## Regional-scale, sector-specific evaluation of global CO2 inversion models using aircraft data from the ACT-America project

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#### Abstract

We use 148 airborne vertical profiles of CO2 for frontal cases from the summer 2016 Atmospheric Carbon and Transport-America (ACT-America) campaign to evaluate the skill of ten global CO2 in situ inversion models from the version 7 Orbiting Carbon Observatory-2 (OCO-2) Model Intercomparison Project (MIP). Model errors (model posterior-observed CO2 dry air mole fractions) were categorized by region (Mid-Atlantic, Midwest, and South), frontal sector (warm or cold), and transport model (predominantly Tracer Model 5 (TM5) and Goddard Earth Observing System-Chemistry (GEOS-Chem)). All inversions assimilated the same CO2 observations. Overall, the median inversion profiles reproduce the general structures of the observations (enhanced / depleted low-level CO2 in warm / cold sectors), but 1) they underestimate the magnitude of the warm / cold sector mole fraction difference, and 2) the spread among individual inversions can be quite large (> 5 ppm). Uniquely in the Mid-Atlantic, inversion biases segregated according to atmospheric transport model, where TM5 inversions biases were-3 to-4 ppm in warm sectors, while those of GEOS-Chem were +2 to +3 ppm in cold sectors. The large spread among the mean posterior CO2 profiles is not explained by the different atmospheric transport models. These results show that the inversion systems themselves are the dominant cause of this spread, and that the aircraft campaign data are clearly able to identify these large biases. Future controlled experiments should identify which inversions best reproduce midlatitude CO2 mole fractions, and how inversion system components are linked to system performance.

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# Regional-scale, sector-specific evaluation of global CO<sub>2</sub> inversion models using aircraft data from the ACT-America project

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### **Key Points:**

- Global inversions capture observed CO<sub>2</sub> variations across North America for summer 2016, but underestimate warm / cold sector differences.
- Inversion variability appears to be mainly driven by differences in inversion system design and/or prior fluxes, not transport model.
- Uniquely in the Mid-Atlantic, transport-dependent CO<sub>2</sub> biases exist, present in both warm and cold sectors.

#### Abstract

We use 148 airborne vertical profiles of  $CO_2$  for frontal cases from the summer 2016 Atmospheric 1 Carbon and Transport – America (ACT-America) campaign to evaluate the skill of ten global CO<sub>2</sub> 2 in situ inversion models from the version 7 Orbiting Carbon Observatory - 2 (OCO-2) Model 3 Intercomparison Project (MIP). Model errors (model posterior – observed CO<sub>2</sub> dry air mole 4 fractions) were categorized by region (Mid-Atlantic, Midwest, and South), frontal sector (warm or 5 cold), and transport model (predominantly Tracer Model 5 (TM5) and Goddard Earth Observing 6 7 System – Chemistry (GEOS-Chem)). All inversions assimilated the same CO<sub>2</sub> observations. Overall, the median inversion profiles reproduce the general structures of the observations 8 (enhanced / depleted low-level CO<sub>2</sub> in warm / cold sectors), but 1) they underestimate the 9 magnitude of the warm / cold sector mole fraction difference, and 2) the spread among individual 10 inversions can be quite large (> 5 ppm). Uniquely in the Mid-Atlantic, inversion biases segregated 11 according to atmospheric transport model, where TM5 inversions biases were -3 to -4 ppm in warm 12 sectors, while those of GEOS-Chem were +2 to +3 ppm in cold sectors. The large spread among 13 the mean posterior CO<sub>2</sub> profiles is not explained by the different atmospheric transport models. 14 These results show that the inversion systems themselves are the dominant cause of this spread, 15 and that the aircraft campaign data are clearly able to identify these large biases. Future controlled 16 experiments should identify which inversions best reproduce midlatitude CO<sub>2</sub> mole fractions, and 17 how inversion system components are linked to system performance. 18

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#### 20 1 Introduction

To mitigate the effects of climate change due to greenhouse gases (GHGs), it is vital that 21 we understand the exchange of CO<sub>2</sub> between the atmosphere, biosphere, and ocean, and how these 22 exchanges might vary with time. Previous studies have shown that the land biosphere is a 23 24 substantial sink of atmospheric CO<sub>2</sub> at present (e.g., Tans et al., 1990; Denning et al., 1995; Gurney et al., 2004; Ballantyne et al. 2012), but much remains unknown about its precise magnitude and 25 long-term trajectory, what accounts for its variability, and to what extent it is tropical or 26 extratropical (Schimel et al., 2015; Crowell et al., 2019). In part, this lack of understanding is due 27 to a gap in observational coverage at regional-to-continental scales (spatially, approximately  $10^3$ 28  $-10^4$  km). While eddy flux towers can provide measurements of surface CO<sub>2</sub> fluxes at local scales 29 (Luyaessert et al., 2007), and hemispheric-to-global fluxes can be constrained by mole fraction 30 measurements at remote sites (Ciais et al., 2019), at intermediate scales neither method is 31 completely satisfactory. Measurements of CO<sub>2</sub> mole fraction from *in situ* stations do not have a 32 straightforward relationship to surface fluxes on intermediate scales (Stephens et al., 2007), and 33 these stations are predominantly located in the mid-latitudes, limiting the assessment of surface 34 fluxes in tropical and high-latitude regions (Peylin et al., 2013). 35

The current state-of-the-art method of generating a full atmospheric analysis of  $CO_2$  mole fractions and surface fluxes on regional-to-global scales is through the use of global flux inversion models (Enting et al., 1995; Bousquet et al., 2000; Peylin et al., 2013; Basu et al., 2018). These Bayesian optimization systems use global atmospheric transport models to simulate mole fraction fields from a given prior distribution of surface fluxes, and then "invert" the transport to find the optimal surface fluxes that minimize errors between observed mole fractions and those that result from forward simulations of those optimal surface fluxes ('posterior  $CO_2$ '; Tarantola, 2005). The

posterior mole fraction field provides an analysis of unobserved CO<sub>2</sub> distributions in synoptic 43 weather systems down to the resolution limit of these models, generally a few degrees of latitude 44 /longitude (e.g. Peters et al. 2007). The accuracy of these global inversions, however, is dependent 45 on choice of data inversion procedure, prior flux model, and transport model used within the global 46 inversions, any of which can vary considerably among different inversion groups (Peylin et al., 47 2013). In particular, this often manifests itself as an inability to associate errors in modeled CO<sub>2</sub> 48 uniquely with transport or surface flux errors. The absence of sufficiently dense independent data 49 networks hinders the ability to support or refute particular inversion results. 50

Tropospheric profiles of GHGs have been obtained for the last two decades at 51 approximately biweekly intervals by the National Oceanographic and Atmospheric Administration 52 (NOAA) light aircraft profiler network at over 22 sites in North America, and have provided 53 independent assessment of the skill of global inversions throughout the troposphere (Sweeney et 54 al., 2015). These data have been used to show evidence of increased CO<sub>2</sub> depletion in the boreal 55 growing season from west-to-east across the North American continent, consistent with a surface 56 biogenic sink and a prevailing eastward motion of air masses, though some of the depletion was 57 attributed to Eurasian sinks upstream (Sweeney et al., 2015; Lan et al., 2017). The CarbonTracker 58 global inversion system, version CT2015 (Peters et al., 2007, with updates documented at 59 http://carbontracker.noaa.gov ) was shown by Lan et al. (2017) to have horizontal gradients of 60 column-averaged  $CO_2$  (XCO<sub>2</sub>) that compare well with the available data. Stephens et al. (2007) 61 used the profiles to evaluate the seasonal vertical  $CO_2$  gradients of 12 global inversions from the 62 Transcom 3 intercomparison experiment (Gurney et al., 2004; Baker et al., 2006), and found that 63 the inversions tended to underestimate the observed positive vertical gradient during the boreal 64 growing season, which was attributed to overestimation of vertical mixing. It was shown on these 65 timescales that overestimated vertical mixing in an inversion that assimilates near-surface mole 66 fraction data led to an overestimated net surface flux of CO<sub>2</sub>, though the correlation was weaker 67 during the growing season. The recent study of Gaubert et al. (2019) found better agreement 68 between inversion vertical CO<sub>2</sub> gradients and oceanic observations in the annual mean, but biased 69 vertical gradients were still present when just boreal summer was considered. 70

