Development and evaluation of CO_2 transport in MPAS-A v6.3

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Abstract.

Chemistry transport models (CTM) play an important role in understanding fluxes and atmospheric distribution of carbon dioxide (CO_2). They have been widely used for modeling CO_2 transport through forward simulations and inferring fluxes through inversion systems. With the increasing availability of high resolution observations, it has been become possible to estimate CO_2 fluxes at higher spatial resolution. ~~However the computational cost of high resolution global model simulation is so high that only major research and operation centers can afford it. In this paper,~~ In this work we implemented CO_2 transport in Model Prediction Across Scales-Atmosphere (MPAS-A). The objective is to use the variable-resolution capability of MPAS-A to enable high resolution CO_2 simulation at limited region with a global model. Treating CO_2 as an inert tracer, we implemented in MPAS-A (v6.3) the CO_2 transport processes, including advection, vertical mixing by boundary layer scheme, and convective transport. We first evaluated the newly implemented model~~by running two sets of simulations over a 60-15 km variable-resolution global domain. The first set of simulations covers four Atmospheric Carbon and Transport-America (ACT-America) aircraft campaign seasons (2016-2018), and the simulated is evaluated using the extensive airborne measurements from ACT~~'s tracer mass conservation and then its CO_2 simulation accuracy. A one-year (2014) MPAS-A simulation is evaluated at the global scale using CO_2 measurements from 50 near-surface stations and 18 Total Carbon Column Observing Network (TCCON) stations. The simulation ~~accuracy~~ is also compared with ~~a 27-km resolution WRF-Chem simulation and CarbonTracker (~~two global models: National Oceanic and Atmospheric Administration (NOAA) CarbonTracker v2019 ~~) covering the same time periods. The second set of simulations covers the month of January and July of 2014, and the results are evaluated using near-surface hourly measurements from 50 surface and tower sites across the globe. This simulation accuracy is compared with~~ (CT2019) and European Center for Medium-Range Weather Forecasts (ECMWF) Integrated Forecasting System (IFS)~~global simulation conducted during the same period. Overall, the evaluation using aircraft measurements indicates that MPAS transport model~~ . A second set of simulation (2016-2018) is used to evaluate MPAS-A at regional scale using Atmospheric Carbon and Transport-America (ACT-America) aircraft CO_2 measurements over the eastern United States. This simulation is also compared with CT2019 and a 27-km WRF-Chem simulation. The global scale evaluations show that MPAS-A is capable of representing the ~~observed atmospheric~~ spatial and temporal CO_2 variation with comparable level of accuracy as IFS of similar horizontal resolution. The regional scale evaluations show that MPAS-A is

capable of representing the observed atmospheric CO_2 spatial structures related with the mid-latitude synoptic weather system, including the warm ~~/~~versus cold sector distinction, boundary layer to free troposphere difference, and ~~enhancements along frontal boundaries. The evaluation using hourly measurements shows that the MPAS transport model is capable of achieving a same level of accuracy as the IFS 80-km resolution simulation~~frontal boundary CO_2 enhancement. MPAS-A's performance in representing these CO_2 spatial structures are comparable with the global model CT2019 and regional model WRF-Chem.

1 Introduction

Carbon dioxide (CO_2) is the most important greenhouse gas, and our knowledge about its sources and sinks still have large gaps. Inversion systems are ~~often used to infer~~ tools for inferring surface CO_2 fluxes based on observations and chemistry transport models (CTM). Two types of CTMs are commonly used: global models and regional models. Global models are commonly used for inferring CO_2 fluxes at coarse spatial ~~scale~~ scales (Patra et al., 2008; Schuh et al., 2019; Jacobson et al., 2007, 2020). With the fast increasing number of atmospheric CO_2 observations, including those acquired by ground based, airborne, and satellite instruments, regional inversion system have been developed and applied to estimate carbon fluxes at higher resolution (Gerbig et al., 2009; Pillai et al., 2012; Lauvaux et al., 2012; Hu et al., 2019; Zheng et al., 2018, 2019).

A major challenge of atmospheric CO_2 inversion modeling is how to partition the model-data mismatch (MDM) among the transport model error, observation error, and prior flux error (Baker et al., 2006). In the Bayesian inversion framework, the error covariance matrix $\mathbf{R}$ is commonly used to represent the combined error of transport model and observations. While it is important to correctly represent the transport model error in an inversion system, it is also important to reduce the error in order to estimate the fluxes with less uncertainty. One approach to reduce the transport model error is to increase ~~simulation resolution~~ the horizontal resolution of a simulation. For instance, Feng et al. (2016) found high-resolution WRF-Chem simulation improved CO_2 model-data comparison because of better resolved planetary boundary layer (PBL) and better representation of spatial variability of CO_2 fluxes. In a recent study, Agusti-Panareda et al. (2019) investigated the impacts of transport model's horizontal resolutions on simulated CO_2 accuracy, and they found that CO_2 variability are generally better represented by higher resolution simulations.

