents the simultaneous correlation maps for each site in terms of the  
12 simulated CO<sub>2</sub> concentration time series. The coverage of the correlation maps is  
13 determined by two factors at the same time: atmospheric transport and surface fluxes.  
14 This method differs from the footprint method (Kort et al., 2013). The footprint method  
15 indicates the influence of the atmospheric transport to the location of the observation  
16 only; no emission pattern was considered. Here both transport and emissions play a role  
17 in the area covered by the observation site. Therefore, the correlation maps are subject to  
18 overestimation of the influence area versus the footprint method, due to the complicated  
19 nature of the atmospheric integrator. As an example, in Figure 10, the coloured grids of  
20 the correlation map are not necessarily *physically* related to the observation site. Those  
21 far from the site may lose the track of the initial sources. Conversely, there is definitely  
22 no *physical* influence from the uncorrelated areas to the observation site. Figure 14 shows  
23 the fraction of the total FFCO<sub>2</sub> emissions over the LA megacity as function of the number  
24 of the observation sites for all of the runs. Because of the reason above, we focus on the  
25 uncorrelated areas only. Assuming that the coverage of the GHG measurement network is  
26 not sufficient if an area is correlated to less than or equal to two sites, then ~28.9 % of  
27 FFCO<sub>2</sub> is potentially under-constrained by the current GHG measurement sites (Figure  
28 14a: WRF-Hestia 1.3-km). These areas include most of the mountains, Santa Monica Bay  
29 and the upwind coast, and the south part of the Central Basin (Figure 11), about 21.1 %  
30 of total area. However, this analysis is a qualitative assessment of the observational



1 constraint. Consideration of errors in the CO<sub>2</sub> emissions needs to be taken into account  
2 for a complete assessment of the network.

3 Figure 14 also reflects the impact of the FFCO<sub>2</sub> emissions used to simulate the CO<sub>2</sub> fields.  
4 In the 1.3-km WRF-Hestia run, there are no areas covered by more than six sites, while  
5 the 1.3-km WRF-Vulcan run shows 39.8 % of FFCO<sub>2</sub> emissions over the LA megacity to  
6 be covered by more than six sites. Additionally, the distribution appears nearly normal  
7 for the 1.3-km WRF-Vulcan run. A similar discrepancy is seen between the 4-km WRF-  
8 Hestia and WRF-Vulcan runs. These differences between the WRF-Hestia and WRF-  
9 Vulcan runs further highlight the importance of using the high-resolution FFCO<sub>2</sub>  
10 emissions product for the urban CO<sub>2</sub> simulation.

11

## 12 **7 Conclusion**

13 A set of WRF configurations varying by PBL scheme, urban surface scheme, and model  
14 resolution has been evaluated by comparing the PBL height determined by aircraft  
15 profiles and ceilometer, wind speed and wind direction measured by radar wind profiler,  
16 and surface atmospheric states measured by NWS stations. The results suggest that, there  
17 is no remarkable difference between the 4-km and 1.3-km resolution simulations in terms  
18 of atmospheric model performances in horizontal, but the 1.3-km model runs resolve the  
19 vertical gradients of wind fields and PBL height somewhat better as demonstrated. The  
20 model inter-comparisons show the model using MYNN\_UCM has overall better  
21 performance than others. Coupled to FFCO<sub>2</sub> emissions products (Hestia-LA and Vulcan  
22 2.2), a land-atmosphere modelling system was built with MYNN\_UCM for studying the  
23 heterogeneity of urban CO<sub>2</sub> emissions over the LA megacity.

24 The Vulcan and Hestia-LA FFCO<sub>2</sub> emission products were used to investigate the impact  
25 of the model representation errors and emission aggregation errors on the modelled CO<sub>2</sub>  
26 concentration. Compared to the in-situ measurements during CalNex-LA, the 1.3-km  
27 modelled CO<sub>2</sub> concentrations clearly outperform the results at 4-km resolution for  
28 capturing both the spatial distribution and the temporal variability of the urban CO<sub>2</sub>  
29 signals due to strong FFCO<sub>2</sub> emission gradients across the LA megacity, even though no  
30 clear improvement in the meteorological evaluation was observed across the basin. The



1 inter-comparison of the WRF-Hestia and WRF-Vulcan runs reinforces the importance of  
2 using high-resolution emission products to represent correct, large spatial gradients in  
3 atmospheric CO<sub>2</sub> concentrations for urban environments.

4 Based on the 1.3-km WRF-Hestia run, the coverage of the current GHG measurement  
5 site over the LA megacity was evaluated using the modelled spatial correlations. Kort et  
6 al. (2013) concluded a network of eight surface observation sites provided the minimum  
7 sampling required for accurate monitoring of FFCO<sub>2</sub> emissions in LA using Vulcan at 4-  
8 km model resolution. In this study, however, using Vulcan FFCO<sub>2</sub> emissions tend to  
9 overestimate the observational constraint spatially, suggesting that the information lies in  
10 multiple fine-scale plumes rather than a single urban dome over the Los Angeles basin.  
11 Thanks to the much finer-resolution model and FFCO<sub>2</sub> emission product Hestia-LA, the  
12 coverage of each observation site seems constrained to a more limited area. Using a high-  
13 resolution emission data product and a high-resolution model configuration is necessary  
14 for accurately assessing the urban measurement network.

15

## 16 **8 Author contributions**

17 S. Feng and T. Lauvaux designed the model experiments, evaluated the model  
18 performance, and developed the assessment of the measuring network; S. Newman  
19 provided the calibrated CO<sub>2</sub> measurements and the support for the model evaluations. P.  
20 Rao, R. Patarasuk, D. O’Keeffe, J. Huang, Y. Song, K.R. Gurney developed and prepared  
21 the Vulcan and Hestia emission products; R. Ahmadov contributed to the developments  
22 of the WRF-VPRM model and relevant guideline; A. Deng provided quality control to  
23 the observations from the National Weather Stations; L.I. Díaz-Isaac tested PBL  
24 algorithms; S. Jeong and M.L. Fischer provided the background CO<sub>2</sub> concentration for  
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27

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## 1 **References**

- 2 Ahmadov, R., Gerbig, C., Kretschmer, R., Koerner, S., Neininger, B., Dolman, A. J., and  
3 Sarrat, C.: Mesoscale covariance of transport and CO<sub>2</sub> fluxes: Evidence from  
4 observations and simulations using the WRF-VPRM coupled atmosphere-biosphere  
5 model, *Journal of Geophysical Research: Atmospheres*, 112, D22107,  
6 10.1029/2007JD008552, 2007.
- 7 Ahmadov, R., Gerbig, C., Kretschmer, R., Körner, S., Rödenbeck, C., Bousquet, P., and  
8 Ramonet, M.: Comparing high resolution WRF-VPRM simulations and two global CO<sub>2</sub>  
9 transport models with coastal tower measurements of CO<sub>2</sub>, *Biogeosciences*, 6, 807-817,  
10 10.5194/bg-6-807-2009, 2009.
- 11 Angevine, W. M., Eddington, L., Durkee, K., Fairall, C., Bianco, L., and Brioude, J.:  
12 Meteorological Model Evaluation for CalNex 2010, *Monthly Weather Review*, 140,  
13 3885-3906, 10.1175/MWR-D-12-00042.1, 2012.
- 14 Asefi-Najafabady, S., Rayner, P. J., Gurney, K. R., McRobert, A., Song, Y., Coltin, K.,  
15 Huang, J., Elvidge, C., and Baugh, K.: A multiyear, global gridded fossil fuel CO<sub>2</sub>  
16 emission data product: Evaluation and analysis of results, *Journal of Geophysical*  
17 *Research: Atmospheres*, 119, 10,213-210,231, 10.1002/2013JD021296, 2014.
- 18 Baker, D. F., Law, R. M., Gurney, K. R., Rayner, P., Peylin, P., Denning, A. S.,  
19 Bousquet, P., Bruhwiler, L., Chen, Y. H., Ciais, P., Fung, I. Y., Heimann, M., John, J.,  
20 Maki, T., Maksyutov, S., Masarie, K., Prather, M., Pak, B., Taguchi, S., and Zhu, Z.:  
21 TransCom 3 inversion intercomparison: Impact of transport model errors on the  
22 interannual variability of regional CO<sub>2</sub> fluxes, 1988–2003, *Global Biogeochemical*  
23 *Cycles*, 20, n/a-n/a, 10.1029/2004GB002439, 2006.
- 24 Baker, K. R., Misener, C., Obland, M. D., Ferrare, R. A., Scarino, A. J., and Kelly, J. T.:  
25 Evaluation of surface and upper air fine scale WRF meteorological modeling of the May  
26 and June 2010 CalNex period in California, *Atmospheric Environment*, 80, 299-309,  
27 <http://dx.doi.org/10.1016/j.atmosenv.2013.08.006>, 2013.
- 28 Bosart, L. F.: Analysis of a California Catalina Eddy Event, *Monthly Weather Review*,  
29 111, 1619-1633, 10.1175/1520-0493(1983)111<1619:AOACCE>2.0.CO;2, 1983.