71 The spatial and temporal scale of the NOAA long-term aircraft profiles are too coarse to 72 resolve structures found in individual synoptic weather systems, which progress over the continent 73 on a time scale of days, and have airstream sectors (warm conveyor belt, cold conveyor belt, dry intrusion) on scales of hundreds to thousands of km. Furthermore, as noted by Sweeney et al. 74 75 (2015), the profiles tend to preferentially sample fair-weather conditions. While over a long time period these observations help constrain CO<sub>2</sub> fluxes and transport, they do not adequately sample 76 the structures of synoptic weather systems that play an important role in vertical and latitudinal 77 transport of  $CO_2$  (Schuh et al, 2019). More recently, field campaigns using more extensive 78 sampling of GHG with aircraft have occurred, such as the Atmospheric Tomography Mission 79 (ATom; Wofsy et al. 2018) and the O<sub>2</sub> / N<sub>2</sub> Ratio and CO<sub>2</sub> Airborne Southern Ocean Study 80 (ORCAS; Stephens et al. 2018), but these generally did not target the North American continent. 81 Tower-based observations (Andrews et al., 2014), while continuous in time are spatially sparse 82 and limited to the planetary boundary layer (PBL). Model-data comparisons using tower-based, 83 in situ CO<sub>2</sub> observations have shown large model-data differences (Diaz-Isaac et al, 2014; 2018) 84 but have not yielded direct insight into the representation of weather systems in atmospheric CO<sub>2</sub> 85 simulations. 86

Recently, dedicated satellites have been launched that can infer XCO<sub>2</sub> from shortwave 87 infrared observations, including the Greenhouse gases Observing SATellite (GOSAT: Kuze et al., 88 2009), and the Observing Carbon Observatory -- 2 (OCO-2) satellite platform launched by NASA 89 90 in 2014 (Crisp 2005; Eldering et al., 2017). These programs have the goals of increasing global data and coverage and reducing observational dependence on vertical mixing. 91 To assess the sensitivity of the global inversions to OCO-2 data as well as to the different choices of inversion 92 procedure mentioned above, NASA organized the OCO-2 Model Intercomparison Project (OCO-93 2 MIP) including 11 global inversion groups (Crowell et al., 2019). As part of the OCO-2 MIP, a 94 set of standardized numerical experiments with varying transport models, optimization techniques, 95 and prior surface fluxes were performed with different sets of common assimilated data; one set 96 of experiments assimilated OCO-2 retrievals, while another set only assimilated standardized in 97 situ observations (henceforth the IS inversions). Among other findings, they found generally 98 small, but positive (< about +1 ppm) biases among the IS inversions relative to independent aircraft 99 observations in the northern extratropics. However, overall variability among models constrained 100 with IS data remained high, and the independent aircraft datasets were deemed too sparse to be 101 able to make more specific assessments of the skill of different inversion techniques. 102

Schuh et al. (2019) noted systematic differences within the OCO-2 MIP between inversions 103 that used the Goddard Earth Observing System – Chemistry (GEOS-Chem; Bey et al., 2001) 104 105 transport model, and those that used the Tracer Model 5 (TM5; Krol et al., 2015). Poleward of 45 N, the GEOS-Chem IS inversions had reduced growing season surface uptake, and overall seasonal 106 flux amplitude, compared with the TM5 inversions, although large variability among the full 107 108 inversion systems precluded strict statistical significance. They then performed controlled experiments, running forward simulations with the same surface fluxes (from CarbonTracker 109 CT2016) for both TM5 and GEOS-Chem. These confirmed that, relative to TM5, GEOS-Chem 110 had a tendency to 'trap' surface flux signals near the surface and advect them poleward; thus 111 GEOS-Chem would require reduced seasonal cycles of local CO<sub>2</sub> surface fluxes to match the same 112 set of near-surface mole fraction measurements. Outstanding questions include which transport 113 model is closer to observations, and whether these characteristics are due to differences in vertical 114 or horizontal mixing (e.g., GEOS-Chem could have greater meridional transport of  $CO_2$  in the 115 midlatitude Ferrel cells (Peixoto and Oort, 1992; Pauluis et al., 2009; Parazoo et al., 2011), 116 increasing relative horizontal mixing but reducing relative vertical mixing of  $CO_2$ ). TM5 vs. 117 GEOS-Chem forward-transport experiments with the anthropogenic tracer  $SF_6$  in Schuh et al. 118 119 (2019) suggested that TM5 transport produced better agreement with marine near-surface observations, but transport model skill above the boundary layer was not known. 120

To increase our understanding of GHG mole fractions and fluxes over North America, the 121 Atmospheric Carbon Transport -- America (ACT-America) NASA Earth Venture Suborbital 2 122 (EVS-2) mission (Davis et al., 2018; Baier et al., 2019; Pal et al. 2020) is using a combination of 123 aircraft, satellite, and tower-based observational platforms, including a series of aircraft field 124 campaigns that cover three focus regions (the U.S. Mid-Atlantic, Midwest, and South) and all four 125 seasons, beginning with summer 2016. During each six-week campaign, flight plans for each 126 individual flight day were designed based on forecast meteorology for either investigating multiple 127 sectors of frontal weather systems, sampling large-scale mole fractions over fair weather boundary 128 layers, or providing under-flights of OCO-2 passages to help evaluate its retrievals. It was 129 hypothesized that the synoptic-scale variability of CO<sub>2</sub> on frontal days would have strong 130 sensitivity to atmospheric transport, and that these data could be used to evaluate transport models 131 132 and determine the optimal ones to use within flux inversion systems.

Pal et al. (2020) presents an observational analysis of the aircraft *in situ*  $CO_2$  and  $CH_4$  mole 133 fractions from the frontal cases of the summer 2016 ACT-America campaign, classifying data as 134 either 'warm sector' or 'cold sector', and within a sector into PBL, lower free troposphere, and 135 upper free troposphere. In the PBL they found systematically greater CO<sub>2</sub> mole fractions in warm 136 sectors than cold sectors (by 5-30 ppm), and, on average, warm / cold sector mole fractions were 137 enhanced / depleted relative to the free troposphere. (Henceforth, 'enhanced' and 'depleted' mean 138 higher / lower mole fractions relative to the free troposphere.) Free troposphere warm sectors also 139 had enhanced CO<sub>2</sub> relative to cold sectors, but by reduced magnitude (5 ppm or less) compared to 140 the PBL. Chen et al. (2019) used the vertical profile data from the summer 2016 and winter 2017 141 campaigns to evaluate the CO<sub>2</sub> mole fractions in a pair of global inversions available during the 142 period -- the ECMWF-based Copernicus Atmosphere Monitoring Service (CAMS) (Agusti-143 Panareda et al., 2014), and CarbonTracker Near-Realtime (CT-NRT), a version of the 144 CarbonTracker system that uses some climatologically-based flux priors and a subset of 145 observations to deliver a product with reduced processing time. They found the two inversions 146 agreed reasonably well with the independent ACT-America data. However, substantial low biases 147 for CT-NRT were noted in summer 2016 over the MidAtlantic region and in winter 2017 over the 148 Midwest; meanwhile, CAMS showed a positive low-level bias in the MidAtlantic during summer 149 2016, and a positive bias throughout the column in winter 2017. The uncertainty as inferred from 150 the model spread was deemed comparable to the model error. 151

