1	How can mountaintop CO <sub>2</sub> observations be used to constrain regional
2	carbon fluxes?
3	John C. Lin <sup>1</sup> , Derek V. Mallia <sup>1</sup> , Dien Wu <sup>1</sup> , Britton B. Stephens <sup>2</sup>
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5	<sup>1</sup> Department of Atmospheric Sciences, University of Utah, Salt Lake City, Utah 84112, USA
6	<sup>2</sup> Earth Observing Laboratory, National Center for Atmospheric Research, Boulder, Colorado
7	80301, USA
8	Correspondence to: John C. Lin (John.Lin@utah.edu)
9	
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#### 11 Abstract.

12 Despite the need for researchers to understand terrestrial biospheric carbon fluxes 13 to account for carbon cycle feedbacks and predict future CO<sub>2</sub> concentrations, knowledge 14 of these fluxes at the regional scale remains poor. This is particularly true in 15 mountainous areas, where complex meteorology and lack of observations lead to large 16 uncertainties in carbon fluxes. Yet mountainous regions are often where significant 17 forest cover and biomass are found—i.e., areas that have the potential to serve as carbon 18 sinks. As CO<sub>2</sub> observations are carried out in mountainous areas, it is imperative that 19 they are properly interpreted to yield information about carbon fluxes. In this paper, we 20 present  $CO_2$  observations at 3 sites in the mountains of the Western U.S., along with 21 atmospheric simulations that attempt to extract information about biospheric carbon 22 fluxes from the CO<sub>2</sub> observations, with emphasis on the observed and simulated diurnal 23 cycles of CO<sub>2</sub>. We show that atmospheric models can systematically simulate the wrong 24 diurnal cycle and significantly misinterpret the CO<sub>2</sub> observations, due to erroneous 25 atmospheric flows as a result of terrain that is misrepresented in the model. This problem 26 depends on the selected vertical level in the model and are exacerbated as the spatial 27 resolution is degraded, and our results indicate that a fine grid spacing of ~4 km or less 28 may be needed to simulate a realistic diurnal cycle of  $CO_2$  for sites on top of the steep 29 mountains examined here in the American Rockies. In the absence of higher resolution 30 models, we recommend coarse-scale models to focus on assimilating afternoon  $CO_2$ 31 observations on mountaintop sites over the continent to avoid misrepresentations of

32 nocturnal transport and influence.

#### 33 1. Introduction

34 Scientific consensus among climate scientists points to carbon dioxide  $(CO_2)$  as 35 the main greenhouse gas leading to climate change (IPCC, 2014). Therefore, a strong 36 need exists to quantify and understand global carbon fluxes, among which the terrestrial 37 biospheric component is the most dynamic, potentially even reversing signs on an annual 38 basis from year to year (Le Quéré et al., 2015; Sarmiento et al., 2010). Yet quantifying 39 and predicting terrestrial biospheric carbon fluxes continue to pose a challenge to 40 researchers, as seen in the large divergence between models in projections of biospheric 41 fluxes into the future (Cox et al., 2000;Friedlingstein et al., 2003;Arora et al., 2013) as 42 well as in hindcast mode, particularly at the regional scale (Sarmiento et al., 43 2010;Stephens et al., 2007;Fisher et al., 2014).

Because mountains cover approximately a quarter of the Earth's land surface (Blyth et al., 2002), it is imperative to quantify and understand carbon fluxes over "complex terrain". Case in point is the Western U.S., where significant amounts of biomass are found above 1000 m elevation (Fig. 1). Similarly, much of the biomass and potential for terrestrial carbon storage in other parts of the world are found in hills or mountains, partly due to the fact that historical deforestation and biomass removal have been most pronounced in easier-to-access, flat regions (Ramankutty and Foley, 1999).

51 Despite the importance of regions with complex terrain in regional to global 52 carbon cycling, these areas have hitherto been under-sampled due to logistical difficulties, 53 harsh environmental settings, and violation of flat terrain assumptions in eddy covariance. 54 Recently, Rotach et al. (2014) argued that current difficulties to balance the terrestrial 55 carbon budget are due to inabilities to handle atmospheric circulations in complex terrain. 56 While these authors presented a strong case for the consideration of flows over complex 57 terrain, they did not quantify the implications of neglecting such flows for interpreting 58 CO<sub>2</sub> observations. However, the significance of complex terrain has led to efforts to start 59 closing this gap, in regions such as Europe (Pillai et al., 2011) and the American Rockies 60 ((Schimel et al., 2002); see below).

61 The American Rockies will be the focus region of this study, which attempts to 62 show how CO<sub>2</sub> concentrations in mountain regions can be properly linked, through 63 atmospheric transport, to biospheric fluxes. While the objective of this paper is to use the 64 American Rockies as a case study to illustrate general aspects of interpreting CO<sub>2</sub> 65 observations in mountainous regions, several other compelling reasons exist for studying 66 this region. Both models and observations have suggested that significant carbon storage 67 can occur in the American Rockies (Fig.1) (Schimel et al., 2002; Monson et al., 68 2002; Wharton et al., 2012), albeit this storage is highly sensitive to environmental drivers 69 such as temperature and water availability (Monson et al., 2006;Schwalm et al., 70 2012; Wharton et al., 2012; Potter et al., 2013) as well as disturbances such as insect 71 infestation (Negron and Popp, 2004) and wildfires (Wiedinmyer and Neff, 2007). These 72 disturbances are also coinciding with rapid population increases in this region (Lang et al., 73 2008), with concomitant rise in urban CO<sub>2</sub> emissions (Mitchell et al., In Review), urban-74 wildland interfaces (Mell et al., 2010), and demands for water resources (Reisner, 75 1993:Gollehon and Ouinby, 2000).

76 Recently, several research efforts have attempted to improve the understanding of 77 carbon fluxes in the American Rockies. Direct eddy covariance-based measurements of 78 carbon fluxes have been carried out in the mountains (Blanken et al., 2009; Yi et al., 79 2008); however, the eddy covariance technique characterizes fluxes only over a small area of  $\sim 1 \text{ km}^2$  (Baldocchi et al., 2001) and requires careful attention to potential biases 80 81 from local advection. Ground-based ecological measurements (Anderegg et al., 82 2012; Tkacz et al., 2008) yield detailed information regarding the ecosystem, but such 83 observations are also limited in spatial coverage and temporal resolution. Atmospheric 84 CO<sub>2</sub> observations can characterize fluxes over hundreds of km (Gerbig et al., 2009), 85 providing important regional scale constraints. Aircraft-based CO<sub>2</sub> measurements in this 86 region have had some success in characterizing regional scale fluxes (Desai et al., 2011), 87 albeit on a sporadic, campaign-based setting. More significantly, a network of accurate 88 CO<sub>2</sub> observations has been maintained on mountaintops in the Rockies for the past 89 decade (Stephens et al., 2011). These observations have been assimilated by

sophisticated global carbon data assimilation systems such as "CarbonTracker" (Peters et
al., 2007) to retrieve biospheric carbon fluxes over the mountainous regions and the rest
of the globe.

93 Due to the expanding number of  $CO_2$  observations in mountainous areas and the 94 need to understand carbon fluxes in such regions, a strong motivation exists to evaluate 95 existing methods in which CO<sub>2</sub> observations are used in atmospheric models to retrieve 96 carbon fluxes. van der Molen et al. (2007) simulated CO<sub>2</sub> variability near a Siberian 97 observational site and showed that even modest terrain variations of ~500 m over 200 km 98 could lead to considerable CO<sub>2</sub> gradients. Due to the dangers of mis-representing 99 terrain/flows and introducing biases into the carbon inversion system, mountaintop CO<sub>2</sub> 100 observations have often been omitted from carbon inversion systems (Rodenbeck, 101 2005;Geels et al., 2007;Peters et al., 2010). In fact, the most recent release of 102 CarbonTracker ("CT-2015") stopped assimilating the three RACCOON sites 103 (http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/).