- 1 Bougeault, P., and Lacarrere, P.: Parameterization of Orography-Induced Turbulence in a  
2 Mesobeta--Scale Model, *Monthly Weather Review*, 117, 1872-1890, 10.1175/1520-  
3 0493(1989)117<1872:POOITI>2.0.CO;2, 1989.
- 4 Bréon, F. M., Broquet, G., Puygrenier, V., Chevallier, F., Xueref-Remy, I., Ramonet, M.,  
5 Dieudonné, E., Lopez, M., Schmidt, M., Perrussel, O., and Ciais, P.: An attempt at  
6 estimating Paris area CO<sub>2</sub> emissions from atmospheric concentration measurements,  
7 *Atmos. Chem. Phys.*, 15, 1707-1724, 10.5194/acp-15-1707-2015, 2015.
- 8 Brioude, J., Angevine, W. M., Ahmadov, R., Kim, S. W., Evan, S., McKeen, S. A., Hsie,  
9 E. Y., Frost, G. J., Neuman, J. A., Pollack, I. B., Peischl, J., Ryerson, T. B., Holloway, J.,  
10 Brown, S. S., Nowak, J. B., Roberts, J. M., Wofsy, S. C., Santoni, G. W., Oda, T., and  
11 Trainer, M.: Top-down estimate of surface flux in the Los Angeles Basin using a  
12 mesoscale inverse modeling technique: assessing anthropogenic emissions of CO, NO<sub>x</sub>  
13 and CO<sub>2</sub> and their impacts, *Atmos. Chem. Phys.*, 13, 3661-3677, 10.5194/acp-13-3661-  
14 2013, 2013.
- 15 C40: Climate 40 Group, <http://live.c40cities.org/>, 2012.
- 16 Chen, D., Li, Q., Stutz, J., Mao, Y., Zhang, L., Pikelnaya, O., Tsai, J. Y., Haman, C.,  
17 Lefer, B., Rappenglück, B., Alvarez, S. L., Neuman, J. A., Flynn, J., Roberts, J. M.,  
18 Nowak, J. B., de Gouw, J., Holloway, J., Wagner, N. L., Veres, P., Brown, S. S.,  
19 Ryerson, T. B., Warneke, C., and Pollack, I. B.: WRF-Chem simulation of NO<sub>x</sub> and O<sub>3</sub>  
20 in the L.A. basin during CalNex-2010, *Atmospheric Environment*, 81, 421-432,  
21 <http://dx.doi.org/10.1016/j.atmosenv.2013.08.064>, 2013.
- 22 Chen, F., and Dudhia, J.: Coupling an Advanced Land Surface-Hydrology Model with  
23 the Penn State-NCAR MM5 Modeling System. Part I: Model Implementation and  
24 Sensitivity, *Monthly Weather Review*, 129, 569-585, 10.1175/1520-  
25 0493(2001)129<0569:CAALSH>2.0.CO;2, 2001.
- 26 Conil, S., and Hall, A.: Local Regimes of Atmospheric Variability: A Case Study of  
27 Southern California, *Journal of Climate*, 19, 4308-4325, 10.1175/JCLI3837.1, 2006.
- 28 Davis, C., Low-Nam, S., and Mass, C.: Dynamics of a Catalina Eddy Revealed by  
29 Numerical Simulation, *Monthly Weather Review*, 128, 2885-2904, 10.1175/1520-



- 1 0493(2000)128<2885:DOACER>2.0.CO;2, 2000.
- 2 Deng, A., Stauffer, D. R., Gaudet, B. J., Dudhia, J., Hacker, J., Bruyere, C., Wu, W.,  
3 Vandenberghe, F., Liu, Y., and Bourgeois, A.: Update on WRF-ARW End-to-End Multi-  
4 scale FDDA System, 10th Annual WRF Users' Workshop, Boulder, CO, June 23, 2009.
- 5 Díaz Isaac, L. I., Lauvaux, T., Davis, K. J., Miles, N. L., Richardson, S. J., Jacobson, A.  
6 R., and Andrews, A. E.: Model-data comparison of MCI field campaign atmospheric  
7 CO<sub>2</sub> mole fractions, *Journal of Geophysical Research: Atmospheres*, 119,  
8 2014JD021593, 10.1002/2014JD021593, 2014.
- 9 Djuricin, S., Pataki, D. E., and Xu, X.: A comparison of tracer methods for quantifying  
10 CO<sub>2</sub> sources in an urban region, *Journal of Geophysical Research: Atmospheres*, 115,  
11 n/a-n/a, 10.1029/2009JD012236, 2010.
- 12 Engelen, R. J., Denning, A. S., and Gurney, K. R.: On error estimation in atmospheric  
13 CO<sub>2</sub> inversions, *Journal of Geophysical Research: Atmospheres*, 107, 4635,  
14 10.1029/2002JD002195, 2002.
- 15 Enting, I. G., Heimann, M., Wigley, T. M. L., Commonwealth, S., and Industrial  
16 Research, O.: Future emissions and concentrations of carbon dioxide: key  
17 ocean/atmosphere/land analyses, Division of Atmospheric Research technical paper ;no.  
18 31, 120 p., CSIRO, Australia, 120 p. pp., 1994.
- 19 Etheridge, D. M., Steele, L. P., Langenfelds, R. L., Francey, R. J., Barnola, J. M., and  
20 Morgan, V. I.: Natural and anthropogenic changes in atmospheric CO<sub>2</sub> over the last 1000  
21 years from air in Antarctic ice and firn, *Journal of Geophysical Research: Atmospheres*,  
22 101, 4115-4128, 10.1029/95JD03410, 1996.
- 23 Gerbig, C., Körner, S., and Lin, J. C.: Vertical mixing in atmospheric tracer transport  
24 models: error characterization and propagation, *Atmos. Chem. Phys.*, 8, 591-602,  
25 10.5194/acp-8-591-2008, 2008.
- 26 Grell, G. A., and Dévényi, D.: A generalized approach to parameterizing convection  
27 combining ensemble and data assimilation techniques, *Geophysical Research Letters*, 29,  
28 38-31-38-34, 10.1029/2002GL015311, 2002.
- 29 Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Baker, D., Bousquet, P.,



- 1 Bruhwiler, L., Chen, Y.-H., Ciais, P., Fan, S., Fung, I. Y., Gloor, M., Heimann, M.,  
2 Higuchi, K., John, J., Maki, T., Maksyutov, S., Masarie, K., Peylin, P., Prather, M., Pak,  
3 B. C., Randerson, J., Sarmiento, J., Taguchi, S., Takahashi, T., and Yuen, C.-W.:  
4 Towards robust regional estimates of CO<sub>2</sub> sources and sinks using atmospheric transport  
5 models, *Nature*, 415, 626-630,  
6 [http://www.nature.com/nature/journal/v415/n6872/supinfo/415626a\\_S1.html](http://www.nature.com/nature/journal/v415/n6872/supinfo/415626a_S1.html), 2002.
- 7 Gurney, K. R., Mendoza, D. L., Zhou, Y., Fischer, M. L., Miller, C. C., Geethakumar, S.,  
8 and de la Rue du Can, S.: High Resolution Fossil Fuel Combustion CO<sub>2</sub> Emission Fluxes  
9 for the United States, *Environmental Science & Technology*, 43, 5535-5541,  
10 10.1021/es900806c, 2009.
- 11 Gurney, K. R., Razlivanov, I., Song, Y., Zhou, Y., Benes, B., and Abdul-Massih, M.:  
12 Quantification of Fossil Fuel CO<sub>2</sub> Emissions on the Building/Street Scale for a Large  
13 U.S. City, *Environmental Science & Technology*, 46, 12194-12202, 10.1021/es3011282,  
14 2012.
- 15 Gurney, K. R., Romero-Lankao, P., Seto, K. C., Hutyra, L. R., Duren, R., Kennedy, C.,  
16 Grimm, N. B., Ehleringer, J. R., Marcutuillio, P., Hughes, S., Pincetl, S., Chester, M. V.,  
17 Runfola, D. M., Feddema, J. J., and Sperling, J.: Climate change: Track urban emissions  
18 on a human scale citation, *Nature*, 525, 179–181, 10.1038/525179a, 2015.
- 19 Haman, C. L., Lefer, B., and Morris, G. A.: Seasonal Variability in the Diurnal Evolution  
20 of the Boundary Layer in a Near-Coastal Urban Environment, *Journal of Atmospheric  
21 and Oceanic Technology*, 29, 697-710, 10.1175/JTECH-D-11-00114.1, 2012.
- 22 Hong, S.-Y., Dudhia, J., and Chen, S.-H.: A Revised Approach to Ice Microphysical  
23 Processes for the Bulk Parameterization of Clouds and Precipitation, *Monthly Weather  
24 Review*, 132, 103-120, 10.1175/1520-0493(2004)132<0103:ARATIM>2.0.CO;2, 2004.
- 25 Hong, S.-Y., Noh, Y., and Dudhia, J.: A New Vertical Diffusion Package with an Explicit  
26 Treatment of Entrainment Processes, *Monthly Weather Review*, 134, 2318-2341,  
27 10.1175/MWR3199.1, 2006.
- 28 Houghton, R. A.: The annual net flux of carbon to the atmosphere from changes in land  
29 use 1850–1990\*, *Tellus B*, 51, 298-313, 10.1034/j.1600-0889.1999.00013.x, 1999.



- 1 Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and  
2 Collins, W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the  
3 AER radiative transfer models, *Journal of Geophysical Research: Atmospheres*, 113, n/a-  
4 n/a, 10.1029/2008JD009944, 2008.
- 5 IPCC: Climate Change 2013. The Physical Science Basis. Contribution of Working  
6 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate  
7 Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A.  
8 Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)], Cambridge University Press,  
9 Cambridge, United Kingdom and New York, NY, USA, 1535pp., 2013.
- 10 Jacob, D. J., Crawford, J. H., Maring, H., Clarke, A. D., Dibb, J. E., Emmons, L. K.,  
11 Ferrare, R. A., Hostetler, C. A., Russell, P. B., Singh, H. B., Thompson, A. M., Shaw, G.  
12 E., McCauley, E., Pederson, J. R., and Fisher, J. A.: The Arctic Research of the  
13 Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design,  
14 execution, and first results, *Atmos. Chem. Phys.*, 10, 5191-5212, 10.5194/acp-10-5191-  
15 2010, 2010.
- 16 Janjić, Z. I.: The Step-Mountain Eta Coordinate Model: Further Developments of the  
17 Convection, Viscous Sublayer, and Turbulence Closure Schemes, *Monthly Weather*  
18 *Review*, 122, 927-945, 10.1175/1520-0493(1994)122<0927:TSMECM>2.0.CO;2, 1994.
- 19 Jeong, S., Hsu, Y.-K., Andrews, A. E., Bianco, L., Vaca, P., Wilczak, J. M., and Fischer,  
20 M. L.: A multitower measurement network estimate of California's methane emissions,  
21 *Journal of Geophysical Research: Atmospheres*, 118, 11,339-311,351,  
22 10.1002/jgrd.50854, 2013.
- 23 Kort, E. A., Frankenberg, C., Miller, C. E., and Oda, T.: Space-based observations of  
24 megacity carbon dioxide, *Geophysical Research Letters*, 39, L17806,  
25 10.1029/2012GL052738, 2012.
- 26 Kort, E. A., Angevine, W. M., Duren, R., and Miller, C. E.: Surface observations for  
27 monitoring urban fossil fuel CO<sub>2</sub> emissions: Minimum site location requirements for the  
28 Los Angeles megacity, *Journal of Geophysical Research: Atmospheres*, 118, 1577-1584,  
29 10.1002/jgrd.50135, 2013.