In this study, we also use in situ CO<sub>2</sub> from vertical profiles during the frontal cases from 152 the summer 2016 ACT-America campaign, but here we apply the data to an assessment of the 153 whole suite of inversion models participating in the version 7 OCO-2 MIP study. We analyze the 154 structures and model errors of the profiles as a function of region, meteorological sector, and 155 transport model. We use only flight segments corresponding to vertical profiles because they 156 provide information about atmospheric vertical structure with a minimum of temporal and spatial 157 variability. In particular, this allows the comparison of modeled and observed CO<sub>2</sub> mole fraction 158 as a function of sector without the complication of mismatches between the model and observed 159 frontal position. For simplicity we only examine the OCO-2 MIP inversions that assimilated a 160 prescribed suite of in situ data (the IS experiment), with a special focus on the potential impact of 161 transport model, which in the OCO-2 MIP was predominantly either TM5 or GEOS-Chem. In 162 section 2 we provide more background on the data and model simulations and describe our 163 experimental procedure. Section 3 presents the resulting statistics of these vertical profiles, while 164 sections 4 and 5 include contain overall discussion and conclusions from the work. 165

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### 167 2 Data and Methodology

168 2.1 ACT-America aircraft measurements

The six-week summer 2016 ACT-America campaign took place from 15 July – 31 August 2016. For three successive two-week periods, the base of operations for the C-130 and Kingair B-200 aircraft was at NASA Langley Research Center / Wallops Flight Facility (Mid-Atlantic region), Lincoln, NE (Midwest region), and Shreveport, LA (South region). A total of 25 research flight days occurred – 7 in the Mid-Atlantic, 9 in the Midwest, and 9 in the South. We analyzed only the frontal case flight days from each region (for a list of case days, see the Supplemental Information, Table S1). Flights were conducted in midday or afternoon hours to focus on wellmixed PBL conditions. We used CO<sub>2</sub> mole fractions derived from the Picarro sensors on board
each aircraft. More information on the data used can be found in Davis et al. (2018) and Pal et al.
(2020).

The aircraft data from the summer 2016 ACT-America campaign was put into Obspack format (see https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/OCO2\_insitu/ for details). ACT-America data are publicly available from the NASA archive (https://www-air.larc.nasa.gov/cgibin/ArcView/actamerica.2016) and the ORNL Distributed Active Archive Center (Yang et al., 2018). Profiles identified for each ACT-America campaign are also available from ORNL (Pal, 2019).

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#### 186 2.2 OCO-2 MIP Suite

We used 10 different models from the version 7 OCO-2 MIP suite in this study, as listed 187 in Table 1. All but one are global CO<sub>2</sub> flux inversion models (i.e., they optimize CO<sub>2</sub> surface 188 fluxes on the basis of assimilated CO<sub>2</sub> observations). Three inversions use the TM5 transport 189 model, which uses ERA-Interim analyses (Berrisford et al., 2011) as a meteorological driver, while 190 four use the GEOS-Chem transport model, based on either the GEOS 'forward processing' 191 (GEOS-FP) system or the 2<sup>nd</sup> version of the Modern Era Retrospective analysis for Research and 192 Applications (MERRA-2) (Bosilovich et al., 2015) driver data. Of the remaining models, one 193 ('GEOS Model') is not an inversion, but uses the parent transport model (GEOS-5) of GEOS-194 Chem (Reinecker et al., 2008). 'CAMS' is an inversion that uses the Laboratoire de Météorologie 195 Dynamique – Zoom, version 3 (LMDz3) transport model (Chevalier et al. 2005), which makes use 196 197 of ERA-Interim, and so should have similar transport to the TM5 family. The PCTM transport model also makes use of MERRA-2 driver data, like the GEOS-Chem members, but differs mainly 198 in its modeling of vertical transport. Four variants of this model with different oceanic fluxes 199 200 participated in the MIP, but since all were similar over our regions of interest, only the variant using the NASA Ocean Biogeochemical Model was used in this study. Other details about the 201 MIP can be found in Crowell et al. (2019). 202

CO<sub>2</sub> mole fractions were extracted from all participating models in Obspack format at the times and locations corresponding to the ACT-America aircraft observations (also available at <u>https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/OCO2\_insitu/</u>).

206 2.3 Method

We used 148 flight segments identified by Pal et al. (2020) as either warm sector or cold sector vertical profiles for analysis. Each frontal day contained both warm sector and cold sector profiles as noted in Table S1. We performed the analysis for each combination of two sectors and three regions, where for simplicity we assumed that the sector classification of each modeled profile was the same as its corresponding observation.

We binned both modeled and observed CO<sub>2</sub> mole fraction into 250-m bins of altitude above ground level (AGL), derived by taking the geopotential height of each model / observation pair and subtracting the average ground elevation for the profile as reported in the NASA ORNL DAAC netCDF files. We performed the analysis for bins greater than 250 m AGL, with the exception that we excluded the 250-500 m AGL bin for times before 1700 UTC, to exclude

- boundary layers that might not be well mixed. We also truncated the profile analysis when bins
- 218 contained two or fewer sounding points.
- 219
- **Table 1**. Global inversions / models participating in OCO-2 MIP (adapted from Crowell et al.
- 221 (2019)).
- 222

| Name        | Transport | Driver  | Method      | Resolution | Reference      |
|-------------|-----------|---------|-------------|------------|----------------|
|             |           |         |             | (deg)      |                |
| CT-NRT      | TM5       | ERA-    | EnKF        | 2x3        | Peters et al., |
|             |           | Interim |             | (1x1 N.    | 2007 (with     |
|             |           |         |             | Amer.)     | online         |
|             |           |         |             |            | updates)       |
| OU          | TM5       | ERA-    | 4DVar       | 4x6        | Crowell et     |
|             |           | Interim |             |            | al., 2018      |
| CSU         | GEOS-     | MERRA-2 | Bayesian    | 1x1        | Schuh et al.,  |
|             | Chem      |         |             |            | 2010           |
| CMS-Flux    | GEOS-     | GEOS-FP | 4DVar       | 4x5        | Liu et al.,    |
|             | Chem      |         |             |            | 2014           |
| TM5-4DVar   | TM5       | ERA-    | 4DVar       | 2x3        | Basu et al.,   |
|             |           | Interim |             |            | 2013           |
| GEOS        | GEOS-5    | Inline  |             | 0.3125 x   | Rienecker et   |
| Model       |           |         |             | 0.25       | al., 2008      |
| CAMS        | LMDZ3     | ERA-    | Variational | 1.875 x    | Chevalier et   |
|             |           | Interim |             | 3.75       | al., 2005      |
| U Toronto   | GEOS-     | GEOS-FP | 4DVar       | 4x5        | Deng and       |
|             | Chem      |         |             |            | Chen, 2011     |
| U Edinburgh | GEOS-     | GEOS-FP | EnKF        | 4x5        | Feng et al.,   |
|             | Chem      |         |             |            | 2016           |
| РСТМ        | РСТМ      | MERRA-2 |             | 6.7x6.7    | Baker et al.,  |
|             |           |         |             |            | 2010           |