However, the absence of mountaintop  $CO_2$  observations to constrain carbon inversion systems is, in effect, throwing away valuable information that could inform carbon exchange in potentially important areas of the world (Fig. 1). Case in point is the Schauinsland  $CO_2$  time series on a mountain in the middle of Western Europe, which as of this writing has collected over 40 years of continuous  $CO_2$  data (Schmidt et al., 2003) but remains excluded from numerous carbon inversion systems (Rodenbeck, 2005;Geels et al., 2007;Peters et al., 2010).

111 As a means to evaluate models' ability to interpret mountaintop CO<sub>2</sub> observations, 112 we specifically adopt the observed diurnal cycle during the summer growing season as a 113 key diagnostic. This is because the diurnal cycle during the growing season, with 114 nighttime respiratory release and daytime photosynthetic drawdown of  $CO_2$ , is a 115 prominent feature in the coupling between biospheric fluxes and the atmosphere and one 116 of the dominant modes in the CO<sub>2</sub> time series (Bakwin et al., 1998;Denning et al., 1996). Furthermore, models tend to either use CO<sub>2</sub> data from the nighttime (Keeling et al., 1976) 117 118 (to sample subsiding air in the mid-troposphere) or from the daytime (during well-mixed 119 conditions), and aspects of the diurnal cycle can provide clues as to whether the model is 120 capturing the link between fluxes and concentrations right at either, both, or neither of 121 these times.

122 The diurnal pattern of  $CO_2$  observed at the Storm Peak Laboratory, Colorado, was 123 examined by one of the first mesoscale modeling studies that investigated the impact of 124 mountain flows on CO<sub>2</sub> concentrations (De Wekker et al., 2009). Although this study 125 adopted an idealized simulation covering only a single day of observations, it nonetheless 126 underscored the role of daytime upslope winds. A common approach is to assimilate 127 mountain observations at night (Peters et al., 2007), favoring subsidence conditions 128 characterizing free tropospheric concentrations and avoiding the need to resolve daytime 129 upslope flows (Keeling et al., 1976).

Recently, Brooks et al. (2016) used pseudo-observations to examine the
detectability of a regional flux anomaly by three mountaintop CO<sub>2</sub> sites in the American
Rockies (including Storm Peak Laboratory). For the atmospheric model they adopted a
time-reversed Lagrangian particle dispersion model (LPDM), which yields the
"footprint", or source region, of the observation sites (Lin et al., 2012). Although this

135 study investigated whether the three mountaintop sites could detect signals of ecosystem

136 disturbance, Brooks et al. (2016) did not specifically examine issues related to erroneous

137 atmospheric transport in complex terrain nor compare modeled CO<sub>2</sub> against observed138 values.

139 In this paper, we will focus on the same 3 mountaintop  $CO_2$  sites in the American 140 Rockies and specifically examine the implications of using nocturnal versus daytime data 141 within models, in light of atmospheric models at various grid spacings—from high 142 resolution regional simulations to coarser global scale simulations. More specifically, we

- resolution regional simulations to coarser global scale simulations. More specifically, we will drive a time-reversed LPDM with various meteorological fields and receptor heights.
- 144 We will probe the implications on the footprint, transport, and the resulting CO<sub>2</sub>
- 145 concentrations as the driving meteorological fields are degraded with coarser grid spacing 146 and also as different vertical levels within the model are used.
- 147 The guiding questions of this paper are, as follows:
- 148 1. How do atmospheric flows in mountainous areas affect CO<sub>2</sub> concentrations and their representation in models?
- 150 2. What are the errors incurred due to the use of coarse-scale atmospheric simulations?
- 151 3. How can mountaintop  $CO_2$  observations be used in an effective manner to constrain
- 152 regional carbon fluxes in complex terrain?

#### 153 **2. Methodology**

#### 154 2.1 RACCOON Observations

155 The Regional Atmospheric Continuous CO<sub>2</sub> Network (RACCOON,

156 http://raccoon.ucar.edu) was established in 2005 and has collected in situ CO<sub>2</sub>

157 measurements at up to six sites over the past decade (Stephens et al., 2011). Here we

158 present and simulate observations from the three longest running high-alpine sites (Fig. 2;

- 159 Table 1). The easternmost site (NWR) is at 3,523 m elevation near the treeline on Niwot
- Ridge, just west of Ward, CO. Niwot Ridge is a LTER site and there is an AmeriFlux
  tower run by the University of Colorado 3 miles east and 500 m lower on the ridge. The

instrumentation reside in the "T-Van" where the U.S. National Oceanic and Atmospheric

- 163 Administration (NOAA)'s Global Monitoring Division has collected weekly flask
- 164 samples for measurement of CO<sub>2</sub>, isotopes, and other species for over 40 years, and daily
- 165 flasks since 2006. The middle site (SPL) is at the Desert Research Institute's Storm Peak
- 166 Lab (3,210 m on Mt. Werner near Steamboat Springs, CO). This mountaintop
- 167 observatory has a long history of measurements related to cloud physics, cloud-aerosol
- 168 chemistry, and air quality. The westernmost site (HDP) is on Hidden Peak (3,351 m,
- above the Snowbird ski resort, Utah). This mountaintop site generally experiences
- regionally well-mixed or free-tropospheric air, but with influences from Salt Lake Cityduring boundary-layer growth and venting periods.
- The RACCOON measurements are based on a LiCor LI-820 single-cell IRGA
  with frequent calibrations. The instruments sample air from one of three inlet lines on a
  tower (two at HDP) and use a suite of four calibration gases plus a fifth surveillance gas.
  All reference gases are rigorously tied to the WMO CO<sub>2</sub> Calibration Scale with use of the
  NCAR CO<sub>2</sub> and O<sub>2</sub> Calibration Facility. 100-second average measurement precision is ±
- 177 0.1 ppm (1  $\sigma$ ), and intercomparability is estimated from several methods to be 0.2 ppm
- 178 (Stephens et al., 2011).

- We applied filtering to the mountaintop CO<sub>2</sub> observations to remove local
  influences and to extract values that are more regionally representative (Brooks et al.,
- 181 2012). Observations were filtered out in which the within-hour standard deviation is
- 182 greater than 1.0 ppm or when the differences between the top two inlets are greater than
- 183 0.5 ppm, which indicate periods when significant influences that are highly localized to
- 184 the site are affecting the observations. This filtering removed 15%, 16%, and 27% of the
- 185 hourly observations at HDP, SPL, and NWR, respectively. Regardless, filtering made
- 186 negligible differences in the observed diurnal cycles in  $CO_2$  (see Supplement).
- 187 Henceforth, we will refer to the filtered observations when discussing the observed CO<sub>2</sub>.

#### 188 2.2 WRF-STILT Atmospheric Model

189 The atmospheric modeling framework adopted in this study is a Lagrangian time-190 reversed particle dispersion model, the Stochastic Time-Inverted Lagrangian Transport 191 (STILT) model (Lin et al., 2003), driven by a mesoscale gridded model, the Weather 192 Research and Forecasting (WRF) model (Skamarock and Klemp, 2008). STILT is a 193 Lagrangian model that simulates the effects of turbulent dispersion using the stochastic 194 motions of air parcels. It has been widely applied to the interpretation of CO<sub>2</sub> and trace 195 gases in general (Lin et al., 2004;Hurst et al., 2006;Göckede et al., 2010;Kim et al., 196 2013; Mallia et al., 2015; Jeong et al., 2012). WRF is a state-of-the-art non-hydrostatic 197 mesoscale atmospheric model that can simulate a variety of meteorological phenomena 198 (Skamarock and Klemp, 2008), gaining widespread acceptance and usage among the 199 atmospheric science community. Careful coupling between WRF and STILT has been 200 carried out, with an emphasis towards physical consistency and mass conservation 201 (Nehrkorn et al., 2010).