- 1 Kretschmer, R., Gerbig, C., Karstens, U., and Koch, F. T.: Error characterization of CO<sub>2</sub>  
2 vertical mixing in the atmospheric transport model WRF-VPRM, *Atmos. Chem. Phys.*,  
3 12, 2441-2458, 10.5194/acp-12-2441-2012, 2012.
- 4 Kretschmer, R., Gerbig, C., Karstens, U., Biavati, G., Vermeulen, A., Vogel, F.,  
5 Hammer, S., and Totsche, K. U.: Impact of optimized mixing heights on simulated  
6 regional atmospheric transport of CO<sub>2</sub>, *Atmos. Chem. Phys.*, 14, 7149-7172,  
7 10.5194/acp-14-7149-2014, 2014.
- 8 Kusaka, H., Kondo, H., Kikegawa, Y., and Kimura, F.: A Simple Single-Layer Urban  
9 Canopy Model For Atmospheric Models: Comparison With Multi-Layer And Slab  
10 Models, *Boundary-Layer Meteorol.*, 101, 329-358, 10.1023/A:1019207923078, 2001.
- 11 Kusaka, H., and Kimura, F.: Thermal Effects of Urban Canyon Structure on the  
12 Nocturnal Heat Island: Numerical Experiment Using a Mesoscale Model Coupled with  
13 an Urban Canopy Model, *Journal of Applied Meteorology*, 43, 1899-1910,  
14 10.1175/JAM2169.1, 2004a.
- 15 Kusaka, H., and Kimura, F.: Coupling a Single-Layer Urban Canopy Model with a  
16 Simple Atmospheric Model: Impact on Urban Heat Island Simulation for an Idealized  
17 Case, *Journal of the Meteorological Society of Japan. Ser. II*, 82, 67-80,  
18 10.2151/jmsj.82.67, 2004b.
- 19 Lac, C., Donnelly, R. P., Masson, V., Pal, S., Riette, S., Donier, S., Queguiner, S.,  
20 Tanguy, G., Ammoura, L., and Xueref-Remy, I.: CO<sub>2</sub> dispersion modelling over Paris  
21 region within the CO<sub>2</sub>-MEGAPARIS project, *Atmos. Chem. Phys.*, 13, 4941-4961,  
22 10.5194/acp-13-4941-2013, 2013.
- 23 Lauvaux, T., Uliasz, M., Sarrat, C., Chevallier, F., Bousquet, P., Lac, C., Davis, K. J.,  
24 Ciais, P., Denning, A. S., and Rayner, P. J.: Mesoscale inversion: first results from the  
25 CERES campaign with synthetic data, *Atmos. Chem. Phys.*, 8, 3459-3471, 10.5194/acp-  
26 8-3459-2008, 2008.
- 27 Lauvaux, T., Pannekoucke, O., Sarrat, C., Chevallier, F., Ciais, P., Noilhan, J., and  
28 Rayner, P. J.: Structure of the transport uncertainty in mesoscale inversions of CO<sub>2</sub>  
29 sources and sinks using ensemble model simulations, *Biogeosciences*, 6, 1089-1102,



- 1 10.5194/bg-6-1089-2009, 2009.
- 2 Lauvaux, T., Schuh, A. E., Bocquet, M., Wu, L., Richardson, S., Miles, N., and Davis, K.  
3 J.: Network design for mesoscale inversions of CO<sub>2</sub> sources and sinks, 2012, 64,  
4 10.3402/tellusb.v64i0.17980, 2012.
- 5 Lauvaux, T., Miles, N. L., Deng, A., Richardson, S. J., Cambaliza, M. O., Davis, K. J.,  
6 Gaudet, B., Gurney, K. R., Huang, J., Karion, A., Oda, T., Patarasuk, R., Razlivanov, I.,  
7 Sarmiento, D., Shepson, P. B., Sweeney, C., Turnbull, J. C., and Wu, K.: High resolution  
8 atmospheric inversion of urban CO<sub>2</sub> emissions during the dormant season of the  
9 Indianapolis Flux Experiment (INFLUX), Journal of Geophysical Research:  
10 Atmospheres, *accepted*.
- 11 Law, R. M., Rayner, P. J., Steele, L. P., and Enting, I. G.: Data and modelling  
12 requirements for CO<sub>2</sub> inversions using high-frequency data, Tellus B, 55, 512-521,  
13 10.1034/j.1600-0889.2003.00029.x, 2003.
- 14 Law, R. M., Peters, W., Rödenbeck, C., Aulagnier, C., Baker, I., Bergmann, D. J.,  
15 Bousquet, P., Brandt, J., Bruhwiler, L., Cameron-Smith, P. J., Christensen, J. H., Delage,  
16 F., Denning, A. S., Fan, S., Geels, C., Houweling, S., Imasu, R., Karstens, U., Kawa, S.  
17 R., Kleist, J., Krol, M. C., Lin, S. J., Lokupitiya, R., Maki, T., Maksyutov, S., Niwa, Y.,  
18 Onishi, R., Parazoo, N., Patra, P. K., Pieterse, G., Rivier, L., Satoh, M., Serrar, S.,  
19 Taguchi, S., Takigawa, M., Vautard, R., Vermeulen, A. T., and Zhu, Z.: TransCom model  
20 simulations of hourly atmospheric CO<sub>2</sub>: Experimental overview and diurnal cycle results  
21 for 2002, Global Biogeochemical Cycles, 22, n/a-n/a, 10.1029/2007GB003050, 2008.
- 22 Le Quéré, C., Peters, G. P., Andres, R. J., Andrew, R. M., Boden, T. A., Ciais, P.,  
23 Friedlingstein, P., Houghton, R. A., Marland, G., Moriarty, R., Sitch, S., Tans, P., Arneth,  
24 A., Arvanitis, A., Bakker, D. C. E., Bopp, L., Canadell, J. G., Chini, L. P., Doney, S. C.,  
25 Harper, A., Harris, I., House, J. I., Jain, A. K., Jones, S. D., Kato, E., Keeling, R. F.,  
26 Klein Goldewijk, K., Körtzinger, A., Koven, C., Lefèvre, N., Maignan, F., Omar, A.,  
27 Ono, T., Park, G. H., Pfeil, B., Poulter, B., Raupach, M. R., Regnier, P., Rödenbeck, C.,  
28 Saito, S., Schwinger, J., Segschneider, J., Stocker, B. D., Takahashi, T., Tilbrook, B., van  
29 Heuven, S., Viovy, N., Wanninkhof, R., Wiltshire, A., and Zaehle, S.: Global carbon  
30 budget 2013, Earth Syst. Sci. Data, 6, 235-263, 10.5194/essd-6-235-2014, 2014.





- 1 Levin, I., Kromer, B., Schmidt, M., and Sartorius, H.: A novel approach for independent  
2 budgeting of fossil fuel CO<sub>2</sub> over Europe by 14CO<sub>2</sub> observations, *Geophysical Research*  
3 *Letters*, 30, n/a-n/a, 10.1029/2003GL018477, 2003.
- 4 Lu, R., and Turco, R. P.: Air pollutant transport in a coastal environment—II. Three-  
5 dimensional simulations over Los Angeles basin, *Atmospheric Environment*, 29, 1499-  
6 1518, [http://dx.doi.org/10.1016/1352-2310\(95\)00015-Q](http://dx.doi.org/10.1016/1352-2310(95)00015-Q), 1995.
- 7 Mahadevan, P., Wofsy, S. C., Matross, D. M., Xiao, X., Dunn, A. L., Lin, J. C., Gerbig,  
8 C., Munger, J. W., Chow, V. Y., and Gottlieb, E. W.: A satellite-based biosphere  
9 parameterization for net ecosystem CO<sub>2</sub> exchange: Vegetation Photosynthesis and  
10 Respiration Model (VPRM), *Global Biogeochemical Cycles*, 22, GB2005,  
11 10.1029/2006GB002735, 2008.
- 12 Description of the modifications made in WRF.3.1 and short user's manual of BEP,  
13 2009.
- 14 Mesinger, F., DiMego, G., Kalnay, E., Mitchell, K., Shafran, P. C., Ebisuzaki, W., Jović,  
15 D., Woollen, J., Rogers, E., Berbery, E. H., Ek, M. B., Fan, Y., Grumbine, R., Higgins,  
16 W., Li, H., Lin, Y., Manikin, G., Parrish, D., and Shi, W.: North American Regional  
17 Reanalysis, *Bulletin of the American Meteorological Society*, 87, 343-360,  
18 10.1175/BAMS-87-3-343, 2006.
- 19 Miller, J. B., Lehman, S. J., Montzka, S. A., Sweeney, C., Miller, B. R., Karion, A.,  
20 Wolak, C., Dlugokencky, E. J., Southon, J., Turnbull, J. C., and Tans, P. P.: Linking  
21 emissions of fossil fuel CO<sub>2</sub> and other anthropogenic trace gases using atmospheric  
22 14CO<sub>2</sub>, *Journal of Geophysical Research: Atmospheres*, 117, n/a-n/a,  
23 10.1029/2011JD017048, 2012.
- 24 Nakanishi, M., and Niino, H.: An Improved Mellor–Yamada Level-3 Model: Its  
25 Numerical Stability and Application to a Regional Prediction of Advection Fog,  
26 *Boundary-Layer Meteorol*, 119, 397-407, 10.1007/s10546-005-9030-8, 2006.
- 27 Nehr Korn, T., Henderson, J., Leidner, M., Mountain, M., Eluszkiewicz, J., McKain, K.,  
28 and Wofsy, S.: WRF Simulations of the Urban Circulation in the Salt Lake City Area for  
29 CO<sub>2</sub> Modeling, *Journal of Applied Meteorology and Climatology*, 52, 323-340,