We estimated the variability of the mean observed mole fraction of each height bin with 224 the conventional standard error of the mean.  $\sigma/\sqrt{N}$ , where  $\sigma$  is the standard deviation of all five-225 226 second observations in each height bin. Choosing N to be the number of observations within each height bin, however, is strictly appropriate for independent observations. In reality, the mole 227 fractions within a particular vertical profile are highly autocorrelated, especially in the boundary 228 layer. We decided that the best estimate of the effective N and hence standard error of the mean 229 observed mole fraction was the number of vertical profiles contributing to each height bin. We 230 thus constructed vertical profiles of mean observed mole fraction by first averaging all data points 231 232 in each bin deriving from a common vertical profile, and then weighting all these individual profile averages equally to form the overall bin average mole fraction. The differences in vertical profiles 233 234 of observed mole fraction found by this method instead of simply averaging all five-second data points within each bin were minor. 235

For each of the global inversions, vertical profiles of mean modeled mole fraction were found using the same method as for the observations. For a given region, sector, and altitude bin, the relative magnitude of the bias (defined as model minus observation) to the standard error of the observations gives an indication of the significance of that model's bias. It is likely this is a conservative estimate of significance, because much of the variance of the observations (i.e., from profile to profile with our method) is correlated with variance in the modeled profile (i.e., the models have nonzero skill at predicting synoptic variability).

Providing an overall assessment of model biases relative to the observations is problematic 243 because the relatively small sample size and large variability in model properties preclude even a 244 rough application of Gaussian statistical methods. However, to give some indication of model-to-245 model consistency in biases, we show vertical profiles of the 25<sup>th</sup> quantile of modeled mole 246 fraction, the 50<sup>th</sup> quantile (i.e., the median), and the 75<sup>th</sup> quantile, along with the observed mole 247 fraction for comparison. We also show the ordered rank of the observation as a function of bin, 248 where a rank of 1 indicates the observation has a smaller mole fraction than any of the ten 249 inversions, and a rank of 11 indicates the observation has a larger mole fraction than any inversion. 250

To help quantify an overall model bias for each of the TM5 / GEOS-Chem inversion 251 groups, in each sector / region combination, we computed median statistics for each transport set 252 of inversions within each 250-m bin as described above. We then averaged the statistics into three 253 layers: below 1500-m AGL, 1500-3000 m AGL, and 3000-4500 m AGL. These layers roughly 254 correspond to the boundary layer (BL) / lower free troposphere (LFT) / upper free troposphere 255 (UFT) classification of Pal et al. (2020) (though in that study observed boundary layer height was 256 used instead of a strict height-based classification). We will henceforth refer to these layers as BL, 257 LFT, and UFT respectively for brevity, with the understanding that these designations are 258 259 approximate (in particular, BL may or may not correspond to the actual boundary layer).

260

### 261 **3 Vertical Profile Summary Statistics**

#### 262 3.1 MidAtlantic (16 Jul -- 31 Jul 2016)

During the Mid-Atlantic phase of the summer campaign, we observed in both warm and cold sectors similar CO<sub>2</sub> vertical profiles above about 1500 m AGL, with CO<sub>2</sub> gradually increasing with height, though the cold sector mole fractions were 2-4 ppm lower on average, and possessed considerably more within-bin and between-bin variability, especially in the UFT (Fig. 1).

Mean inversion posterior  $CO_2$  profiles are broadly consistent with the observations. Above 3000 m AGL, there is little difference between the model median warm and cold sector profiles (compare Fig. 2a and Fig 2b). There is a noticeable overall model negative bias in the upper-level median warm sector compared to observations (up to -2 ppm at 4500 m AGL).

Below 3000 m, the overall bias of median profiles is relatively small (1-2 ppm or less) and the median warm-sector profile captures the observed BL CO<sub>2</sub> enhancement quite well. However, for both sectors the interquantile BL model spread is very large (greater than 5 ppm). Individual inversions can have even greater deviations from the observations (e.g., Fig. 1). The fact that individual model BL variability is far larger than that observed in the UFT strongly suggests that the model spread is due to regional flux variability among the inversions, rather than variability in continental upstream boundary conditions.

While not completely accounting for the overall spread, when the inversions are grouped 278 279 by transport model, as shown in Fig. 1, systematic transport-related differences, on the order of the model biases, are apparent. Specifically, the TM5 models are lower in CO<sub>2</sub> mole fraction than 280 the GEOS-Chem models. For the warm sector profiles, all of the TM5 models had significant 281 negative CO<sub>2</sub> biases relative to observations below 3000 m AGL (see Fig. 1a), ranging from 2-7 282 ppm; in contrast, only one other inversion (a GEOS-Chem member) had a significant negative CO<sub>2</sub> 283 bias below 3000 m AGL, and most of the other inversions had significant positive biases. In 284 Table 2, we quantify regional and sector biases as a function of layer and transport model. The 285 corresponding absolute mole fractions are tabulated in Table S2 in the Supplement. We find that 286 the overall TM5 warm sector biases are significantly more negative (-3.9 ppm and -3.1 ppm for 287 the BL and LFT, respectively) than those found in GEOS-Chem (-1.2 ppm and -0.8 ppm). For the 288 cold sectors, the TM5 models are again around 2-5 ppm lower in mole fraction than the GEOS-289 Chem models below 3000 m AGL, if one outlier GEOS-Chem member is excluded (Fig. 1b); 290 however, in these cases it is the TM5 models which are closer to the observations (BL and LFT 291 biases are -1.3 ppm and -0.6 ppm for TM5, but 2.8 ppm and 2.0 ppm for GEOS-Chem). 292

It is of interest to examine model skill in reproducing the observed horizontal mole fraction 293 contrast across fronts, as well as the vertical boundary layer / free troposphere mole fraction 294 contrast within each sector, to gain insight into how well the models capture horizontal and vertical 295 transport. To quantify these respective contrasts, we can use the warm sector mole fraction - cold 296 sector mole fraction at each level in Table 2 (henceforth, "sector difference"), and the BL mole 297 fraction - LFT mole fraction for each sector in Table S2 (henceforth "vertical difference"), based 298 on Pal et al. (2020). We also include values of BL mole fraction – UFT mole fraction in Table 299 S2, which can be considered to represent the continental flux signal relative to the continental 300 background. What we find is that in the BL and LFT the observed sector difference (+4.1 ppm 301 302 and +2.4 ppm) is substantially underestimated by both sets of transport models (TM5: +1.9 ppm and -0.1 ppm; GEOS-Chem: 0.5 ppm and -0.4 ppm). On the other hand, the vertical differences 303 of the observations (-1.4 ppm in warm sectors, -3.5 ppm in cold sectors) are within 1 ppm of each 304 set of transport models (TM5: -2.2 ppm and -4.2 ppm; GEOS-Chem: -1.8 ppm, -2.7 ppm). Thus 305 the models in the MidAtlantic exhibit moderately large, transport-dependent biases in mole 306 fraction and mole fraction sector contrast, relatively consistent up to 3000 m. But the BL - UFT 307 308 differences are considerably larger for the TM5 models than either GEOS-Chem or the

observations (-6.5 ppm warm sectors and -7.2 cold sectors, vs. GEOS-Chem: -4.9 ppm warm and
 -5.0 ppm cold sectors, observations: -4.3 ppm warm sectors and -5.3 ppm cold sectors). This
 reflects large depletions of TM5 LFT CO<sub>2</sub> relative to its upper level background.