202 For this study, we ran WRF in a two-way nested mode centered between Utah and 203 Colorado where the RACCOON sites are located (Fig. 2). The grid spacing was refined 204 in factors of 3, from 12 km grid spacing covering the entire Western U.S. to 4 km and 205 then to 1.3 km in the innermost domain that covers all of the RACCOON sites. 41 206 vertical levels were adopted, with 10 of these levels within 1 km of the ground surface, 207 following Mallia et al. (2015). Comprehensive testing of different WRF settings have 208 been carried out as part of a previous publication (Mallia et al., 2015) and adopted here: 209 i.e., the MYJ, Grell-Devenyi Ensemble, and Purdue Lin schemes for parameterizing the 210 planetary boundary layer (PBL), cumulus convection, and microphysics, respectively. In 211 addition to the testing reported in Mallia et al. (2015), we have also carried out evaluation 212 of the WRF fields specifically using meteorological measurements on mountaintops, near 213 the RACCOON sites. These evaluations reveal that errors in the simulated 214 meteorological fields are reasonable when compared against other atmospheric 215 simulations evaluated in less complex terrain (Mallia et al., 2015), and biases are 216 especially small for the WRF-1.3km fields (Table 2). In this paper, we will examine the 217 resulting differences in meteorological and CO<sub>2</sub> simulations when STILT is driven by 218 WRF fields at three different grid spacings.

In addition to the three WRF domains, we drove STILT with a fourth
 meteorological field, from NCEP's Global Data Assimilation System (GDAS). GDAS is
 archived at 1°×1° grid spacing, at 6 hourly intervals and at 23 vertical pressure levels.
 Driving STILT with GDAS was a means by which we attempted to construct an

223 atmospheric model to resemble the NOAA CarbonTracker product, which was also at

 $1^{\circ} \times 1^{\circ}$  resolution (and 25 vertical levels) over North America. More details about CarbonTracker can be found in the next section.

226 Driven by the various meteorological fields, STILT released 2000 air parcels 227 every 3 hours (00, 03, 06, ...21 UTC) for the months of June, July, and August 2012 from the RACCOON sites and transported for 3 days backward in time. An example of 228 229 STILT-simulated air parcel trajectories can be found in Fig. S1. The choice of 2000 230 parcels followed from results from sensitivity tests in a previous study, also over the 231 Western U.S. (Mallia et al., 2015). In the case of WRF, STILT has the capability to 232 transport the parcels in a nested fashion. So when we refer to "WRF 1.3km simulations", 233 it actually means that the atmosphere in the innermost domain (Fig. 2) was simulated at 234 1.3 km, switching to 4km grid spacing when the parcel left the 1.3km domain; likewise, 235 the 12km winds were used when the parcel left the 4km domain. For the "WRF 4km 236 simulations" we started with the 4 km fields as the innermost domain, and then 12 km in 237 the outer domain.

238 For each site, we released STILT parcels using two different ways to determine 239 starting levels. When we refer to "AGL", we mean that the starting height was set at the 240 level of the inlet above the ground surface (Table 1), following the local terrain as 241 resolved in the meteorological model (whether at 1.3-, 4-, 12-km, or 1° grid spacing). 242 The alternative method, referred to as "ASL", means that the starting level was set to the 243 elevation above sea level. For instance, the HDP site is located at 3351 m above sea level. 244 The ground height as resolved by the 12km WRF model is at 2357 m, so the starting 245 height was placed at 994 (=3351 - 2357) m above the resolved terrain. CarbonTracker, 246 as well as many other global-scale models (Geels et al., 2007; Peters et al., 2010) places 247 the observation site at an internal model level following the ASL method, so the "GDAS-248 ASL" runs were a means by which we attempted to mimic the global model configuration 249 and to illuminate potential errors that could result from such a configuration. We also 250 tested the AGL height for GDAS, at HDP only. As shown later, these runs were highly 251 erroneous, so we did not carry them out for the other two sites.

252 The STILT-simulated air parcels were tracked as they were transported 253 backwards in time from the RACCOON receptors (see example in Fig. S1); when they 254 were in the lower part of the PBL, the locations of the parcels and amount of time the 255 parcels spend in the lower PBL were tallied. This information was used in calculation of 256 the "footprint"-i.e., the sensitivity of the receptor to upwind source regions (in units of 257 concentration per unit flux). For more details, see Lin et al. (2003). The footprints, 258 encapsulating the atmospheric transport information, were then combined with gridded 259 fluxes from the biosphere and anthropogenic emissions, which are described in the next 260 sections.

#### 261 2.3 CarbonTracker CO<sub>2</sub> Concentrations and Biospheric Fluxes

CarbonTracker is a carbon data assimilation system covering the whole globe that
retrieves both oceanic and terrestrial biospheric carbon fluxes (Peters et al., 2007).
Observed atmospheric CO<sub>2</sub> concentrations are assimilated by CarbonTracker, which
adjusts carbon fluxes to minimize differences with the observed CO<sub>2</sub> using an ensemble
Kalman filter methodology.

We took three-dimensional CO<sub>2</sub> fields from CarbonTracker to initialize CO<sub>2</sub> concentrations at the end of the 3-day back trajectories from STILT. CarbonTrackerderived biospheric fluxes, along with anthropogenic and fire emissions (Sect. 2.4), were

- also multiplied with STILT-derived footprints and combined with the initial CO<sub>2</sub>
   concentrations to yield simulated CO<sub>2</sub> at the RACCOON receptors.
- CarbonTracker is maintained and continues to be developed by the NOAA's Earth System Research Laboratory. For this paper, we adopt the "CT-2013b" version. CT-2013b provides multiple prior estimates of the oceanic, terrestrial, and fossil carbon fluxes, with each combination yielding separate posterior fields of carbon fluxes and CO<sub>2</sub> distributions. CT-2013b results are presented as an average across the suite of prior fluxes and CO<sub>2</sub> fields.

278 CT-2013b resolves atmospheric transport and fluxes at  $1^{\circ} \times 1^{\circ}$  over North America and  $3^{\circ}$ -lon  $\times 2^{\circ}$ -lat in the rest of the globe, with 25 vertical levels. The driving 279 280 meteorological fields come from the European Centre for Medium-Range Weather 281 Forecasts's ERA-interim reanalysis. The ensemble Kalman filter system within 282 CarbonTracker solves for scaling factors on weekly timescales to adjust upward or 283 downward biospheric carbon fluxes. The adjustments were made over "ecoregions" on 284 land, rather than attempting to adjust fluxes within individual gridcells, as way to reduce 285 the dimensions of the inversion problem within CarbonTracker. More details regarding 286 the CarbonTracker system can be found in Peters et al. (2007) and on-line at 287 http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2013B/.

288 Since CarbonTracker was designed for global carbon cycle analyses to retrieve 289 large-scale fluxes, the adjustment to biospheric carbon fluxes could result in artifacts at 290 the local to regional scales. More specifically, the attempt to match CO<sub>2</sub> observations 291 with a single scalar can result in flipped diurnal cycles, causing carbon uptake during the 292 night that is partly offset by enhanced respiration in a nearby ecoregion (Fig. S2). For this 293 paper, we implemented a fix that removed this artifact by detecting these reversed diurnal 294 patterns, adjusting them while preserving the 24-hour integrated carbon flux. See the 295 Supplement and Fig. S3 for details.

#### 296 2.4 Anthropogenic and Fire Emissions

297 Anthropogenic CO<sub>2</sub> emissions were obtained from the Emission Database for 298 Global Atmospheric Research (EDGAR) (European Commission, 2009), which resolves 299 emissions globally at 0.1°×0.1° annually. In order to temporally downscale the annual 300 emissions, hourly scaling factors were obtained from the Vulcan emission inventory 301 (Gurney et al., 2009) and applied to the EDGAR annual emissions. Lastly, CO<sub>2</sub> emissions 302 from EDGAR for Year 2010 were extrapolated to 2012 using population growth rates 303 across the U.S. since 2010, as this was the last year in which EDGAR emissions were 304 available.