- 1 10.1175/JAMC-D-12-061.1, 2012.
- 2 Newman, S., Xu, X., Affek, H. P., Stolper, E., and Epstein, S.: Changes in mixing ratio  
3 and isotopic composition of CO<sub>2</sub> in urban air from the Los Angeles basin, California,  
4 between 1972 and 2003, *Journal of Geophysical Research: Atmospheres*, 113, n/a-n/a,  
5 10.1029/2008JD009999, 2008.
- 6 Newman, S., Jeong, S., Fischer, M. L., Xu, X., Haman, C. L., Lefer, B., Alvarez, S.,  
7 Rappenglueck, B., Kort, E. A., Andrews, A. E., Peischl, J., Gurney, K. R., Miller, C. E.,  
8 and Yung, Y. L.: Diurnal tracking of anthropogenic CO<sub>2</sub> emissions in the Los Angeles  
9 basin megacity during spring 2010, *Atmos. Chem. Phys.*, 13, 4359-4372, 10.5194/acp-  
10 13-4359-2013, 2013.
- 11 Newman, S., Xu, X., Gurney, K. R., Hsu, Y. K., Li, K. F., Jiang, X., Keeling, R. F., Feng,  
12 S., O'Keefe, D., Patarasuk, R., Wong, K. W., Rao, P., Fisher, M. L., and Yung, Y. L.:  
13 Toward consistency between bottom-up CO<sub>2</sub> emissions trends and top-down atmospheric  
14 measurements in the Los Angeles megacity, *Atmos. Chem. Phys. Discuss.*, 1-45,  
15 10.5194/acpd-15-1-2015, 2015.
- 16 Pataki, D. E., Alig, R. J., Fung, A. S., Golubiewski, N. E., Kennedy, C. A., McPherson,  
17 E. G., Nowak, D. J., Pouyat, R. V., and Romero Lankao, P.: Urban ecosystems and the  
18 North American carbon cycle, *Global Change Biology*, 12, 2092-2102, 10.1111/j.1365-  
19 2486.2006.01242.x, 2006.
- 20 Pataki, D. E., Xu, T., Luo, Y. Q., and Ehleringer, J. R.: Inferring biogenic and  
21 anthropogenic carbon dioxide sources across an urban to rural gradient, *Oecologia*, 152,  
22 307-322, 10.1007/s00442-006-0656-0, 2007.
- 23 Pillai, D., Gerbig, C., Marshall, J., Ahmadov, R., Kretschmer, R., Koch, T., and Karstens,  
24 U.: High resolution modeling of CO<sub>2</sub> over Europe: implications for representation errors  
25 of satellite retrievals, *Atmos. Chem. Phys.*, 10, 83-94, 10.5194/acp-10-83-2010, 2010.
- 26 Pillai, D., Gerbig, C., Ahmadov, R., Rödenbeck, C., Kretschmer, R., Koch, T.,  
27 Thompson, R., Neininger, B., and Lavrié, J. V.: High-resolution simulations of  
28 atmospheric CO<sub>2</sub> over complex terrain – representing the Ochsenkopf mountain tall  
29 tower, *Atmos. Chem. Phys.*, 11, 7445-7464, 10.5194/acp-11-7445-2011, 2011.



- 1 Rao, P., Gurney, K. R., Patarasuk, R., Song, Y., Miller, C. E., Duren, R. M., and  
2 Eldering, A.: Spatio-temporal Variations in Onroad Vehicle Fossil Fuel CO<sub>2</sub> Emissions  
3 in the Los Angeles Megacity, *Environmental Science and Technology*, submitted, 2015.
- 4 Riley, W. J., Hsueh, D. Y., Randerson, J. T., Fischer, M. L., Hatch, J. G., Pataki, D. E.,  
5 Wang, W., and Goulden, M. L.: Where do fossil fuel carbon dioxide emissions from  
6 California go? An analysis based on radiocarbon observations and an atmospheric  
7 transport model, *Journal of Geophysical Research: Biogeosciences*, 113, n/a-n/a,  
8 10.1029/2007JG000625, 2008.
- 9 Rödenbeck, C., Gerbig, C., Trusilova, K., and Heimann, M.: A two-step scheme for high-  
10 resolution regional atmospheric trace gas inversions based on independent models,  
11 *Atmos. Chem. Phys.*, 9, 5331-5342, 10.5194/acp-9-5331-2009, 2009.
- 12 Rogers, R. E., Deng, A., Stauffer, D. R., Gaudet, B. J., Jia, Y., Soong, S.-T., and  
13 Tanrikulu, S.: Application of the Weather Research and Forecasting Model for Air  
14 Quality Modeling in the San Francisco Bay Area, *Journal of Applied Meteorology and  
15 Climatology*, 52, 1953-1973, 10.1175/JAMC-D-12-0280.1, 2013.
- 16 Ryerson, T. B., Andrews, A. E., Angevine, W. M., Bates, T. S., Brock, C. A., Cairns, B.,  
17 Cohen, R. C., Cooper, O. R., de Gouw, J. A., Fehsenfeld, F. C., Ferrare, R. A., Fischer,  
18 M. L., Flagan, R. C., Goldstein, A. H., Hair, J. W., Hardesty, R. M., Hostetler, C. A.,  
19 Jimenez, J. L., Langford, A. O., McCauley, E., McKeen, S. A., Molina, L. T., Nenes, A.,  
20 Oltmans, S. J., Parrish, D. D., Pederson, J. R., Pierce, R. B., Prather, K., Quinn, P. K.,  
21 Seinfeld, J. H., Senff, C. J., Sorooshian, A., Stutz, J., Surratt, J. D., Trainer, M.,  
22 Volkamer, R., Williams, E. J., and Wofsy, S. C.: The 2010 California Research at the  
23 Nexus of Air Quality and Climate Change (CalNex) field study, *Journal of Geophysical  
24 Research: Atmospheres*, 118, 5830-5866, 10.1002/jgrd.50331, 2013.
- 25 Sarrat, C., Noilhan, J., Dolman, A. J., Gerbig, C., Ahmadov, R., Tolk, L. F., Meesters, A.  
26 G. C. A., Hutjes, R. W. A., Ter Maat, H. W., Pérez-Landa, G., and Donier, S.:  
27 Atmospheric CO<sub>2</sub> modeling at the regional scale: an intercomparison of 5 meso-scale  
28 atmospheric models, *Biogeosciences*, 4, 1115-1126, 10.5194/bg-4-1115-2007, 2007.
- 29 Scarino, A. J., Obland, M. D., Fast, J. D., Burton, S. P., Ferrare, R. A., Hostetler, C. A.,



- 1 Berg, L. K., Lefer, B., Haman, C., Hair, J. W., Rogers, R. R., Butler, C., Cook, A. L., and  
2 Harper, D. B.: Comparison of mixed layer heights from airborne high spectral resolution  
3 lidar, ground-based measurements, and the WRF-Chem model during CalNex and  
4 CARES, Atmos. Chem. Phys. Discuss., 13, 13721-13772, 10.5194/acpd-13-13721-2013,  
5 2013.
- 6 Strong, C., Stwertka, C., Bowling, D. R., Stephens, B. B., and Ehleringer, J. R.: Urban  
7 carbon dioxide cycles within the Salt Lake Valley: A multiple-box model validated by  
8 observations, Journal of Geophysical Research: Atmospheres, 116, n/a-n/a,  
9 10.1029/2011JD015693, 2011.
- 10 Tarantola, A.: Inverse problem theory and methods for model parameter estimation,  
11 Book, Whole, Society for Industrial and Applied Mathematics, Philadelphia, PA, 2005.
- 12 Turnbull, J., Rayner, P., Miller, J., Naegler, T., Ciais, P., and Cozic, A.: On the use of  
13  $^{14}\text{CO}_2$  as a tracer for fossil fuel  $\text{CO}_2$ : Quantifying uncertainties using an atmospheric  
14 transport model, Journal of Geophysical Research: Atmospheres, 114, n/a-n/a,  
15 10.1029/2009JD012308, 2009.
- 16 Turnbull, J. C., Miller, J. B., Lehman, S. J., Tans, P. P., Sparks, R. J., and Southon, J.:  
17 Comparison of  $^{14}\text{CO}_2$ , CO, and SF<sub>6</sub> as tracers for recently added fossil fuel CO<sub>2</sub> in the  
18 atmosphere and implications for biological CO<sub>2</sub> exchange, Geophysical Research  
19 Letters, 33, n/a-n/a, 10.1029/2005GL024213, 2006.
- 20 Turnbull, J. C., Karion, A., Fischer, M. L., Faloona, I., Guilderson, T., Lehman, S. J.,  
21 Miller, B. R., Miller, J. B., Montzka, S., Sherwood, T., Saripalli, S., Sweeney, C., and  
22 Tans, P. P.: Assessment of fossil fuel carbon dioxide and other anthropogenic trace gas  
23 emissions from airborne measurements over Sacramento, California in spring 2009,  
24 Atmos. Chem. Phys., 11, 705-721, 10.5194/acp-11-705-2011, 2011.
- 25 Ulrickson, B. L., and Mass, C. F.: Numerical Investigation of Mesoscale Circulations  
26 over the Los Angeles Basin. Part II: Synoptic Influences and Pollutant Transport,  
27 Monthly Weather Review, 118, 2162-2184, 10.1175/1520-  
28 0493(1990)118<2162:NIOMCO>2.0.CO;2, 1990.
- 29 UN: World Urbanization Prospects e Revision 2005, Factsheet 7: Mega-cities, 2006.



- 1 United Nations, Department of Economic and Social Affairs, Population Division. World  
2 Urbanization Prospects: The 2005 Revision. Working Paper No. ESA/P/WP/200, 2006.
- 3 UN: World Urbanization Prospects: The 2009 Revision, 2010.
- 4 Wennberg, P. O., Mui, W., Wunch, D., Kort, E. A., Blake, D. R., Atlas, E. L., Santoni, G.  
5 W., Wofsy, S. C., Diskin, G. S., Jeong, S., and Fischer, M. L.: On the Sources of  
6 Methane to the Los Angeles Atmosphere, *Environmental Science & Technology*, 46,  
7 9282-9289, 10.1021/es301138y, 2012.
- 8 Wong, K. W., Fu, D., Pongetti, T. J., Newman, S., Kort, E. A., Duren, R., Hsu, Y. K.,  
9 Miller, C. E., Yung, Y. L., and Sander, S. P.: Mapping CH<sub>4</sub> : CO<sub>2</sub> ratios in Los Angeles  
10 with CLARS-FTS from Mount Wilson, California, *Atmos. Chem. Phys.*, 15, 241-252,  
11 10.5194/acp-15-241-2015, 2015.
- 12 Wu, L., Bocquet, M., Lauvaux, T., Chevallier, F., Rayner, P., and Davis, K.: Optimal  
13 representation of source-sink fluxes for mesoscale carbon dioxide inversion with  
14 synthetic data, *Journal of Geophysical Research: Atmospheres*, 116, n/a-n/a,  
15 10.1029/2011JD016198, 2011.
- 16 Wunch, D., Wennberg, P. O., Toon, G. C., Keppel-Aleks, G., and Yavin, Y. G.:  
17 Emissions of greenhouse gases from a North American megacity, *Geophysical Research*  
18 *Letters*, 36, L15810, 10.1029/2009GL039825, 2009.
- 19 Xiao, X., Hollinger, D., Aber, J., Goltz, M., Davidson, E. A., Zhang, Q., and Moore Iii,  
20 B.: Satellite-based modeling of gross primary production in an evergreen needleleaf  
21 forest, *Remote Sensing of Environment*, 89, 519-534,  
22 <http://dx.doi.org/10.1016/j.rse.2003.11.008>, 2004.
- 23 Yver, C. E., Graven, H. D., Lucas, D. D., Cameron-Smith, P. J., Keeling, R. F., and  
24 Weiss, R. F.: Evaluating transport in the WRF model along the California coast, *Atmos.*  
25 *Chem. Phys.*, 13, 1837-1852, 10.5194/acp-13-1837-2013, 2013.
- 26 Zhou, Y., and Gurney, K.: A new methodology for quantifying on-site residential and  
27 commercial fossil fuel CO<sub>2</sub> emissions at the building spatial scale and hourly time scale,  
28 *Carbon Management*, 1, 45-56, 10.4155/cmt.10.7, 2010.
- 29



Table 1. Common elements of the WRF-Chem configuration used in all runs.