Figure 1. Vertical profiles of mean observed (black) and modeled (colored) mole fraction as a function of altitude AGL for Mid-Atlantic warm sectors (a) and cold sectors (b), from 16-31 Jul 2016. Blue indicates OCO-2 MIP models using GEOS-Chem offline transport; red indicate models using TM5 offline transport; light blue indicates the GEOS-5 inline transport member; orange indicates the CAMS member; green indicates the PCTM transport member. Bars on the

observations indicate  $\pm \sigma/\sqrt{N}$ , as described in text. Statistics generated from 36 total warm sector profiles and 12 total cold sector profiles.





Figure 2. Vertical profiles of mean observed mole fraction (black) and model spread of mole fraction (colored) as a function of altitude AGL for Mid-Atlantic warm sectors (a) and cold sectors (b), from 16-31 Jul 2016. Vertical colored curve indicates median (50<sup>th</sup> quantile) value of model mole fraction; bars indicate location of 25<sup>th</sup> and 75<sup>th</sup> quantile of model mole fraction. Numbers to right indicate ordered rank of observations relative to models, with 1 indicating observed value is less than all 10 model values, and 11 indicating observed value is greater than all 10 model values.

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- **Table 2**. Mole fractions biases of CO<sub>2</sub> in layers below 1500 m AGL (BL), 1500 3000 m AGL
- 330 (LFT), and 3000 m 4500 m AGL (UFT) for both TM5 and GEOS-Chem inversion, within each
- of the three regions in warm and cold sectors. Also shown are sector differences (warm sector
- mole fractions minus cold sector mole fractions) for each of GEOS-Chem, TM5, and the
- 333 observations for each layer and region. Values are the average of each 250-m bin median value
- 334 within each layer.

| Layer                       | Median<br>TM5<br>Warm<br>Sector<br>Bias | Median<br>GEOS-<br>Chem<br>Warm<br>Sector<br>Bias | Median<br>TM5<br>Cold<br>Sector<br>Bias | Median<br>GEOS-<br>Chem<br>Cold<br>Sector<br>Bias | TM5<br>Sector<br>Difference | GEOS-<br>Chem<br>Sector<br>Difference | Observed<br>Sector<br>Difference |
|-----------------------------|---|---|---|---|-----------------------------|---------------------------------------|----------------------------------|
| Mid-Atlantic                |   |   |   |   |                             |                                       |                                  |
| Below<br>1500 m (BL)        | -3.9                                    | -1.2  | -1.3                                    | +2.8  | +1.9                        | +0.5                                  | +4.5                             |
| 1500 m –<br>3000 m<br>(LFT) | -3.1                                    | -0.8  | -0.6                                    | +2.0  | -0.1                        | -0.4                                  | +2.4                             |
| 3000 m –<br>4500 m<br>(UFT) | -1.7                                    | -0.6  | +0.6                                    | +2.5  | +1.2                        | +0.4                                  | +3.5                             |
| Midwest                     |   |   |   |   |                             |                                       |                                  |
| Below<br>1500 m (BL)        | -0.8                                    | -1.5  | +3.6                                    | +3.3  | +8.7                        | +8.3                                  | +13.1                            |
| 1500 m –<br>3000 m<br>(LFT) | -0.3                                    | -0.5  | -1.6                                    | -2.3  | +5.7                        | +6.2                                  | +4.4                             |
| 3000 m –<br>4500 m<br>(UFT) | -0.5                                    | -0.9  | -0.6                                    | -0.8  | +1.9                        | +1.7                                  | +1.8                             |
| South                       |   |   |   |   |                             |                                       |                                  |
| Below<br>1500 m (BL)        | -0.8                                    | -1.3  | +2.3                                    | +1.8  | +4.0                        | +4.0                                  | +7.1                             |

| 1500 m –<br>3000 m<br>(LFT) | +0.4 | 0.0  | 0.0  | +0.1 | +3.7 | +3.2 | +3.3 |
|-----------------------------|------|------|------|------|------|------|------|
| 3000 m –<br>4500 m<br>(UFT) | +0.2 | -0.1 | -0.1 | +0.4 | +2.7 | +1.9 | +2.4 |

#### 337 3.2 Midwest (1 Aug -- 16 Aug 2016)

Unlike the MidAtlantic, in the Midwest the observed warm and cold sector profiles show little resemblance to each other. In the warm sectors, (Fig. 3a), little vertical variation of mean  $CO_2$  (1 ppm or less) is seen above 1500 m AGL. Below 1500 m AGL, many (though not all) warm sectors show a few ppm enhancement of  $CO_2$ . In contrast, the Midwest cold sectors (Fig. 3b) show the greatest low-level  $CO_2$  depletion relative to upper levels of any observed profile subset (almost 20 ppm), and vertical gradients of the mean  $CO_2$  profile exist throughout the whole atmosphere above 1000 m AGL.

345 Individual model biases for the Midwest warm sectors are on the order of 3 ppm or less at all levels in the profile, and the bias for the overall median model profile is on the order of 1 ppm. 346 The biases that are present seem to be significant, in that the model spread is even smaller, at least 347 above 1500 m. Between 3500 m and 7000 m AGL, the model mole fractions are too low in 348 virtually every case (Fig. 4a). Below 1000 m AGL the biases are small compared to observational 349 variability (Fig. 3a), but virtually all inversion members have lower mole fractions than the median 350 observed mole fraction, as seen by the high rank of the observations in Fig. 4a. While BL CO<sub>2</sub> 351 enhancements were not observed in every warm-sector case, when they were observed the models 352 underestimated them. The transport model differences appear to be about half of the overall bias 353 in both the UFT and BL (Table 2). But in terms of overall magnitude (< 1 ppm), these transport 354 model differences are substantially less than those in the MidAtlantic. 355

Though the observational standard errors are large, in the Midwest cold sectors above 1500 m AGL the modeled  $CO_2$  is lower than the observations for virtually all inversions (Fig. 4b). Below 1500 m, the model biases reverse sign, and become positive for virtually all members between 750 – 1250 m AGL. Thus the models systematically underestimate the mole fraction difference between about 1000 and 2500 m AGL. By 3000 m AGL, the Midwest cold sector negative observation-relative biases are reduced in magnitude, and remain relatively constant with additional height.



364

Figure 3: Same as Figure 1, but for the Midwest region (1-15 Aug 2016). Statistics generated from 29 total warm sector profiles and 29 total cold sector profiles.





Figure 4: Same as Figure 2, but for the Midwest region (1-15 Aug 2016).

In the BL, the observed Midwest sector difference is much larger in the Midwest than in the Mid-Atlantic (13 ppm). The models underestimate it by about 4 ppm (8.7 ppm in TM5, 9.0 ppm in GEOS-Chem). Interestingly, the Midwest sector difference in the LFT is actually overestimated by the models (5.7 ppm in TM5, 6.2 ppm in GEOS-Chem, vs. 4.4 ppm in the observations). This is most easily explained by the models transporting too much of the cold sector BL depletion signal into the LFT.