Wildfire emissions for CO<sub>2</sub> were obtained from the Wildland Fire Emissions
Inventory (WFEI) (Urbanski et al., 2011). Since these emissions were only reported daily,
three-hourly diurnal scaling factors were obtained from Global Fire Emissions Database
v3.1 and applied to the daily WFEI emissions to downscale the emissions to sub-daily
timescales (Mu et al., 2011;van der Werf et al., 2010).

310 Contributions from anthropogenic and wildfire emissions, on average, to the mean 311  $CO_2$  diurnal cycle observed at all the mountain sites were secondary in comparison to the 312 biosphere (Fig. S4). In particular, the wildfire contributions were episodic and averaged 313 out to negligible contributions over Jun~Aug 2012 (Fig. S4). Because of this, we will not 314 touch upon wildfires in the remainder of the paper.

#### 315 **3. Results**

#### 316 **3.1 Observed versus Simulated Diurnal Cycle**

The observed and simulated diurnal cycles of CO<sub>2</sub> for the three selected RACCOON sites are shown in Fig. 3. These diurnal patterns were calculated from averaging the 3-hourly simulated time series from different model setups, which exhibit significant variability at multi-day synoptic timescales and correlations with different meteorological variables between HDP, SPL, and NWR (Fig. S5, Table S1). Due to this complexity we are focussing the analysis on the average diurnal pattern.

The observed diurnal cycle exhibits an amplitude of  $\sim 2$  ppm, on average, with more elevated concentrations at night and depleted values during the day. In contrast to the observed diurnal cycles, the simulated CO<sub>2</sub> extracted from the site's altitude within CarbonTracker's output (Table 1) exhibits a different cycle. Instead of peaking at night, CO<sub>2</sub> in CarbonTracker reaches its maximum during the afternoon at HDP. At SPL and NWR, the diurnal cycle is significantly attenuated, with nighttime values barely elevated over the background instead of the nighttime enhancement in the observed values.

It appears that the erroneous diurnal pattern at HDP within CarbonTracker can partly be due to the diurnal reversal in the original biospheric fluxes, which showed strong uptake of CO<sub>2</sub> even at night for the gridcell where HDP is located (Fig. S2). This resulted in erroneous diurnal patterns at all of the lowest 8 levels of CarbonTracker (Fig. S6), with the bottom 2 levels exhibiting strong depletions in CO<sub>2</sub> at night and enhancements during the day, pointing to unrealistic nighttime uptake and daytime release.

However, the diurnal reversal in biospheric fluxes alone does not completely
explain the erroneous diurnal pattern. Differences in the diurnal pattern between GDASASL simulations after introducing the diurnal fix in biospheric fluxes were not as
pronounced at SPL and NWR.

The GDAS-ASL simulations show a pronounced peak of  $CO_2$  in the morning that is missing from observations at all three sites (Fig. 3). We will discuss this feature, also seen in other coarse-scale simulations of mountaintop  $CO_2$  (Geels et al., 2007), in Sect. 3.2 below.

345 In contrast to GDAS-ASL and CarbonTracker, the WRF-driven simulations better 346 reproduce the shape of the observed diurnal cycle (Fig. 3), with nighttime enhancements 347 and daytime depletions of  $CO_2$ . Considerable differences in nocturnal  $CO_2$ 348 concentrations are found, however, in the WRF-STILT runs at various grid spacings. 349 WRF-12km significantly overestimates CO<sub>2</sub> at night, while WRF-1.3km and -4km 350 produced similar CO<sub>2</sub> concentrations that correspond much more closely to observed 351 values. While GDAS simulations started near the ground ("GDAS-AGL") also exhibit nighttime enhancements and daytime depletions of CO<sub>2</sub>, the nighttime values are grossly 352 353 estimated, exceeding even the values in WRF-12km. Therefore, we do not present 354 GDAS using the AGL configurations at the other two sites.

Part of the error in all the simulations against the observations could arise from errors in the CarbonTracker boundary condition imposed at the end of the STILT back trajectories. Evaluations of CT-2013b against aircraft vertical profiles (which were not assimilated into CarbonTracker) at the Trinidad Head and Estevan Point sites on the West Coast of the North American continent carried out by the CarbonTracker team (http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2013B/profiles.php) indicate that

- 361 CT-2013b overestimates  $CO_2$  concentrations by at most 1.0 ppm, on average, during the 362 summer season. Thus, the fact that GDAS and CarbonTracker underestimate  $CO_2$  at
- 363 night likely cannot be attributed solely to a biased boundary condition.

#### 364 **3.2 Differences in Simulated Transport to Mountaintop Sites**

#### 365 3.2.1 Footprint Patterns

In order to isolate the impact of differences in atmospheric transport on the simulated
CO<sub>2</sub>, we examine the average diurnal pattern of the footprint strength over Jun~Aug
2012 (Fig. 4). At each hour of the day we summed the spatially explicit map of the
average footprint that marks out the source region of each RACCOON site—shown in
Figs. 5~6 for HDP and in the Supplemental Information for the other 2 sites. The result
shows the diurnal pattern of the sensitivity of the receptor concentration to upwind fluxes.

372 To a large extent, the diurnal variation in footprint strength mirrors the simulated 373 CO<sub>2</sub> concentrations. Nocturnal enhancements in the footprints are seen in the WRF-374 driven simulations, with the WRF-12km exhibiting the strongest nocturnal footprints. 375 Footprints from WRF-1.3km and WRF-4km are weaker at night than from WRF-12km and closely resemble each other. GDAS-AGL footprints (only shown at HDP) are the 376 377 highest among all models at night, leading to the drastic overestimation in CO<sub>2</sub> in Fig. 3. 378 In contrast, GDAS-ASL footprints exhibit a peak in the morning and are generally 379 smaller in value than their WRF counterparts at other times of the day at HDP and NWR. 380 At SPL, the GDAS-ASL footprint strengths are stronger and more in line with values 381 from the other models.

Footprints are weaker during the daytime, and in contrast to the nighttime,
 differences between footprint strengths simulated by different models are significantly
 smaller. In particular, the differences are minimized in the afternoon.

385 These patterns are also seen in the footprint maps. We further examine 386 differences in the spatial patterns of average footprints produced from the various WRF 387 and GDAS configurations. The spatial patterns are contrasted at two different times of 388 the day, associated with the nighttime and afternoon hours: 0200MST (0900UTC) and 389 1400 MST (2100UTC), respectively. Only HDP is shown for these two hours of the day 390 in Figs. 5 and 6; similar figures for SPL and NWR can be found in the Supplementary 391 Information (Figs. S7~S10). The footprint maps show marked differences at night (Fig. 392 5): the WRF-12km footprints are clearly stronger than their counterparts from the other 3 393 model configurations, with higher values covering the Wasatch Range near the HDP site. 394 Meanwhile, the GDAS-ASL footprint at 0200 MST shows a striking contrast, with very 395 low values around HDP and the Wasatch area in general.

The afternoon footprints at 1400MST (Fig. 6) display much more similarity with each other. Not only do the spatial patterns between the WRF and GDAS runs resemble one another; the significant differences in footprint strengths, with overestimation by WRF-12km and underestimation by GDAS-ASL, are no longer found. The aforementioned nighttime divergence and afternoon correspondence between footprint patterns are repeated at the SPL and NWR sites (Figs. S7~S10).