Option	Description
Microphysics	WSM5 (Hong et al., 2004)
Longwave radiation	RRTMG (Iacono et al., 2008)
Shortwave radiation	RRTMG (Iacono et al., 2008)
Land surface	Noah land surface model (Chen and Dudhia, 2001)
Cumulus scheme	Grell-3 (Grell and Dévényi, 2002) applied to 12-km domain (d01) only

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Table 2. WRF configurations used for the sensitivity runs.

Configuration	PBL scheme	Urban surface scheme	Grid spacing (km)
BouLac_BEP_d02	BouLac	BEP	4
BouLac_BEP_d03	BouLac	BEP	1.3
BouLac_UCM_d02	BouLac	UCM	4
BouLac_UCM_d03	BouLac	UCM	1.3
MYJ_d02	MYJ	None	4
MYN_d03	MYJ	None	1.3
MYJ_UCM_d02	MYJ	UCM	4
MYJ_UCM_d03	MYJ	UCM	1.3
MYNN_d02	MYNN	None	4
MYNN_d03	MYNN	None	1.3
MYNN_UCM_d02	MYNN	UCM	4
MYNN_UCM_d03	MYNN	UCM	1.3

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Table 3. Locations of the 2015-era GHG measurement sites in the model domain<sup>Ⓜ</sup>

Code*	Name	Type	Lat. (° N)	Lon. (° E)
GH	Granada Hills	Tower	34.28	-118.47
Pasadena	Pasadena	Building top	34.14	-118.13
MWO	Mt. Wilson	Mountain top	34.22	-118.06
USC	University of South California	Building top	34.02	-118.29
Compton	Compton	Tower	33.87	-118.28
CSUF	California State University, Fullerton	Building top	33.88	-117.88
Ontario	Ontario	Tower	34.06	-117.58
SB	San Bernardino	Tower	34.09	-118.35
Dryden <sup>†</sup>	Dryden	TCCON	34.95	-117.89
VV	Victorville	Tower	34.61	-117.29
UCI	University of California, Irvine	Building top	33.64	-117.84
SCI	San Clemente Island	Tower	32.92	-118.49
PV	Palos Verdes	In-situ non-standard	33.74	-118.35

\*La Jolla site is operating but not included in this paper

\*Codes used in this paper

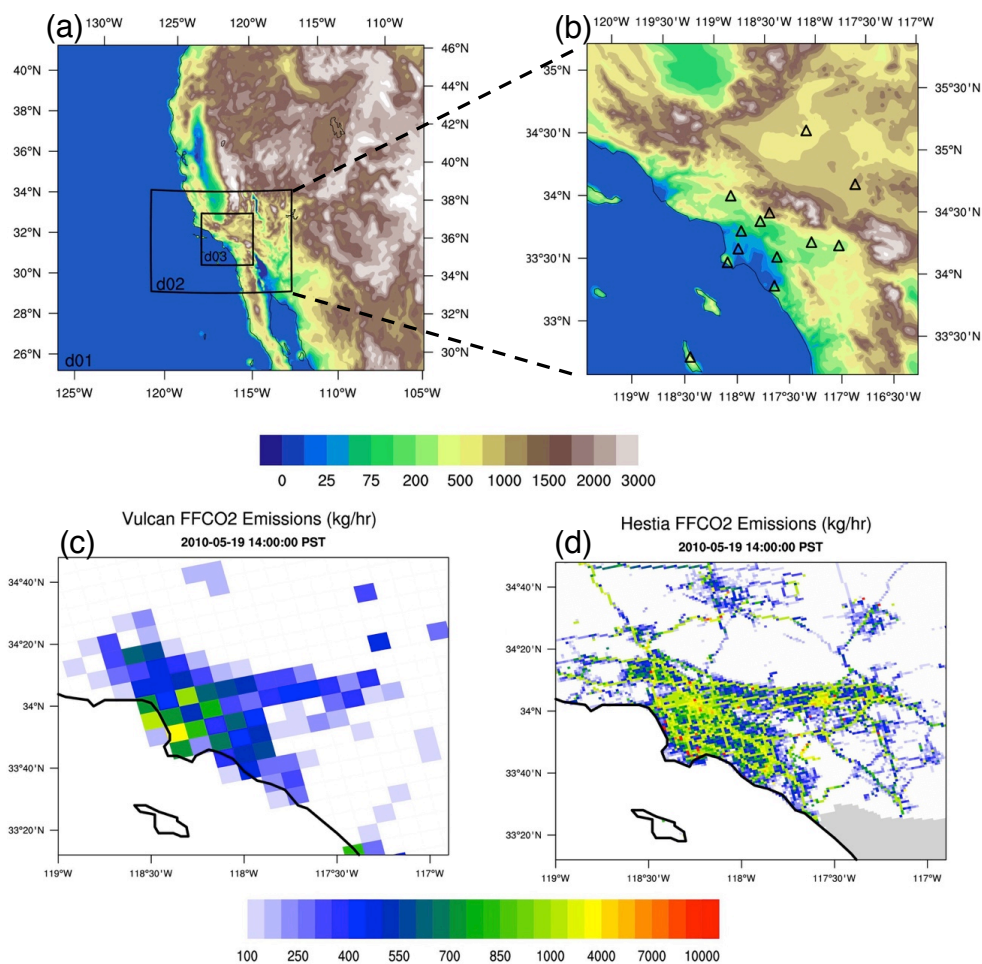
<sup>†</sup> In the analysis, we assume Dryden site is a near-surface point measurement like other sites rather than a column observation for simplicity.

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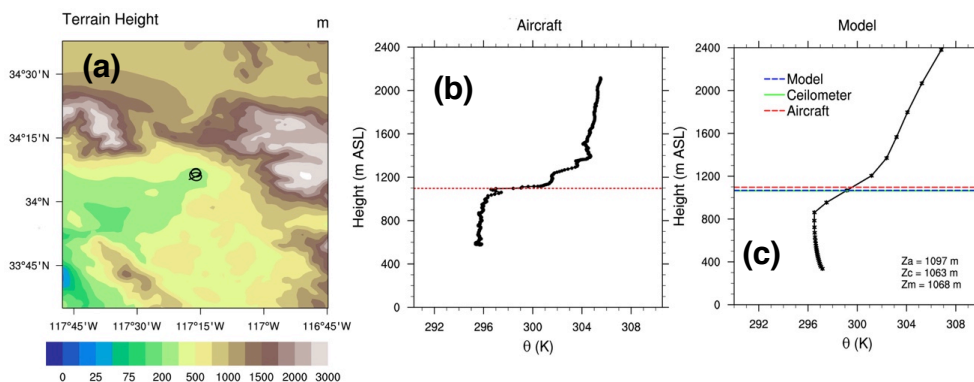
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3 Figure 1. (a) Model domains. Contours are terrain height (unit: m). (b) The 1.3-km  
4 model domain (d03) and terrain height (unit: m). Triangles represent the locations of the  
5 GHG measurement sites. (c and d) Snapshots of the Vulcan and Hestia FFCO<sub>2</sub> emissions  
6 (unit: kg/hr) over the LA megacity at 14:00 PST on 15 May 2010.

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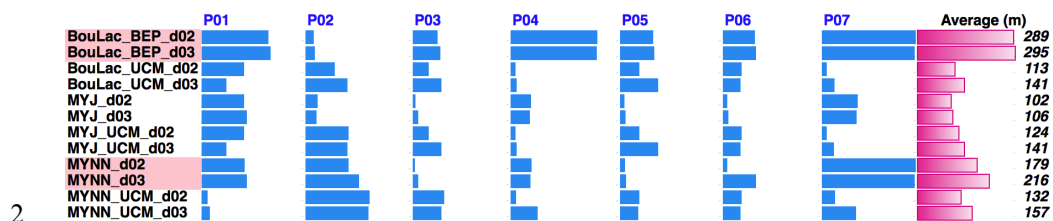
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4 Figure 2. A case selected on 19 May 2010 at 12:25 (PST) (a) Location of the vertical  
5 profile flown by the CalNex aircraft and the neighbouring terrain heights (units: m). (b)  
6 In-situ potential temperature profile measured by the aircraft . The red dashed line at  
7 ~1100 m is the PBL height calculated based on the vertical gradient of potential  
8 temperature  $\Theta$ (K). (c) Modelled potential temperature profile from the  
9 MYNN\_UCM\_d02 configuration. The red dashed line is the aircraft-determined PBL  
10 height ( $Z_a$  in masl). The solid green line is the PBL height measured by the Caltech  
11 ceilometer ( $Z_c$  in masl). The blue line is the modelled PBL height ( $Z_m$  in m).