These characteristics resemble the growing-season average vertical profiles of the 381 inversions used in Stephens et al. (2007), in which model underestimation of the vertical gradient 382 between 1000 m and 4000 m was used to infer model overestimation of vertical mixing. A similar 383 inference can be made here using BL – LFT (-8.8 ppm observations vs. -3.6 ppm TM5 and -3.2 384 ppm GEOS-Chem). The reduced observation-relative biases in the UFT vs. the LFT suggest that 385 the LFT communicates more via vertical mixing with the BL than with the UFT. We hypothesize 386 that the enhanced vertical mixing is mainly a result overestimated boundary layer mixing in the 387 models, which would not be expected to exceed 3000 m AGL very frequently. 388

While the bias of the overall vertical differences relative to the observations are quite large, there is little transport model dependence to this bias. The systematic differences between TM5 and GEOS-Chem transport members in the Midwest cold sectors are even less than they are in the Midwest warm sectors, and are much smaller than the corresponding observational-relative biases. From Table 2, the biases in the BL are -3.6 ppm for TM5 and -3.3 for ppm for GEOS-Chem.

394 Stephens et al. (2007) went on to show that larger vertical gradients for their seasonal 395 inversion profiles were correlated with decreased magnitudes of net surface posterior fluxes. But that doesn't seem to be the case here. Among the individual inversions, BL mole fractions do not 396 seem to be strongly constrained. The variation of the vertical difference magnitude is large, 397 ranging approximately that of the observations to near zero, and sometimes of opposite sign to the 398 observations – a feature that would be hard to explain solely by vertical mixing differences. 399 Furthermore, while one might expect vertical mixing to be mainly a function of transport model, 400 little dependence of BL – LFT mole fractions on transport model is apparent. 401

402

403 3.3 South (16 Aug - 31 Aug 2016)

In the South warm sectors, there is even less vertical structure in the observed mole fraction profiles above 1500 m AGL than in the Midwest warm sectors (Fig. 5a). Below 1500 m AGL, the clearly observed tendency is for  $CO_2$  mole fractions to be greater than in the free troposphere, by 2-3 ppm. This low-level warm sector enhancement was noted and discussed by Pal et al. (2020), and suggests that a net upwind source of  $CO_2$ .

The inversion posterior  $CO_2$  mole fraction profiles for the South warm sectors also show very little vertical variability with height in the mid- and upper-troposphere, with biases that are extremely small in magnitude (< 2 ppm even for the worst-case model above 2 km AGL, and for the median profile on the order of tenths of a ppm at most). But there does seem to be a tendency in Table 2 for the TM5 models to be about 0.3 ppm greater in upper level mole fraction than the GEOS-Chem models, which while small is actually greater than the overall model bias. Nearer the surface, many of the models reproduce but underestimate the enhancement of  $CO_2$  (Fig. 6a).

The number of cold sector profiles in the South was the least numerous of any region, and so the composite profile is somewhat noisy and difficult to interpret (Fig. 5b). Both observed variability and model spread are very small (1 ppm or less) above 3000 m AGL, but extremely
large (4-5 ppm) below 3000 m AGL (Fig. 6b). While the median model mole fraction below 1500
m AGL is about 2 ppm greater than the observations, the observed profile lies well within the total
model spread (Fig. 6b), and the TM5 and GEOS-Chem biases are within 0.5 ppm of each other
(Table 2). The spread of the GEOS-Chem models is substantially larger than for the TM5 models
(see Fig. 5b).

424



Figure 5: Same as Figure 1, but for the South region (16-31 Aug 2016). Statistics generated from
31 total warm sector profiles and 11 total cold sector profiles.





| 12 | Λ |
|----|---|
| 45 | 4 |

| 435 | Figure 6: | Same as Figure 2, | but for the South | n region (15-31 | Aug 2016). |
|-----|-----------|-------------------|-------------------|-----------------|------------|
|-----|-----------|-------------------|-------------------|-----------------|------------|

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The observed BL sector difference in South (Table 2, 7.1 ppm) is underestimated by both 438 439 the TM5 and GEOS-Chem models (4 ppm). As in the Midwest, both sets of transport models are biased negative in the BL warm sector by about 1 ppm (-0.8 ppm in TM5, -1.3 ppm in GEOS-440 Chem), so the larger contribution to the underestimated BL sector gradient is the positive model 441 442 bias in the cold sector (2.3 ppm in TM5, 1.8 ppm in GEOS-Chem). The magnitude of the observed warm sector vertical difference (+2.0 ppm) is underestimated in both model means (+0.8 ppm for 443 the TM5 models and +0.7 ppm in GEOS-Chem). As in other regions, individual inversion profiles 444 can be very different from the mean, or median, profiles, with some warm sectors even showing 445 low-level depletion. 446

447 In the South cold sectors, as was the case in the Midwest, the variation of BL mole fractions and vertical gradients among individual inversions is very large (Fig. 5b), and the median 448 interguantile spread at low levels is on the order of 7 ppm (Fig. 6b). Like the Midwest, the overall 449 median inversion bias for cold sector BL is positive (about +2.0 ppm), although considerably less 450 than the spread among inversions; also like the Midwest, the dependence of this bias on transport 451 model is considerable less than the overall bias itself (i.e., BL cold sector biases of +2.3 ppm for 452 TM5, +1.8 ppm for GEOS-Chem in Table 2). Meanwhile, for the cold sector LFT, inversion biases 453 are virtually non-existent (0.0 ppm for TM5; +0.1 ppm for GEOS-Chem). Though overall the 454 inversion cold sector LFT is unbiased, the observed profile in Fig. 5b shows a sharp drop in mole 455 fraction as one descends from 2000 m to 1500 m AGL that is not present in any of the inversions. 456 As in the Midwest, this suggests that the inversions are overestimating vertical mixing at least to 457 the 2000 m AGL level. 458

459

### 460 **4 Discussion**

As documented in Pal et al. (2020), the ACT observations from summer 2016 show BL CO<sub>2</sub> mole fractions are enhanced in warm sectors and depleted in cold sectors throughout all regions. We note that our values for these enhancements / depletions differ from, and in the warm sector case are substantially less than, the values in Pal et al. (2020). This is in part because they included horizontal transects in their analysis, and in part because in their vertical analysis they used boundary layer height rather than 1500 m AGL to define their lowest layer.

Except for the warm sectors of the Midwest and South, the spread among individual inversions is quite large compared to the median inversion biases, especially near the surface. (For Midwest cold sectors, the near-surface model spread is large, but the positive model bias is even larger.) The low-level observational variability is also much less than the total inversion model spread. Thus, in these cases ACT-America observations can serve to select out particular inversion members that have more realistic profiles than the others.

Errors in modeled regional CO<sub>2</sub> mole fraction can be attributed to errors in regional surface fluxes, errors in transport (vertical and horizontal), and/or errors in the regional upstream boundary conditions (e.g., those of the inflow marine air masses to the North American continent). The UFT mole fractions may be used as a proxy for the continental upstream boundary conditions, under the assumption that those boundary conditions are vertically well-mixed, and that the UFT has been little affected by the regional flux signal. These assumptions are consistent with the aircraft climatology from Sweeney et al., (2015) that shows vertically homogeneous summer  $CO_2$  profiles at the west and Gulf coasts, the boundary conditions for most of the air we are sampling, and relatively small mole fraction differences (a few ppm in the BL) between the Gulf and west coast air masses. Our measurements show the Gulf background at least is 401-402 ppm.