To further understand the nighttime divergence between model configurations, we now examine the average air parcel trajectories within Figs 5 and 6. It is worth noting that these trajectories differ from conventional mean wind trajectories that do not incorporate effects from turbulent dispersion (Lin, 2012). Instead, these mean trajectories 406 are determined by averaging the 2000 stochastic air parcel trajectories from STILT used 407 for simulating transport arriving at a specific hour at a particular site, and then averaging 408 over the ~90 days spanning June~August 2012. Thus there are ~180,000 stochastic 409 trajectories averaged into generating the mean trajectory, thereby incorporating the net 410 effect of turbulence on atmospheric transport. An example showing a subset of stochastic 411 air parcels giving rise to the average trajectory is given in Fig. S1 for NWR, for 1400 412 MST

413 Similar to the footprints, average trajectories differ much more at night than in the 414 afternoon. Differences in average air parcel trajectories and the underlying resolved mountainous terrain are further examined in the next section. 415

#### 416 3.2.2 Three-dimensional Terrain and Trajectories

417 The 3D terrain plots in Figs. 7, 9, and 11 illustrate the degradation in terrain 418 resolved by coarser grid spacings and the resulting differences in average STILT-derived 419 stochastic air parcel trajectories started at night (0900UTC) from the three sites. The 420 afternoon (2100UTC) plots are shown in the Supplementary Information (Figs. S11~S13). 421 The PBL heights, which determine whether air parcels are affected by surface fluxes (and 422 lead to nonzero footprint values) are also plotted as blue lines in the same plots. Note 423 that the apparent intersection of the PBL height with the ground in Figs. 7 and 9 is an 424 artifact from averaging of multiple PBL heights along stochastic trajectories (Fig. S1).

425 Despite terrain smoothing compared against WRF-1.3km, WRF-4km produced 426 STILT trajectories that are very similar to those from WRF-1.3km at all three sites, 427 suggesting that salient features of the mountain flows resolved with 1.3km spacing are 428 also found in the 4km spacing. In contrast, WRF-12km and GDAS-ASL both differed 429 significantly from the more finely-gridded WRF simulations. Not only did the 430 trajectories deviate from the higher resolution counterparts; the relationships between the 431 trajectory vis-à-vis the PBL height, critical for determining footprints and simulating CO<sub>2</sub> 432 changes (Sect. 2.2), also differ. The WRF-12km trajectories spend more time within the 433 PBL, while GDAS-ASL trajectories are found much less within the PBL, because they 434 start at a greater height above ground level.

435 An alternative perspective is to view the trajectory and PBL heights relative to the 436 ground surface ("AGL") instead of above sea level, at each time step backward in time 437 from the receptor (Figs. 8, 10, 12). These figures highlight the fact that while PBL 438 dynamics in the three WRF configurations are similar, the heights of the trajectories 439 relative to the PBL height differ. The trajectory exits above the nocturnal PBL one hour 440 backward in time, on average, while the WRF-12km trajectory spends several hours 441 within the PBL.

442 The difference in the trajectory behavior can be explained by the differing terrain. 443 In mountainous terrain, PBL heights generally follow the terrain elevations, albeit with attenuated amplitude (Steyn et al., 2013). Thus in WRF-1.3km and 4km, the more highly 444 445 resolved terrain produced shallow nocturnal PBL height that descend in the valley (Fig. 7) 446 while the corresponding trajectory hovers above it. Viewed relative to the ground surface 447 (Fig. 8), the trajectory originating from HDP appears to have exited above the nocturnal 448 PBL in WRF-1.3km and 4km. In contrast, due to the significantly "flattened" mountains 449 in WRF-12km and in GDAS, the PBL heights exhibit less spatial variation near the 450 mountaintop receptor, since the terrain itself was smoothed. Consequently, WRF-12km 451 trajectories, unlike the WRF-1.3km or -4km cases, travel closer to the ground surface,

452 within the nighttime PBL, even as it is advected away from the three RACCOON sites 453 (Figs. 7, 8). This resulted in stronger nighttime footprints in WRF-12km as seen in Figs. 454 4 and 5. Another effect of the proximity of the air parcels to the model's ground surface 455 is the slower windspeeds from surface drag, causing the air parcel trajectories to remain close to the 3 sites until the previous day; for HDP and SPL, the mean trajectories spiral 456 457 toward the site at the surface, following an "Ekman wind spiral" pattern (Holton, 1992). 458 In WRF-1.3km and WRF-4km, the measurement sites are at significantly higher 459 elevations above the resolved valleys in the area surrounding the sites, and the air parcels 460 are found above the shallow nocturnal boundary layer hugging the valley floor, on 461 average (Fig. 7).

462 Although both WRF-12km and GDAS poorly resolve the mountains, a key 463 difference in the case of GDAS-ASL is that the air parcels were released at a site's 464 elevation above sea level (following what is generally done in CarbonTracker, and other 465 global models), much higher above ground than the release used in WRF-12km, which 466 was selected to be the height in AGL above the flattened mountain. Therefore, the 467 GDAS-ASL trajectories were significantly higher than the PBL height in the model (particularly at HDP and NWR), which followed the flattened ground surface in the  $1^{\circ} \times 1^{\circ}$ 468 469 grid spacing. Another noticeable difference in GDAS-ASL trajectory was the 470 significantly higher daytime PBL heights (Figs. 8, 10, 12). We suspect this is because of 471 the greatly reduced vertical resolution within GDAS (23 levels versus 41 levels in WRF): 472 since STILT diagnoses the PBL height to correspond to a model level, a higher PBL 473 height was chosen for GDAS because of the thicker vertical level. Another subtle artifact 474 of the coarse resolution within GDAS can be seen in the anomalously low daytime PBL 475 height just in the vicinity of HDP (Figs. 13, S11). It appears that the GDAS model set an 476 entire  $1^{\circ} \times 1^{\circ}$  grid box near HDP to be water body (the Great Salt Lake), thereby 477 suppressing the PBL height.

The three-dimensional plots can explain the higher nighttime footprint strengths at SPL (Figs. 4, S7). This result appears to be a consequence of the relative elevation of the site and surrounding terrain. The elevation of the surrounding valley floor at SPL is closer to that of the mountaintop location of SPL (Fig. 9); therefore, air parcels released from SPL would have a stronger tendency to reside within the PBL even over the surrounding valleys, unlike the steeper dropoff--i.e., deeper valley--upwind of HDP (Fig. 7) and NWR (Fig. 11).

485 As already found in the footprints (Fig. 5), the afternoon (2100 UTC) differences 486 in air parcel trajectories are much smaller (Figs. S11~S13). We suspect that this is due to 487 the fact that the deeper daytime PBL height causes the trajectories to reside within the 488 PBL, and stronger mixing within the daytime PBL minimize the relative terrain 489 differences. A previous modeling study focusing on the SPL area has also suggested the 490 daytime afternoon PBL depth to extend above the mountaintop (De Wekker et al., 2009), 491 indicating that differences between terrain resolution and the resulting flows could be 492 reduced due to the strong mixing taking place within the deep afternoon PBL. 493 Consequently, simulations in the afternoon show much smaller divergence between 494 various model configurations, resulting in similar footprint strengths and CO<sub>2</sub> values 495 (Figs. 3 and 4). More evidence of the convergence in afternoon simulated  $CO_2$  can be 496 found in the small differences in CO<sub>2</sub> modeled at CarbonTracker's different levels during 497 this time (Fig. S6).

498 A few studies have specifically focused on the flows and atmospheric transport 499 around the NWR site. These authors have pointed to thermally driven flows, particularly 500 downslope drainage flow events at night (Sun et al., 2007;Sun and De Wekker, 501 2011;Blanken et al., 2009). Daytime upslope events, while weaker, were also noted (Sun 502 and De Wekker, 2011;Blanken et al., 2009;Parrish et al., 1990). It may seem that the 3D trajectories in Fig. 11 and Fig. S13 run counter to the presence of such thermally driven 503 504 flows. We suspect that this is because the thermally driven flows induced by the terrain 505 cannot be discerned in the mean trajectories, which also reflect the larger scale flows that 506 can be stronger than the local scale thermally driven flows (Zardi and Whiteman, 2013). 507 When one examines the stochastic trajectories from which the mean trajectories are based 508 (Fig. S1), it is clear that some upslope trajectories can be detected.