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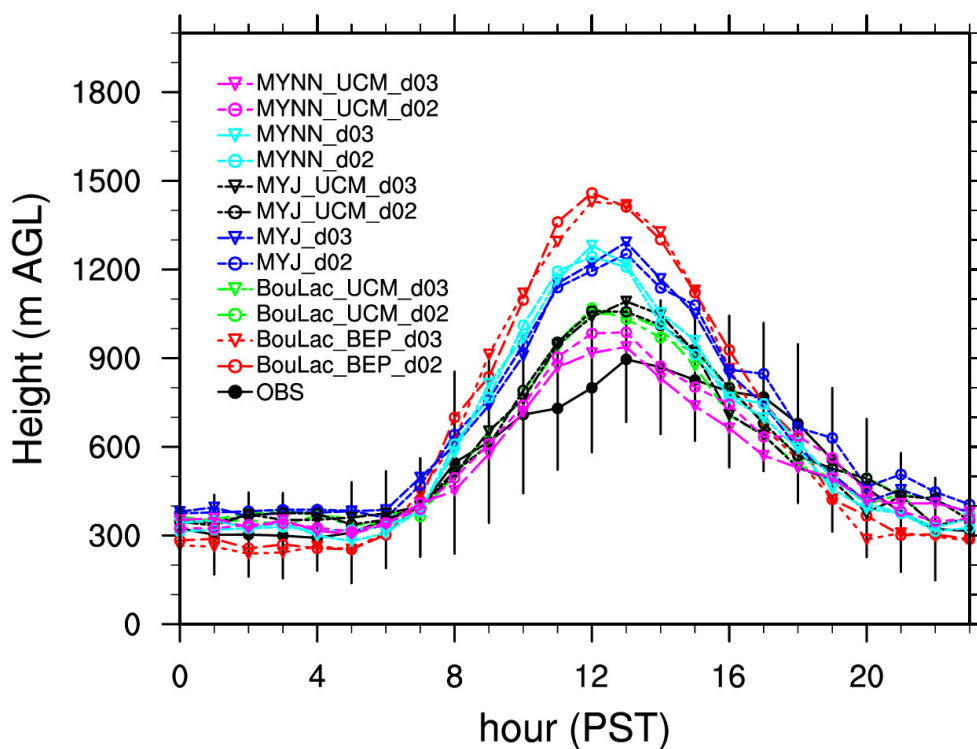
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4 Figure 3. Absolute difference between the aircraft-determined and modelled PBL height  
 5 for each profile: P01, P02, ..., and P07 (blue bars). The pink bars in the last column  
 6 represent the averaged bias over all of the profiles for each configuration. Note that the  
 7 shorter the bar is, the better agreement the model has with the observations.

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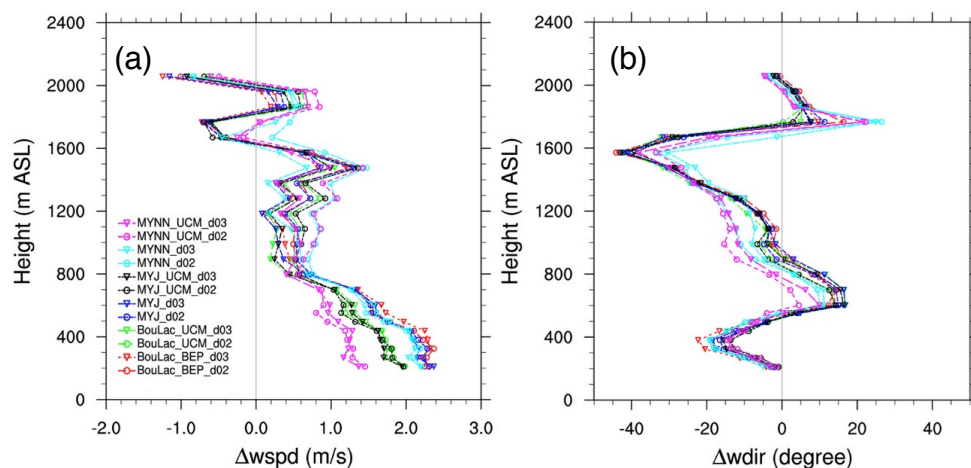


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3 Figure 4. Average diurnal variation of the ceilometer-measured and modelled PBL  
4 heights at California Institute of Technology (Caltech) in Pasadena, CA during 15 May  
5 through 15 June 2010. Error bars indicate standard deviations of the means of the  
6 ceilometer measurement.

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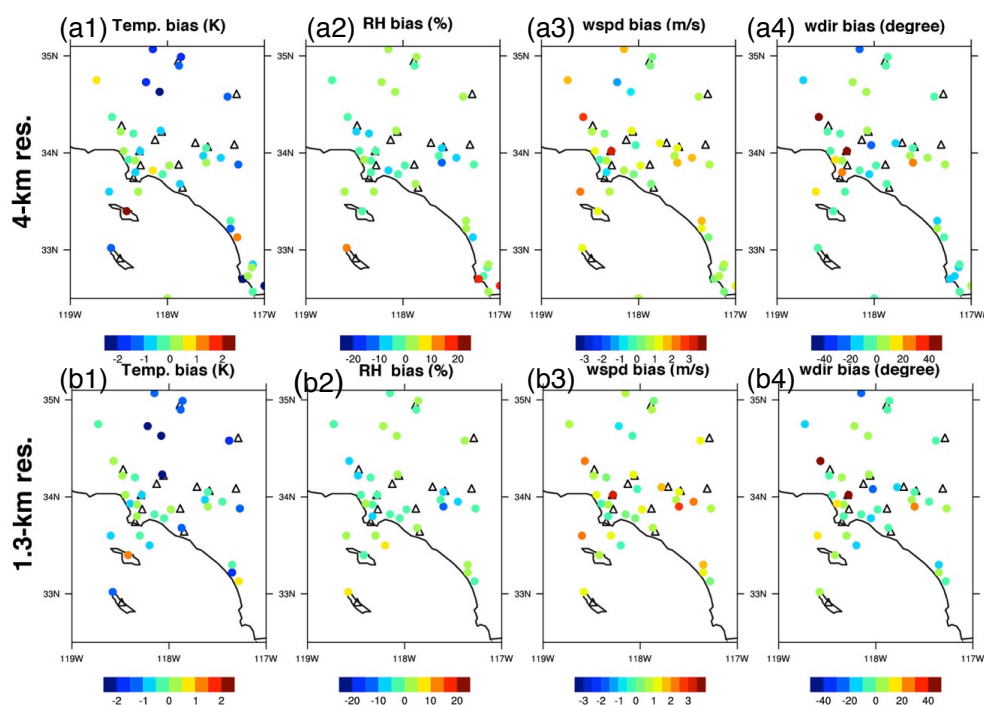
3 Figure 5. Average differences of wind profiles between the simulations and observations  
4 (model – wind radar profiler) at the Los Angeles International Airport (LAX). (a) The  
5 difference for wind speed (unit: m/s); (b) for wind direction (unit: degree).

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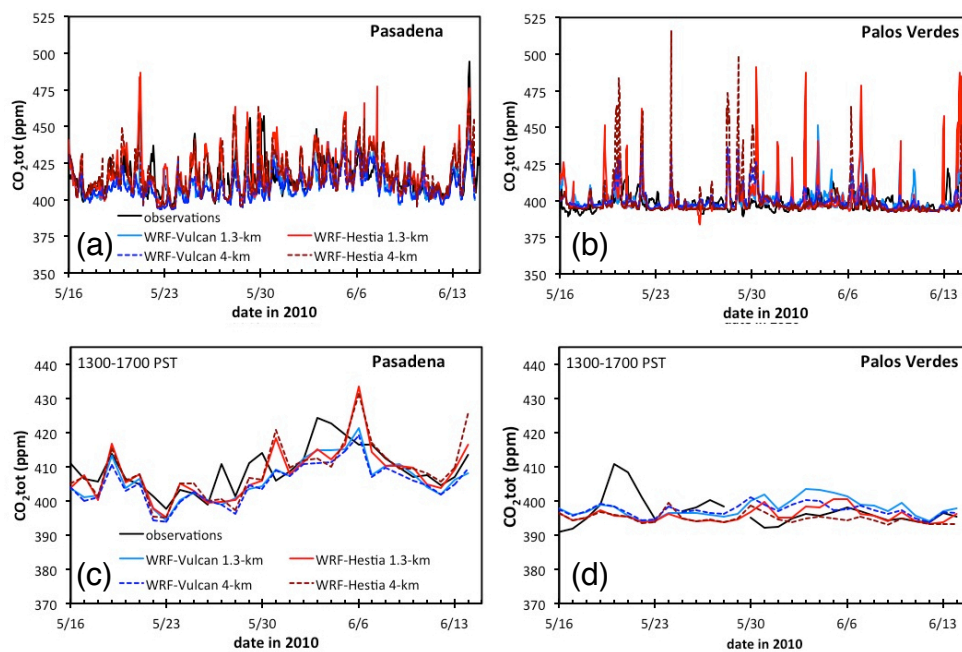
4 Figure 6. Bias maps of the MYNN\_UCM runs versus National Weather Stations (NWS)  
5 over the LA megacity (Model – NWS): (a1-a4) 4-km run; (b1 – b4) 1.3-km run. Black  
6 triangles indicate the locations of the GHG measurement sites.

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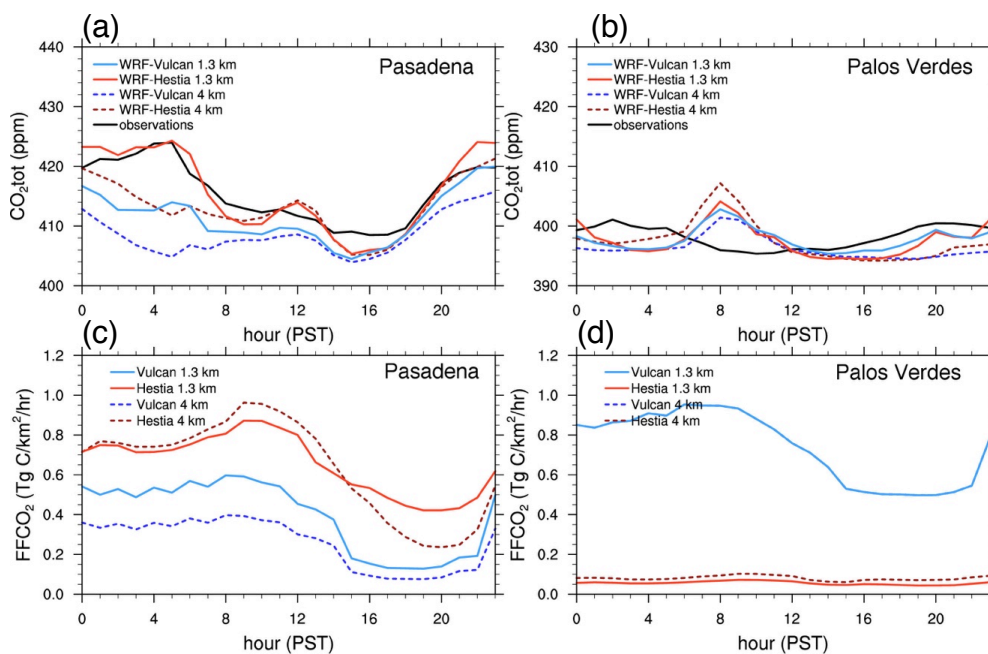
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3 Figure 7. Comparison of the observed and modelled CO<sub>2</sub> concentrations at the (a and c)  
4 Pasadena and (b and d) Palos Verdes sites: (a and b) is hourly time series, (c and d) is  
5 daily afternoon average over 1300 – 1700 PST.