Large inversion-to-inversion differences in the posterior CO<sub>2</sub> profiles, with little 483 correlation to atmospheric transport model, suggest that the differences of the regional posterior 484 fluxes among the inversions must be quite large. For cases where the profile model spread near 485 the surface is large (i.e., over 5 ppm), the corresponding UFT model spread is far less (no more 486 than 2 ppm). This (2 ppm) is also comparable in magnitude to UFT bias of the median model 487 relative to the observations. It is clear that the magnitude of the variability among the inversion 488 profiles is too large to be due to variability in their upstream boundary conditions. This variability 489 is also too large to be attributed solely to transport model differences (TM5 / GEOS-Chem 490 differences are 3-4 ppm in the MidAtlantic and even less elsewhere). Therefore, only large 491 variation in the regional surface posterior fluxes among the inversions can explain the profile 492 variability. 493

The systematic tendencies in the Midwest and South suggest an overestimate in vertical 494 mixing that is common for both the TM5 and GEOS-Chem transport models. These two regions 495 496 are observed to have enhanced BL mole fractions relative to the LFT in warm sectors, and depleted BL mole fractions in model cold sectors; the models reproduce this tendency, but underestimate 497 the magnitude of the BL / LFT vertical difference. The corresponding BL sectoral differences are 498 underestimated relative to the observations, while the LFT sectoral difference is either unbiased, 499 or overestimated. This suggests that model overestimation of vertical mixing (in particular for the 500 cold sectors) likely contributes to the underestimation of the BL sectoral difference. While it is 501 possible that the inverse flux estimates are biased in such a way as to cause underestimation of the 502 BL sectoral differences (e.g., sinks in the cold sector and sources in the warm sector both 503 underestimated), one would then have to explain how the Midwest cold sector LFT could have 504 biases of the opposite sign as the BL. 505

The systematic error characteristics of the MidAtlantic are quite different from those of the 506 other regions. While in the Mid-Atlantic some enhancement of near-surface CO<sub>2</sub> in warm sectors 507 can be seen in the observations (near 750 m AGL in Fig. 1a), the vertical profiles of warm and 508 cold sectors are more similar to each other than they are in the other regions. For the models, the 509 warm and cold sector profiles are even more similar to each other - in fact, for both TM5 and 510 GEOS-Chem median models in Table 2, LFT warm sectors actually have lower mole fractions 511 than cold sectors. As noted previously, contrary to other regions the GEOS-Chem - TM5 512 differences in the MidAtlantic are consistently positive and significant (2-4 ppm in the BL and 513 LFT; 1-2 ppm in the UFT) and exceed the median model sectoral differences, Consequently, the 514 TM5 models have large negative mole fraction biases at all levels in warm sectors, while the 515 GEOS-Chem models have large positive mole fraction biases at all levels in cold sectors. 516

Also unlike the other regions, in the MidAtlantic the model BL - LFT vertical differences are relatively unbiased, even when the mole fractions in the BL and LFT separately are biased. This does not necessarily mean that the model vertical mixing is correct. It does suggest that the upstream source of the model biases seen in the MidAtlantic has had time to vertically mix up to at least the LFT. This implies that the source of the error is either in the upstream boundary conditions, or in regional fluxes far enough upstream to have a MidAtlantic LFT signal. North American uptake signals that progressively increase west-to-east have been noted previously (Sweeney et al. 2015; Lan et al. 2017; Chen et al. 2019).

For the MidAtlantic warm sectors, the UFT mole fraction biases are of the same sign as 525 those of the LFT, but reduced in magnitude, especially for the TM5 models. But for GEOS-Chem 526 a large part of the biases could be due to errors in the background. For TM5, the value of LFT – 527 UFT is much more negative than that of either GEOS-Chem or the observations (-4.3 ppm vs. -3.1 528 ppm GEOS-Chem, -2.9 ppm observations), which leads to TM5 having BL - UFT much more 529 negative as well (-6.5 ppm vs. -4.9 ppm GEOS-Chem, -4.3 ppm observations). This may seem to 530 contradict the results in Schuh et al. (2019) that suggested TM5 had more vertical mixing than 531 GEOS-Chem. However, if little local vertical mixing happens between the LFT and UFT in either 532 models or observations, then increased vertical gradients between them imply little further about 533 local vertical mixing. Rather, they would suggest in the MidAtlantic warm sectors that TM5 has 534 too much upstream vertical mixing between the LFT and BL near the region of upstream surface 535 536 uptake.

For the MidAtlantic cold sectors, the positive GEOS-Chem biases are again quite 537 consistent with height up to the UFT, suggesting most of the (large) biases are in the background. 538 TM5 has reduced mole fraction biases, but again has more negative LFT - UFT and BL – UFT 539 than GEOS-Chem or the aircraft (LFT - UFT: -3.0 ppm for TM5 vs. -2.3 ppm for GEOS-Chem 540 541 and -1.8 ppm for observations; BL – UFT: -7.2 ppm vs. -5.0 ppm and -4.9 ppm). We would again argue that the TM5 vs. GEOS-Chem vertical gradient differences reflect more upstream vertical 542 mixing of a surface uptake signal to the LFT in TM5. The observational vertical gradient might 543 suggest the vertical mixing of GEOS-Chem is more realistic, but with much large positive 544 background bias. But it should also be noted that the observational variability of MidAtlantic cold 545 sectors in the UFT and higher is large, with different layers ranging from 398 – 401 ppm. If the 546 547 'true' upper level background were 401 ppm, one could well conclude that TM5 has the more realistic vertical mixing to go along with a large negative background bias. 548

549 To better understand the differences between regions, we also did backward trajectories using the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model 550 (Stein et al., 2015; Rolph et al., 2017) from the location of soundings from each combination of 551 sector and region. This is similar to the procedure of Pal et al. (2020), except that we took the 552 trajectories back to 120 hours, and for each sounding location we start backward trajectories at 553 1000 m, 2000 m, and 4000 m AGL, to represent the BL, LFT, and UFT layers. To minimize the 554 sampling uncertainty associated with initiating a single backwards trajectory, at each sounding 555 location we initiate a 3 x 3 matrix of backwards trajectories for a 0.2 x 0.2 degree square centered 556 at the sounding location. In the Supplement, we show representative trajectories from each region 557 / sector combination. 558

Though considerable variability exists among back-trajectories within each region, in general for the Midwest and South warm sectors they originate from the Gulf of Mexico at all levels, as might be expected from their vertical homogeneity. For the cold sectors of these regions, the back-trajectories tend to derive from the north or northwest at low levels, and from southwesterly to northwesterly directions at higher levels. For the MidAtlantic, warm sector backtrajectories, we found parcels from about 1000 m AGL tend to derive from the U.S. Southeast and Gulf Coast, or the Atlantic; at higher levels, parcels derive from more westerly directions, the 566 Appalachians and upper Midwest (Ohio and Mississippi River Valleys). The MidAtlantic cold 567 sector back trajectories are generally from the west or northwest at all levels.

The relatively small MidAtlantic airmass sectoral differences (at least above 1000 m AGL) 568 are plausibly a consequence of the relatively similar origins of the back-trajectories for warm sector 569 and cold sector air. The LFT back-trajectories indicate that for both warm and cold sectors the 570 origin is in the eastern Midwest (Mississippi River Valley), a region with evidence of large 571 growing season uptake (Miles et al., 2012; Lokupitiya et al., 2016). So one hypothesis for the 572 biases in the MidAtlantic is that they are driven by either biases in this source region's uptake flux 573 or biases in the transport of this uptake signal. By contrast, warm sector and cold sector air masses 574 in the other regions may be little affected by upstream sources (e.g., they arrive from the Gulf, the 575 Pacific, or the sparsely-vegetated U.S. West), so their BL and LFT fluxes are being largely driven 576 by flux signals from the immediate vicinity. 577

The observations suggest the TM5 representation of how the MidAtlantic cold sectors 578 become depleted in CO<sub>2</sub> is better which may indicate that the TM5 representation of upstream 579 vertical mixing is more realistic. However, the constancy of the cold sector GEOS-Chem positive 580 bias with height suggests that the GEOS-Chem errors are not so much due to vertical mixing errors, 581 but to not properly representing the general flow from the Midwest surface sinks to the 582 MidAtlantic. Similarly, the large TM5 negative biases for the warm sectors could derive from 583 improper advection of the Midwest depletions in these scenarios, especially near the surface where 584 we found the general flow was from the Gulf Coast states. But it may also be that the shallow 585 586 nature of many MidAtlantic warm sectors BLs may make them particularly difficult for inversions with too much vertical mixing to represent. More definite assessment of the skill of transport 587 model vertical mixing may require more disentangling of model vertical and horizontal transport 588 589 errors.