509 We now examine the reason for the erroneous daytime peak in simulated  $CO_2$ 510 from GDAS-ASL that does not show up in the observations (Fig. 3). We specifically 511 focus on this feature because the daytime peak was also found in other coarse-scale 512 simulations of CO<sub>2</sub> for mountaintop sites--e.g., in Europe (Geels et al., 2007). Focusing 513 on the three-dimensional plots at the hours of 0800 and 1100 MST (Fig. 13), when the 514 simulated peaks are found at SPL and both NWR/HDP, respectively, the peaks coincide 515 with times when average trajectories are found within a relatively shallow morning PBL. 516 As the air parcels move backward in time, when the morning transitions backward in 517 time to the nighttime, many of them would still be found within the shallow nighttime 518 PBL. Due to the shallowness of the nocturnal PBL, the footprint values for the air 519 parcels found there would be high. These parcels would also be sampling the nighttime  $CO_2$  release and therefore lead to enhancements in  $CO_2$ . In other words, the erroneous 520 521 daytime peak reflects enhanced CO<sub>2</sub> that is vented up to the observing height within the 522 model during the day. We suspect that something similar is taking place in other global 523 models, leading to similar erroneous daytime CO<sub>2</sub> peaks (Geels et al., 2007).

#### 524 **4. Discussion**

This study has sought to answer the question: how can mountaintop  $CO_2$  observations be used to constrain regional scale carbon fluxes, given the complex terrain and flows in the vicinity of mountaintop sites? To address this question, we have driven a Lagrangian particle dispersion model simulating the transport of turbulent air parcels arriving at 3 mountaintop  $CO_2$  sites in the Western U.S. We then examined potential differences in simulated results as the atmospheric simulations are driven by meteorological fields resolved with differing grid spacings and at different vertical levels.

532 We found that the observed average diurnal  $CO_2$  pattern is better reproduced by 533 simulations driven by WRF-1.3km and WRF-4km ("AGL" configuration), with minimal 534 differences between the two configurations (Fig. 3). The coarser-scale models (WRF-535 12km AGL, GDAS-1°, and CarbonTracker) fail to reproduce the observed diurnal pattern at all 3 sites. The problem is especially severe at night, when both GDAS-ASL 536 537 and CarbonTracker lack the nocturnal enhancements. In contrast, WRF-12km (AGL) 538 shows nocturnal CO<sub>2</sub> buildup that is clearly too strong. The overestimation problem is 539 exacerbated when both coarser grid spacing and "AGL" configuration are adopted, as 540 seen in GDAS-AGL at HDP (Fig. 3).

541 The overestimate in nighttime  $CO_2$  from WRF-12km (AGL) is due to the 542 preponderance of simulated air parcels found within the nocturnal PBL (Figs. 7~9),

- 543 which can be traced to the fact that air parcels are closer to the ground surface when
- mountains are flattened. Conversely, when released at "ASL" levels air parcels are found
- 545 much higher above the nocturnal PBL due to the flattening of mountains in a coarse-scale
- 546 global model like GDAS, resulting in minimal sensitivity to nighttime biospheric fluxes
- and lack of CO<sub>2</sub> buildup. Such large errors in estimated carbon fluxes due to lack of
- ability to resolve patterns have also been found in earlier studies in Europe (Pillai et al.,
  2011;Peters et al., 2010).
- 550 The natural question, then, is what can researchers do with mountaintop  $CO_2$ 551 observations, given the difficulty in resolving the terrain and flows in complex terrain?

## 4.1 Approach 1: Adjust vertical level of simulations from which to compare against observed values

- 554 The diurnal cycle simulated within CarbonTracker varies significantly as a function of
- the vertical level (Fig. S6) from which  $CO_2$  is extracted, particularly at night. The
- strongly attenuated diurnal cycle in the interpolated level corresponding to the ASL
- elevation of the mountaintop sites (orange dashed) is found at higher levels within
- 558 CarbonTracker too, away from the first few levels near the ground. At HDP, the
- nighttime depletion of  $CO_2$  at lower levels appears to be due to the erroneous nighttime photosynthetic uptake in the gridcell where HDP is located (Fig. S2).
- 561 Interestingly, at SPL and NWR the diurnal pattern at a level between Levels 2 and 3 562 appears to correspond more closely to the overall observed  $CO_2$  diurnal cycle, perhaps 563 due to the presence of nighttime enhancements closer to the model surface that is absent 564 from the higher levels closer to the ASL elevation. The closer correspondence to observed patterns may call for researchers to adjust the vertical level to maximize 565 566 resemblance to observations. This was carried out at Jungfraujoch (Folini et al., 2008), 567 where the authors simulated carbon monoxide (CO) at multiple heights and arrived at a 568 height of 80 m above the model's ground surface as the best correspondence with the 569 observed CO, which was measured closer to the ground (Rinsland et al., 2000). Instead, 570 a different study simulating observations at the same site adopted a height of 830 m 571 above the model ground surface (Tuzson et al., 2011). This example illustrates the 572 divergence in researchers' choices for the vertical level in the midst of mountainous 573 terrain.

574 It is worth noting that the introduction of additional degrees of freedom in the vertical 575 level in "fitting" the measured  $CO_2$  diurnal cycle within a carbon assimilation system is 576 potentially problematic. The reason is that the assimilation system seeks to solve for 577 carbon fluxes by examining the mismatch between observed versus simulated CO<sub>2</sub> 578 concentrations. If the mismatch is due to erroneous fluxes, the introduction of additional 579 degrees of freedom in the vertical level would compensate for erroneous fluxes. For 580 instance, if the nighttime carbon fluxes are overestimated in the model, this should show 581 up as an enhanced CO<sub>2</sub> concentration that is larger than observed values. However, this 582 overestimation in CO<sub>2</sub> would be reduced by picking a higher vertical level rather than 583 fixing the overly large efflux in the model. The optimal level could differ between night 584 and day as well; for instance, a level higher than Level 2 would fit better against 585 observations during the daytime at SPL and NWR (Fig. S6). If different levels are 586 adopted at different times of the day, the degrees of freedom that can be adjusted would 587 be even larger, and model-data mismatches would be used in vertical level adjustments 588 instead of correcting erroneous biospheric fluxes.

- Regardless, there is some role for vertical level adjustments to remove the gross mismatch in the observed vs simulated diurnal cycles. If the vertical level is indeed adjusted in a carbon inversion system, we suggest that additional information (e.g., comparisons to meteorological observations or other tracers) is used rather than maximizing the match to the target species (i.e., CO<sub>2</sub>, in the case of a carbon inversion
- 594 system).

The  $CO_2$  values at multiple levels within CarbonTracker show that unlike the nighttime, differences between vertical levels are much smaller during the afternoon at SPL and NWR (Fig. S6), suggesting that the simulated  $CO_2$  values are not as sensitive to the choice of vertical level. We suspect that the large differences between vertical levels at HDP are due to the flipped diurnal cycle in biospheric fluxes within CarbonTracker (Fig. S3). Otherwise, the lack of sensitivity to the choice of vertical level suggests that

601 coarse-scale models should assimilate afternoon observations, rather than nighttime 602 observations (see "Approach 3" below).

#### 603 **4.3 Approach 2: Assign errors to account for model errors**

604 Instead of neglecting the mountaintop CO<sub>2</sub> observations altogether, an alternative

- approach is to make use of the observations, but assigning them errors within the model-
- 606 measurement discrepancy error covariance matrix to account for model deficiencies (Lin
- and Gerbig, 2005;Gerbig et al., 2008). One estimate of the model-measurement
- discrepancy error is the root-mean-square error (RMSE), which ranges from less than 3
- ppm for WRF-1.3km to over 7 ppm for WRF-12km (Fig. S5). In this way, the inversion
- 610 system would assign less weight to observations that the model has difficulties simulating.
- 611 Given the systematic misrepresentation of the diurnal cycle in coarse-scale models, 612 particularly at night (Fig. 3), this approach will effectively throw away much of the data
- 613 as noise, due to inadequacies in the model. This naturally leads to the next possible
- as noise, due to inadequacies in the model. This naturally leads to the next possible approach of just having coarse-scale models assimilate afternoon observations
- approach of just having coarse-scale models assimilate afternoon observations.