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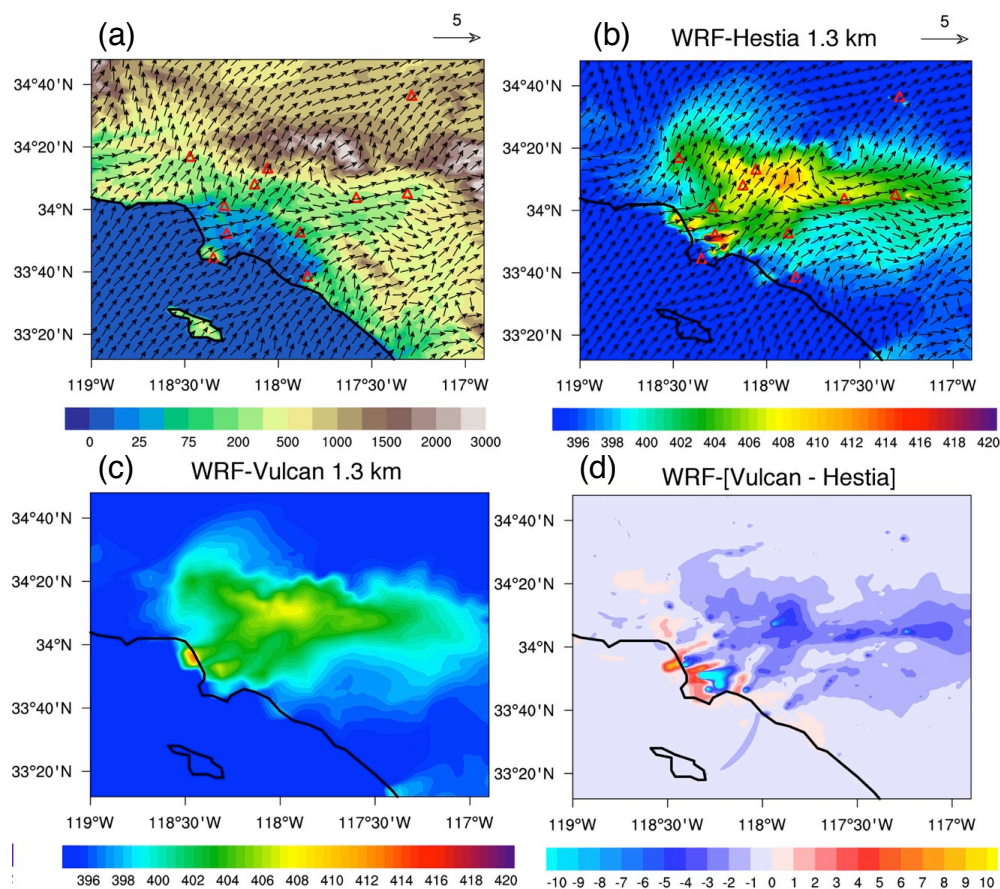
3 Figure 8. Averaged diurnal variation of observed and modelled CO<sub>2</sub> concentration and  
4 FFCO<sub>2</sub> emissions for the (a and c) Pasadena and (b and d) Palos Verdes sites during  
5 CalNex-LA.

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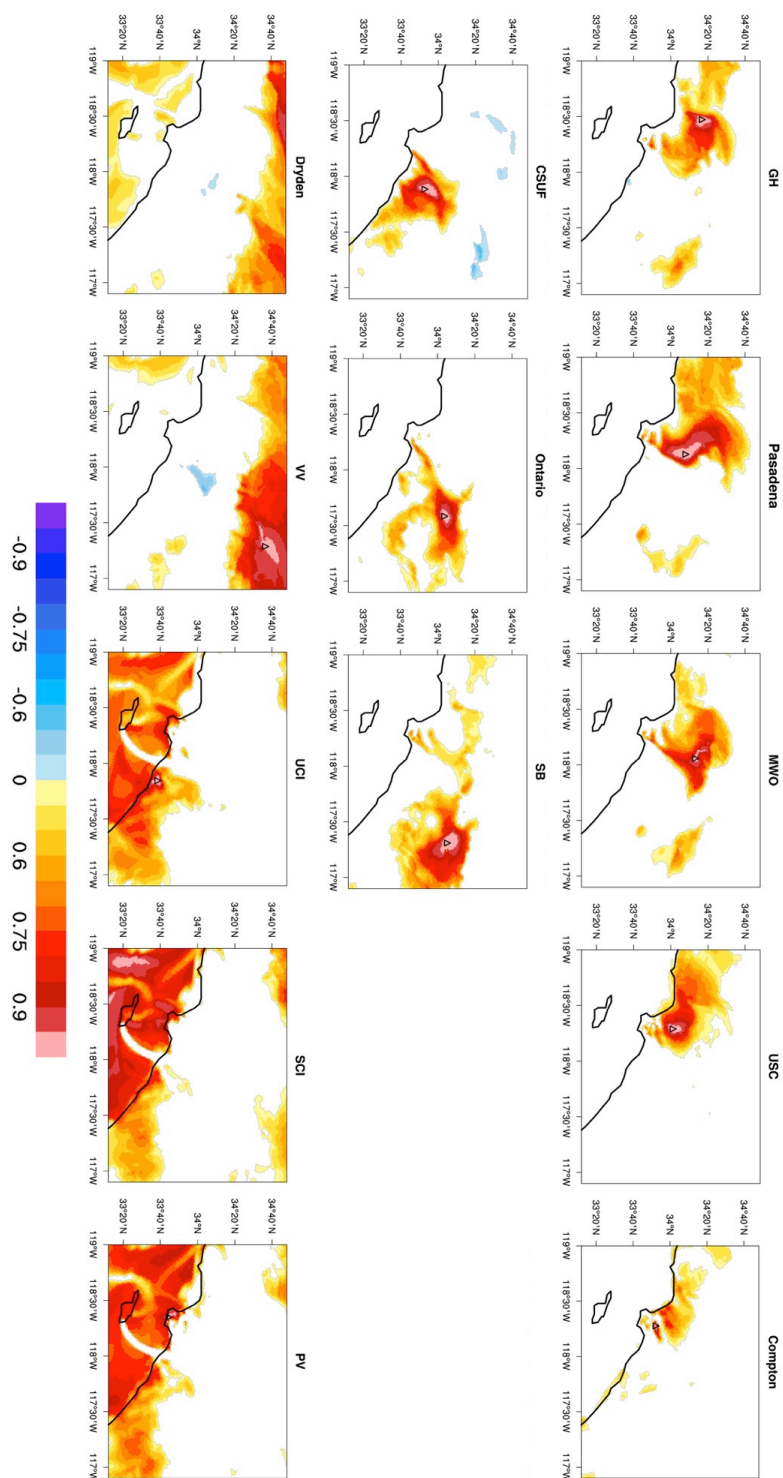


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3 Figure 9. (a and b) The first empirical orthogonal function (EOF 1) for the surface wind  
4 pattern simulated by MYNN\_UCM\_d03 at 1400 PST during CalNex-LA. EOF 1  
5 accounts for 48.1 % of the variance in the average winds. Contours: (a) terrain height  
6 (unit: m); (b) the modelled surface CO<sub>2</sub> concentration (unit: ppm) from the 1.3-km WRF-  
7 Hestia run. The red triangles indicate the locations of the GHG measurement sites. (c)  
8 The modelled CO<sub>2</sub> concentration from the 1.3-km WRF-Vulcan run (unit: ppm). (d) The  
9 difference of the modelled CO<sub>2</sub> concentration between the 1.3-km WRF-Hestia and  
10 WRF-Vulcan runs (unit: ppm).

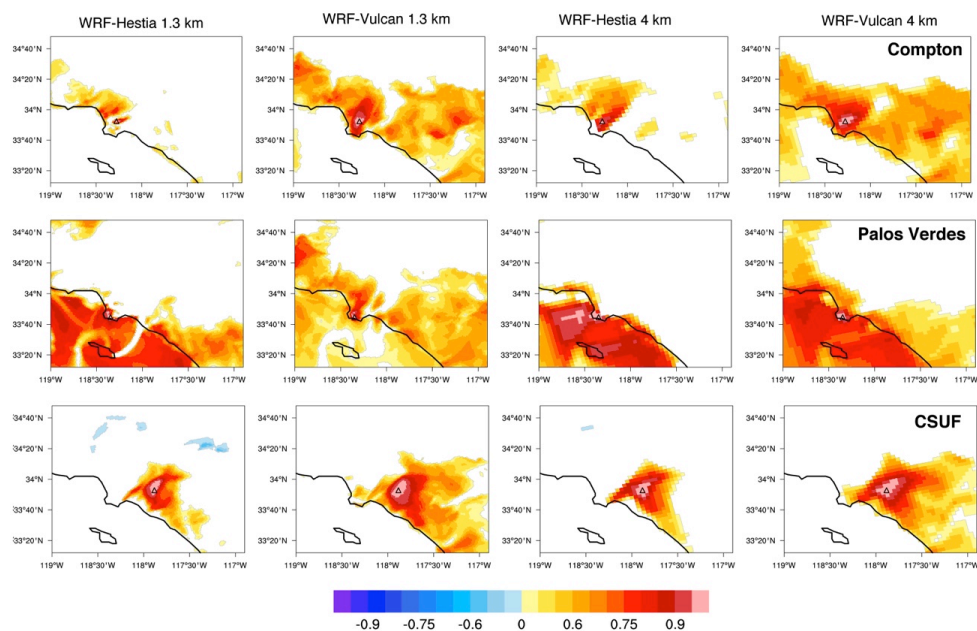




1 Figure 10. The spatial correlation map ( $R$ ) of the 1.3-km WRF-Hestia simulated  $\text{CO}_2$   
2 concentration between each site and the remainder of the domain at 1400 PST during the  
3 CalNex-LA campaign. The correlation map was constructed by calculating the  
4 simultaneous correlation of the site  $\text{CO}_2$  to the  $\text{CO}_2$  over rest of the LA megacity. Note  
5 that only those pixels that pass the  $t$ -test at the significance level of 0.01 ( $|R| \geq 0.46$ ) are  
6 coloured.