Horizontal transport dependence could be most important in the MidAtlantic because of 590 the relatively small differences between warm and cold sector trajectories, which make it difficult 591 to properly represent low-level air streams in inversions with the resolution of global models (i.e., 592 1 deg x 1 deg for CT-NRT, and coarser for the other inversions), as well as topography (the 593 Appalachians) not found in other regions. But sharp boundaries between air masses may still exist 594 in these scenarios, such that small horizontal transport errors may lead to substantial mole fraction 595 errors. Experiments using inversions with a common horizontal resolution might help to reduce 596 some of the horizontal transport differences. 597

Another outstanding issue, if increased TM5 vertical mixing relative to GEOS-Chem is in 598 599 fact responsible for its larger LFT  $CO_2$  depletion in the MidAtlantic, is why in the Midwest no transport-dependent vertical mixing differences were apparent. Possibly vertical mixing for the 600 source regions of MidAtlantic and Midwest surface uptake just has different characteristics. Or it 601 602 could be that local boundary-layer mixing in the Midwest is overestimated by all models, but downstream vertical mixing (e.g., by convection) may be transport-model-dependent. Even if 603 boundary layer mixing were in fact transport-model dependent, the fact that the posterior fluxes 604 are not consistent across all transport members could act to suppress clear transport-dependent 605 signals in inversion vertical mixing. A more controlled experiment such as that in Schuh et al. 606 (2019), where only transport model, or surface flux, is varied would lead to more understanding 607 of inversion biases for the ACT-America cases. Current ongoing research, including direct 608 comparison of the simulated vs. observed ACT boundary layer heights, and more controlled 609

modeling experiments such as more rigorous transport model comparison tests with  $SF_{6}$ , will hopefully provide clarification to the realism of vertical mixing in the transport models.

In the warm sectors, BL enhancements of CO<sub>2</sub> were observed in all regions, corresponding 612 to trajectories from the Gulf Coast. (In the MidAtlantic, enhancements were seen in the lowest 613 levels of the warm sectors, possessing Gulf Coast origins; at slightly higher levels, depleted mole 614 fractions were present in warm sectors for trajectories that had a more Midwestern origin.) The 615 models reproduced the enhancement to some degree, suggesting they had local positive net CO<sub>2</sub> 616 fluxes, but generally underestimated it. By contrast, Schuh et al. (2019) reported that these OCO-617 2 inversions had negative mean net  $CO_2$  fluxes between the equator and 45 N during Aug 2016. 618 However, they also found the fluxes equatorward of 45 N to be substantially reduced from those 619 poleward of 45 N at this time, and the poleward of 45 N seasonal net uptake was rapidly declining 620 (see also Crowell et al., 2019). It is possible that by the time of summer 2016 campaign, the zone 621 of maximum net surface uptake had moved northward from the Gulf Coast states, leading to higher 622 average mole fractions in the northward-moving warm sectors than the southward-moving cold 623 sectors over the ACT focus regions. Ongoing analysis of the last ACT-America campaign from 624 July 2019 should help to address this question. 625

626

### 627 **5 Conclusions**

We have shown that the differences in inversion methodology and/or prior fluxes, as 628 opposed to choice of transport model, are the dominant source of variability among the version 7 629 OCO-2 MIP in situ inversion systems analyzed here. In addition, we have shown that the ACT 630 campaign aircraft observations are clearly able to identify biases, and thus are a valuable tool for 631 testing flux inversions. We have found, after segregating models and observations by altitude and 632 airmass within synoptic weather systems, that biases in posterior CO<sub>2</sub> among the individual 633 inversions can be very large compared with the standard error of the observations, suggesting that 634 these biases are significant. Thus, the posterior CO<sub>2</sub> fluxes from these inversions are also likely 635 systematically biased by large amounts. 636

The median of the OCO-2 MIP4 *in situ* inversions, while containing some systematic biases, captures the observed  $CO_2$  profile characteristics in different regions and sectors fairly well. This analysis suggests the advantage of using central estimates from a suite of models such as the OCO-2 MIP, as opposed to any single member.

The significant systematic biases in the model median mean cross-frontal and LFT-ABL posterior CO<sub>2</sub> differences in the Midwest and South suggest some caution in using the central estimates. These biases are likely a combination of underestimate of cold sector net uptake, warm sector net source, and an overestimate of BL-LFT vertical mixing in the U.S. midcontinent. These biases do not appear to be related to the differences in the TM5 vs. GEOS-Chem atmospheric transport.

647 Significant differences between TM5 vs. GEOS-Chem based inversions in the MidAtlantic 648 point toward the importance of deeper tropospheric mixing farther downwind in the continent. 649 They suggest that TM5 has greater vertical mixing of the upstream depletion signal reflected in 650 much of the MidAtlantic cold sector profile, and may suggest that TM5 models this process more 651 realistically. These conclusions are similar to Schuh et al., (2019), who found more limited vertical 652 mixing vs. horizontal mixing in GEOS-Chem relative to TM5. However, TM5 model errors are 653 much larger than those of GEOS-Chem for MidAtlantic warm sectors, possibly due to horizontal 654 transport errors or excessive local TM5 mixing near the surface in these situations. Biases in 655 inversions based on both atmospheric transport models make it difficult to reach a definitive 656 conclusion.

Because members of the version 7 OCO-2 MIP suite consist of different permutations of 657 prior fluxes and transport models, clear attribution of these systematic differences is not possible. 658 Experiments that can more fully diagnose the origin of the posterior CO<sub>2</sub> biases and provide insight 659 into which inversions provide more realistic posterior CO<sub>2</sub> flux estimates are needed. This can be 660 done with controlled experiments using common surface fluxes but different transport models (e.g. 661 Diaz-Isaac et al, 2014; 2018; Chen et al., 2019; Schuh et al., 2019), and common atmospheric 662 transport but different fluxes (e.g. Feng et al., 2019). Further investigation about the cause of 663 divergence among fluxes can be achieved with comparisons of aircraft CO<sub>2</sub> to inversions that use 664 the same atmospheric CO<sub>2</sub> observations but different inversion methods, or the same inversion 665 methods but different observations – including of course the use of OCO-2 XCO<sub>2</sub> retrievals, a key 666 component of the OCO-2 MIP itself (Crowell et al., 2019). Efforts to expand ACT model-data 667 comparisons in all of these directions, and to include evaluation of atmospheric winds and BL 668 depth (e.g. Diaz-Isaac et al., 2018), are underway. Simulations (Feng et al., 2019) and 669 observational methods (Baier et al., 2019) that can differentiate the origins of continental CO<sub>2</sub> 670 provide yet another tool for diagnosing model performance. Finally, comparisons that span all 671 four seasons will be needed to gain insight into annual flux inversion results. These experiments 672 are all underway as part of the analysis of the ACT-America flight campaigns. 673

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