## 4.4 Approach 3: Have coarse-scale models assimilate afternoon observations instead of nighttime

617 Our results show that the simulated  $CO_2$  values are more in accordance with observed 618 values in the afternoon (Fig. 3). This follows from the fact that afternoon trajectories and 619 footprints match their higher resolution counterparts (Figs. 6, S8, S10, S11~S13), likely

- 620 due to the deeper afternoon PBL depth and the reduction of terrain effects (Stevn et al.,
- 621 2013). In other words, relative differences in PBL depth associated with flattening of
- mountains are lessened when the PBL is deeper; thus the impact on whether an air parcel
- sampled by the mountaintop site falls within the PBL is also attenuated under afternoonvigorous mixing conditions.
- Based on these results, and in lieu of better transport, we suggest coarse-scale models may be better served to assimilate afternoon observations over the continent at their above sea level elevation. This is contrary to what has been commonly practiced by researchers, when nighttime mountaintop observations were assimilated (Peters et al., 2007;Keeling et al., 1976) to avoid daytime upslope flows and when nocturnal
- 629 2007; Keeling et al., 1976) to avoid daytime upslope flows and when nocturnal
- 630 observations that represent free tropospheric conditions would better match coarse 631 resolution models. We have found that sampling coarse-scale (1 deg) models at the
- 632 corresponding ASL height have significant difficulties simulating nighttime CO<sub>2</sub>, since it
- 633 appears that the model failed to represent the strength of the nocturnal footprint at the 3

RACCOON mountaintop sites (Figs. 4, 5). Thus the inability of coarse-scale models to
simulate the transport and PBL depths result in the lack of nocturnal enhancements and
thereby the wrong diurnal cycle (Fig. 3). Conversely, sampling the 12-km simulation at
the AGL height also has significant difficulties simulating nighttime CO<sub>2</sub>, because it
overestimates the nocturnal footprint.

639 However, careful attention needs to be paid to upslope flows in the afternoon and 640 the potential mis-interpretation of more localized biospheric signals or anthropogenic 641 signals from below the mountain. A study from Jungfraujoch in Europe suggested that as 642 much as ~40% of the days in a year are influenced by thermally driven flows (Griffiths et 643 al., 2014). During the afternoon, the mountaintop site would then be influenced by 644 thermally driven upslope winds, as also pointed out by a number of studies around NWR, 645 along the Colorado Front Range (Sun et al., 2010; Sun and De Wekker, 2011; Parrish et al., 646 1990) as well as SPL (De Wekker et al., 2009). For sites like HDP and NWR, which 647 have large nearby urban areas at lower elevation, upslope conditions can be of particular 648 concern if not properly accounted for. If these sites experience elevated CO<sub>2</sub> in the 649 afternoon from pollution sources, and this transport is not captured by the models, then 650 natural CO<sub>2</sub> sources can be significantly overestimated.

651 We found it encouraging that despite the proximity of significant population and 652 anthropogenic emissions from the Salt Lake and Denver area to the HDP and NWR sites, 653 respectively, the WRF-1.3km model suggests that the additional contribution of 654 anthropogenic  $CO_2$  in the afternoon, over and beyond the nighttime signal is less than 655 1ppm, on average (Fig. S4). Presumably this is because of the high elevation of HDP and 656 NWR in relation to the urban area and the dilution of signals as they move up slope; the 657 afternoon urban signal would be enhanced if the sites were placed at lower peaks.

658 Regardless, it is prudent to consider mountaintop sites as not necessarily "pristine" 659 sites and to consider potential contributions from surrounding anthropogenic emissions 660 on these observations. It has been estimated that as of the year 2000, over 10% of the 661 world population live in mountainous areas (Huddleston et al., 2003), meaning that any 662 mountaintop site could very well see anthropogenic signatures. We recommend additional tracers to be measured in conjunction with the mountaintop CO<sub>2</sub> sites. For 663 instance, combustion tracers such as  $C^{14}$  and CO (Levin and Karstens, 2007) have been 664 measured alongside  $CO_2$  at mountaintop sites in Europe. Another promising tracer is 665 Rn<sup>222</sup> (Griffiths et al., 2014), which provides a measure of surface exchange and would 666 667 help provide constraints on the exchange of air measured at the mountaintop with the surface. Co-located meteorological observations-whether in-situ or remotely-sensed 668 669 (e.g., radar, sodar, lidar)—to probe atmospheric flows and turbulent mixing would also 670 be of significant value in helping to interpret the tracer observations (Rotach et al., 671 2014;Banta et al., 2013).

#### 672 4.5 Approach 4: Adopt high-resolution modeling frameworks

673 The least problematic, though potentially costly in terms of computational time, approach

674 to reduce modeling errors when interpreting mountaintop  $CO_2$  observations is to adopt a

high resolution modeling framework. This conclusion was also arrived at by previous

- 676 studies (Pillai et al., 2011;van der Molen and Dolman, 2007;De Wekker et al., 2009).
- From our results, it appears that meteorological fields from WRF at 4-km grid spacing,
- driving a Lagrangian particle dispersion model, can reproduce most features from a 1.3-
- 679 km simulation, and generate a CO<sub>2</sub> diurnal cycle that qualitatively matches the observed

pattern. Once the WRF fields are degraded to 12-km grid spacing, the model fails tocapture such features.

- 682 While at least 4-km resolution in the meteorological fields is needed for the sites
- 683 examined here in the American Rockies, we anticipate that the minimum resolution
- would depend on the level of complexity in the terrain, the height of the observational
- 685 site, and relationship with surrounding sources/sinks.

#### 686 **5.** Conclusions

- Given the large extent of the Earth's surface covered by hills and mountains and the large 687 688 amount of biomass and potential for carbon storage in complex terrain (Fig. 1), we call 689 for expanded efforts in observing and modeling CO<sub>2</sub> and other tracers on mountaintop 690 sites. This study has illustrated the potential for even coarse-scale models to extract 691 information from these observations when focusing on the daytime, afternoon values, and 692 the ability of high resolution models to simulate the general features of the summertime 693 diurnal  $CO_2$  cycle even in the midst of significant terrain complexity. However, we 694 acknowledge that even the highest resolution model adopted in this paper undoubtedly is 695 subject to limitations of its own, and that deviations between simulated versus observed 696 CO<sub>2</sub> diurnal cycles arise from errors in both atmospheric transport as well as the 697 biospheric fluxes. Due to the focus on atmospheric transport in this paper, errors in the 698 simulations caused by shortcomings in the biospheric fluxes remain outside the scope of
- this study (except for corrections to the flipped diurnal cycle; Fig. S3)
- Even though current models remain imperfect, we call for sustained and expanded observations of  $CO_2$  and other tracers (e.g., CO, <sup>222</sup>Rn, and the isotopes of  $CO_2$ ) colocated with meteorological observations on mountaintop sites to create enhanced datasets that can be further utilized by modeling frameworks of the future. Finally, we call for testing and gathering of three-dimensional  $CO_2$  observations over complex terrain, as revealed by intensive airborne campaigns like the Airborne Carbon in the Mountains Experiment (Sun et al., 2010).
- 707
- 708

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#### 722 Figure Captions

#### 723 **Fig. 1**

Aboveground biomass [mega-tons of carbon] from the North American Carbon Program baseline dataset for year 2000 (Kellndorfer et al., 2013) overlaid on topographic surface

in the Western U.S., resolved at  $0.5^{\circ} \times 0.5^{\circ}$  grid spacing.