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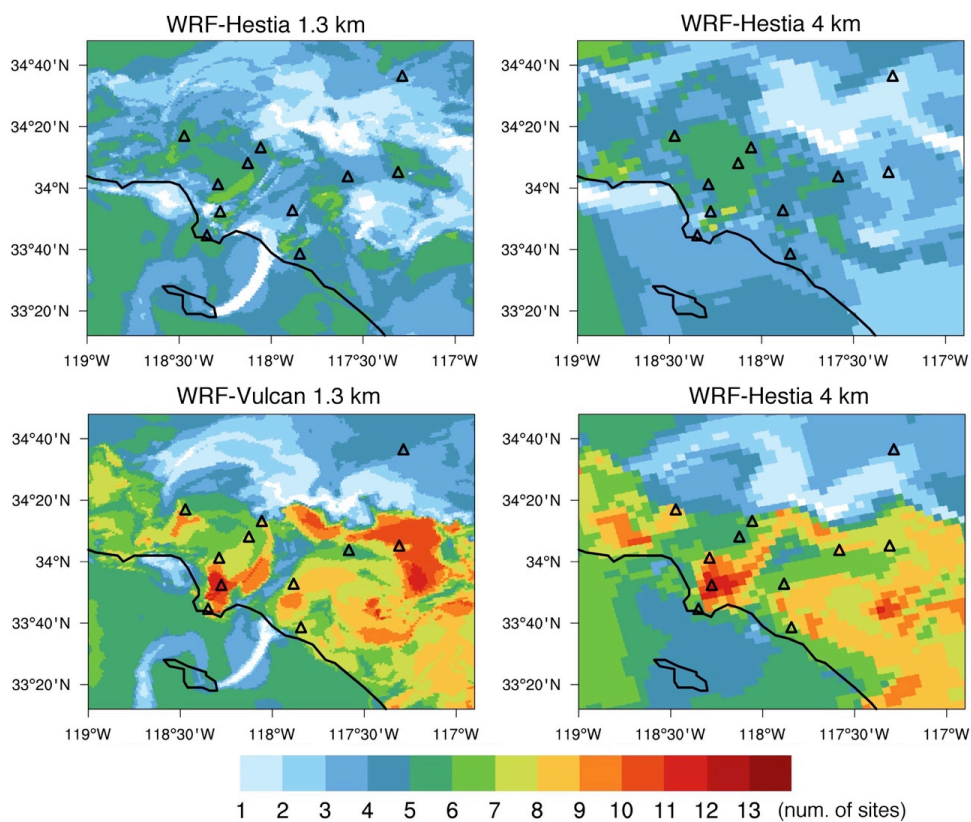


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3 Figure 11. Same as Figure 10 but for the Compton (upper row), Palos Verdes (middle  
4 row), and CSUF (lower row) sites only. Shown are the correlation maps of these three  
5 measurement sites for the 1.3-km WRF-Hestia (first column), 1.3-km WRF-Vulcan  
6 (second column), 4-km WRF-Hestia (third column), and 4-km WRF-Vulcan runs. Note  
7 that only those pixels that pass the  $t$ -test at the significance level of 0.01 ( $|R| \geq 0.46$ ) are  
8 coloured.



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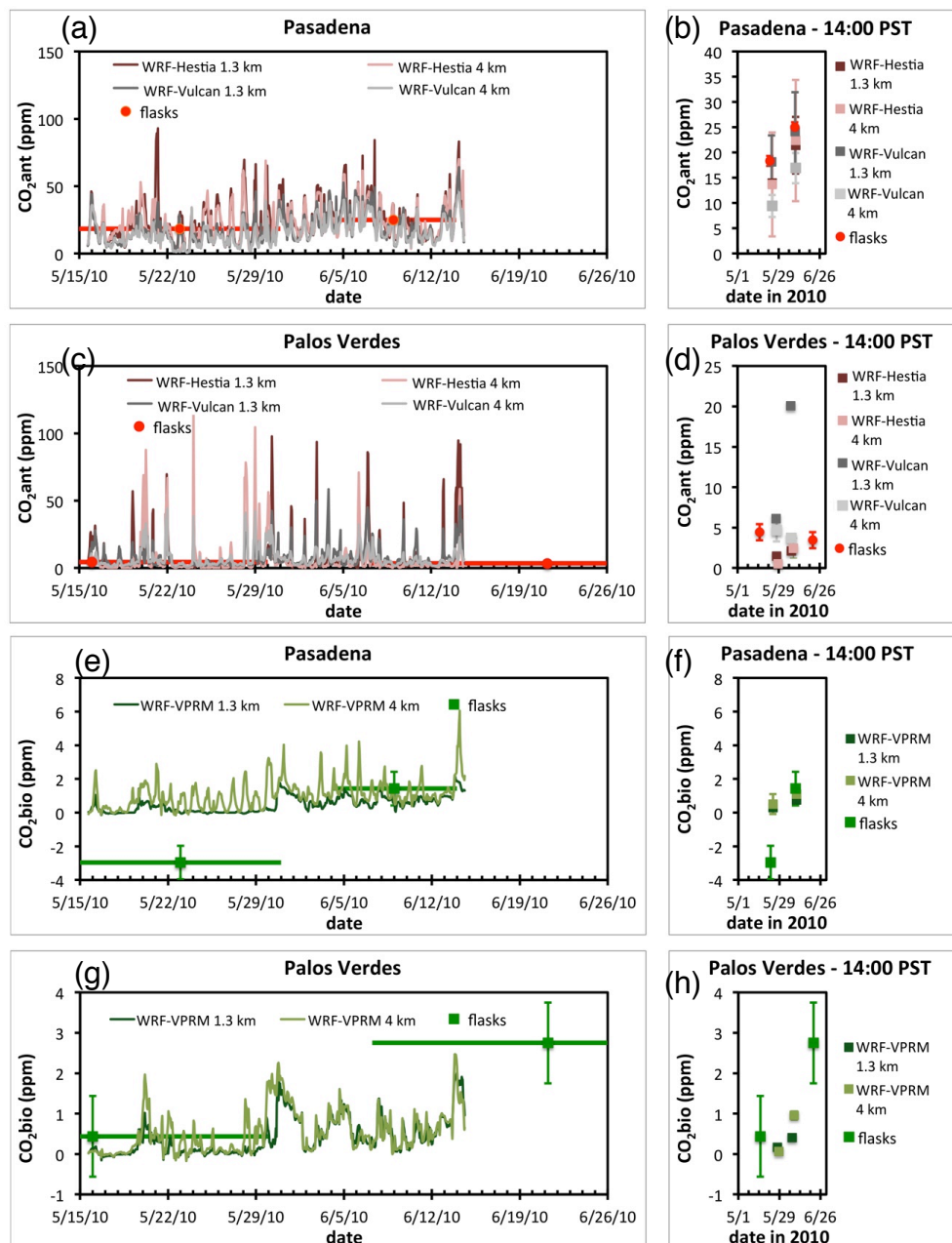
3 Figure 12. The composite maps of spatial correlation ( $R$  in Figure 10 and 11) for the 1.3-  
4 km WRF-Hestia, 1.3-km WRF-Vulcan, 4-km WRF-Hestia, and 4-km WRF-Vulcan runs.  
5 The composite map was constructed by determining the number of the observation sites  
6 for which  $|R|$  is greater than 0.46 at each grid cell.  $|R| = 0.46$  is the critical value at the  
7 significance level of 0.01 of  $t$ -test. Specifically, white cells indicate that no sites are  
8 correlated well at the location; dark red cells indicate that over 13 sites have good  
9 correlation at the location. The SCI and Dryden sites are not shown on these maps.

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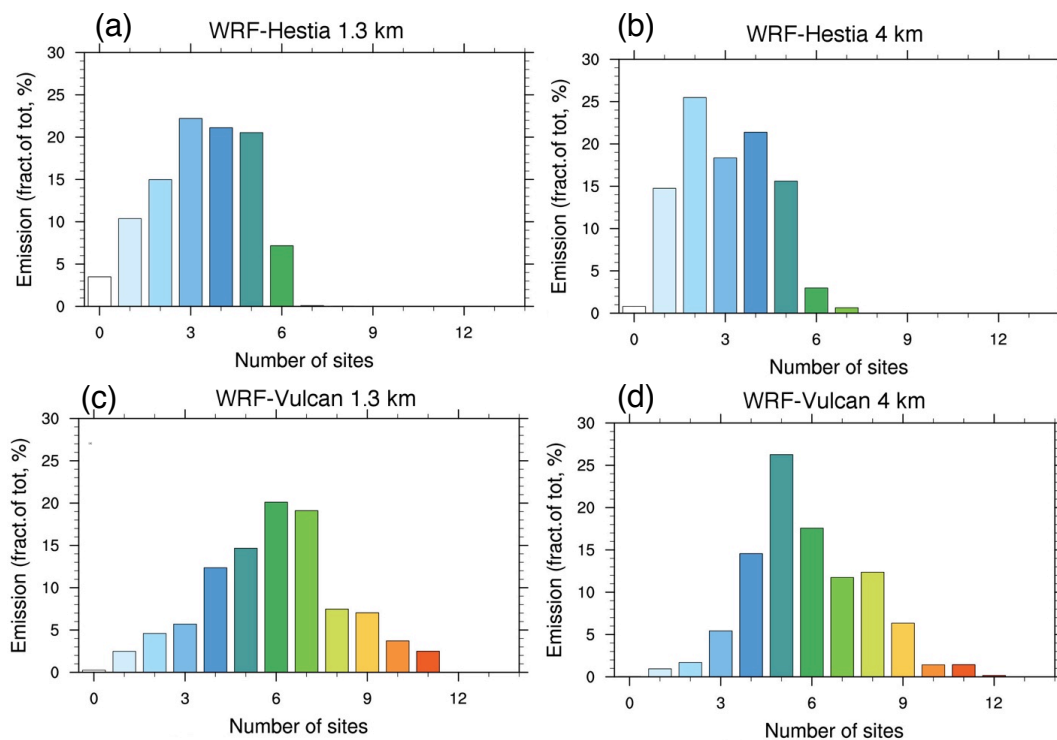
2 Figure 13. Comparisons of flask-sampled and modelled (a-d) anthropogenic fossil fuel  
3 and (e-h) biogenic CO<sub>2</sub> concentration. Left column: hourly time series. The horizontal  
4 error bars on the flask-sampled data points indicate the range of dates combined in each  
5 sample. Note that much of the time periods for the  $\Delta^{14}\text{C}$  samples at the Palos Verdes site



- 1 are before or after the modelling period. Right column: Averages at 1400 PST during
- 2 CalNex-LA. See Newman et al. (2015) for details about the sites and sampling
- 3 information.



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3 Figure 14. The fraction of the FFCO<sub>2</sub> emission over the LA megacity as function of the number  
4 of the GHG measurement sites that covers the area for (a) 1.3-km WRF-Hestia, (b) 4-km WRF-  
5 Hestia, (c) 1.3-km WRF-Vulcan, and (d) 4-km WRF-Vulcan runs during CalNex-LA.

6