#### 727 Fig. 2

- The WRF simulation domain, covering the Western U.S. with a series of nests with 12-,
- 4-, and 1.3-km grid spacing. These WRF meteorological fields are used to drive air
- 730 parcel trajectories within the STILT model.

#### 731 Fig. 3

- The average diurnal CO<sub>2</sub> pattern during June~August 2012 as observed at the 3
- mountaintop sites in the RACCOON network: Hidden Peak (HDP), Storm Peak
- 734Laboratory (SPL), and Niwot Ridge (NWR).Compared against the observations are
- simulated diurnal CO<sub>2</sub> patterns from different models: CarbonTracker, STILT driven
- vith WRF at different grid spacings, and STILT driven with GDAS. Multiple GDAS-
- driven STILT model configurations are shown, including runs without fixes to the
- biospheric fluxes ("biofluxorig"; see Supplemental Information), as well as releasing air
- parcels at the elevations of the sites above mean seal level ("ASL") or, for HDP only, at
- the inlet height (Table 1) above the model's ground level ("AGL"). All of the WRF-
- driven STILT runs place the release point of air parcels following the AGL configuration.
- Error bars denote standard errors of the diurnal averages.

#### 743 **Fig. 4**

- The average diurnal footprint strengths at HDP, SPL, and NWR over June~August 2012
- from STILT, driven with different meteorological fields and release heights (ASL vs
- AGL). The footprint strength was derived by summing over the spatial distribution of
- 747 footprint values (Fig. 5).

#### 748 Fig. 5

- The average footprint (shown in  $log_{10}$ ) for the Hidden Peak (HDP) site in Utah, at night: 0200 MST (0900 UTC), gridded at  $0.1^{\circ} \times 0.1^{\circ}$ . The site is denoted as a triangle. The
- 750 = 0200 MST (0900 UTC), gridded at  $0.1 \times 0.1^{\circ}$ . The site is denoted as a triangle. The
- average back trajectory (averaged over the stochastic STILT trajectories) is drawn as a
- line, with points indicating trajectory locations every hour, as the trajectory moves backfrom the site indicated as points. Magenta parts of the trajectory refer to the nighttime
- from the site indicated as points. Magenta parts of the trajectory refer to the nighttime
   (1900~0700 MST), while pink portions indicate the daytime (0700~1900 MST). Parts of
- the trajectory are shaded with blue when it is found below the average height of the PBL
- 755 the trajectory are shad 756 along the trajectory.

#### 757 **Fig. 6**

758 Similar to Fig. 5, but for the afternoon: 1400 MST (2100 UTC) at HDP.

#### 759 **Fig. 7**

- 760 Three dimensional plots of the terrain over a domain of  $\sim 1^{\circ} \times 1^{\circ}$  surrounding HDP, as
- resolved by the WRF and GDAS models at various grid spacings. The HDP site is
- denoted as a triangle. Also shown is the average back trajectory, derived by averaging

- 763 locations of the numerous stochastic trajectories simulated by STILT, driven by the
- various WRF meteorological fields and the global GDAS field. Back trajectories were
- started from HDP at 0200 MST (0900 UTC). Points indicate trajectory locations every
- hour, as the trajectory moves back from the site indicated as points. Magenta portions of
- the trajectory refer to the nighttime (1900~0700 MST), while pink portions indicate the
- daytime (0700~1900 MST). In addition, the PBL heights averaged along the
- 769 backtrajectory are shown as the blue line.

#### 770 Fig. 8

- 771 Time series of the average back trajectory and PBL heights relative to the ground surface
- ("AGL") instead of above sea level, at each time step backward in time from the receptor
- 773 (triangle). Magenta portions of the trajectory refer to the nighttime (1900~0700 MST),
- while pink portions indicate the daytime (0700~1900 MST). The PBL heights averaged
- along the backtrajectory are shown as the blue line. The nighttime PBL height is
- indicated in dark blue, while the daytime portion is in light blue. The height of the site is
- indicated by the black triangle at the starting time of the back trajectory.

#### 778 **Fig. 9**

779 Similar to Fig. 7, but for the Storm Peak Laboratory (SPL) site.

#### 780 Fig. 10

781 Similar to Fig. 8, but for the Storm Peak Laboratory (SPL) site.

#### 782 Fig. 11

783 Similar to Fig. 9, but for the Niwot Ridge (NWR) site.

#### 784 Fig. 12

785 Similar to Fig. 10, but for the Niwot Ridge (NWR) site.

#### 786 **Fig. 13**

- 787 Similar to three-dimensional terrain and trajectory plots as shown in Figs. 7, 9, and 11,
- but for just the GDAS 1 deg. ASL simulations and for the morning hours of 0800 MST
- and 1100 MST.

#### Tables 792

	Hidden Peak (HDP)	Storm Peak Lab (SPL)	Niwot Ridge (NWR)
Latitude/Longitude	40° 33' 38.80" N 111° 38' 43.48" W	40° 27' 00" N 106° 43' 48" W	40° 03' 11" N 105° 35' 11" W
Top Inlet Height	17.7 m	9.1 m	5.1 m
Site Altitude [m above sea level]	3351 m	3210 m	3523 m
Site Altitude as Resolved by Models [m above sea level]:			
WRF-1.3km	2996 m	3038 m	3411 m
WRF-4km	2918 m	2818 m	3382 m
WRF-12km	2357 m	2724 m	3076 m
GDAS	1856 m	2757 m	2333 m
CarbonTracker	2004 m	2582 m	2276 m

795 
**Table 1**. Characteristics of RACCOON mountaintop sites examined in this paper, as well as the representation of terrain in different meteorological files at these sites.

797

Site	SPL			NWR				
Run type	1.3- km WRF	4-km WRF	12-km WRF	GDAS	1.3- km WRF	4-km WRF	12-km WRF	GDAS
u-wind BIAS [m/s]	-0.5	-1.5	-0.9	2.3	0.1	-0.3	-1.4	-0.2
v-wind BIAS [m/s]	-0.6	-0.3	-0.2	1	0.2	0.4	0.9	1.1
u-wind RMSE [m/s]	3.1	3.8	3.2	3.7	3.5	3.4	3.4	3.2
v-wind RMSE [m/s]	2.7	2.7	2.3	2.5	2.2	2.1	2.2	3

798

799 Table 2. Comparisons of different meteorological files driving STILT against hourly-

800 averaged wind observations at Storm Peak Laboratory (-106.74 W; 40.45 N) and at

801 Niwot Ridge (-105.586 W; 40.053 N; 3502 m ASL) (Knowles, 2015), near the

802 RACCOON CO<sub>2</sub> site. Meteorological observations were not available at the Hidden Peak

site. Error statistics are presented separately for the west-to-east component ("u-wind")

and south-to-north component ("v-wind") of the wind velocity vector.

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### Above-ground Biomass in the Western U.S.





## **WRF** Domains







HDP ave footprint: WRF-1.3km (AGL)

#### HDP ave footprint: WRF-4km (AGL)



HDP ave footprint: WRF-4km (AGL)





Fig. 7



HDP: Mean 3D Trajectory of Stochastic Particles & PBL ht (above ground level) 0900 UTC (0200 MST)



#### SPL: Mean 3D Trajectory of Stochastic Particles & PBL ht for Different Runs 0900 UTC (0200 MST)



# SPL: Mean 3D Trajectory of Stochastic Particles & PBL ht (above ground level) MST 0900 UTC (0200 MST) MST



#### NWR: Mean 3D Trajectory of Stochastic Particles & PBL ht for Different Runs





## NWR: Mean 3D Trajectory of Stochastic Particles & PBL ht (above ground level) MST 0900 UTC (0200 MST) 14:00 16:00 18:00 22:00 00:00 02:00 14:00 16:00 18:00 20:00 02:00



### Mean 3D Trajectory of Stochastic Particles & PBL ht