Global high resolution CO_2 simulations require large computational resources~~which may not be affordable outside major research and operational centers~~. Regional (limited area) models, which ~~circumvent the high computational cost of global models~~have lower computational cost than their global model counterpart at the same horizontal resolution, are often used for high resolution CO_2 transport (Feng et al., 2016; Diaz-Isaac et al., 2019, 2018) and inverse modeling (Sarrat et al., 2007; Gerbig et al., 2008; Lauvaux et al., 2012; Zheng et al., 2019). However a regional model requires CO_2 transported from outside its model domain to be prescribed. For a CO_2 inversion system, having lateral boundaries increase the size of the control vector to be optimized (Rayner et al., 2019). A number of approaches have been applied to the CO_2 lateral boundary problem, such as assuming the boundary inflow is perfectly known (Gockede et al., 2010), correcting the lateral boundary condition using

observation prior to inversion (Lauvaux et al., 2012; Schuh et al., 2013), or jointly optimizing flux and lateral boundary condition (Zheng et al., 2018). When CO_2 lateral boundary is optimized, an inversion system adjusts its CO_2 fields at the boundary prescribed by a parent global model in addition to adjusting surface fluxes. This could be problematic for inversion systems that use satellite derived column averaged CO_2 measurements (XCO_2) because model-data mismatches in the free troposphere (FT) are often originated from outside a regional model's limited area domain (Feng et al., 2019; Lauvaux and Davis, 2014).

The objective of the present paper is to provide an alternative high-resolution CO_2 transport modeling approach to regional transport models. This approach is to use a global variable-resolution model which allows for local grid refinement that enables high resolution simulation over an interested region without incurring the prohibitively high computational cost or the lateral boundary condition. Variable-resolution through local grid refinement has been widely used in Numerical Weather Prediction (NWP) models, such as ~~MPAS~~ MPAS-A (Skamarock et al., 2012), Ocean-Land-Atmosphere Model (OLAM) (Walko and Avissar, 2008a, b), Energy Exascale Earth System Model (E3SM) (Golaz et al., 2019), and Finite-Volume Cubed-Sphere model(FV3) (Putman and Lin, 2007). One benefit of local mesh refinement is enabling regional high-resolution modeling without incurring the lateral boundary condition and its associated problems, such as solution mismatches between the driving global model and the evolving regional model (Davies, 2014).

Model Prediction Across Scales-Atmosphere (MPAS-A) is a fully compressible non-hydrostatic global atmospheric model which uses finite-volume numeric solver discretized on centroidal Voronoi mesh with C-grid staggering of its prognostic variables (Skamarock et al., 2012; Thuburn, 2007; Ringler et al., 2010). The centroidal Voronoi mesh allows for local refinement and variable-resolution horizontal mesh which can be gradually changed from coarse to fine resolutions (Skamarock et al., 2012; Ringler et al., 2008).

To enable CO_2 transport modeling, we implemented ~~in MPAS-A (v6.3) the~~ atmospheric CO_2 transport processes, including advection, vertical mixing by ~~Planetary Boundary Layer (PBL)~~ PBL scheme, and convective transport ~~.~~ in MPAS-A v6.3. Because the CO_2 transport processes are fully integrated into the model's meteorological time steps, the resulting MPAS-A CO_2 is an online CTM. We used the newly developed model to conduct two sets of simulations over a 60-15 km variable-resolution global domain. Then the simulation results are evaluated using an extensive set of airborne observations over the eastern United States and near-surface observations from surface and tower stations across the globe. The simulation accuracy of ~~MPAS~~ MPAS-A is compared with three established CO_2 modeling systems based on the same observational data: WRF-Chem (Skamarock et al., 2008; Feng et al., 2019), Carbontracker (v2019, CT2019 hereafter) (Jacobson et al., 2020), and ECWMF IFS (Agusti-Panareda et al., 2014, 2019).

~~The remainder of the paper is organized as follows. Section 2 details the implementation of transport in MPAS, Section 3 describes MPAS simulation experiments and evaluation using airborne and near-surface observations, and comparison with WRF-Chem, CT2019, and IFS. Section 4 provides a summary of the model development and evaluations.~~

2 Implementation of CO_2 transport in ~~MPAS~~MPAS-A

This section describes the major modifications to ~~MPAS~~ MPAS-A that we made to implement CO_2 tracer transport. We represent CO_2 by its dry air mixing ratio (q_{co_2}) and model its atmospheric transport by adding its continuity equation in ~~MPAS~~ MPAS-A following Eq. 7 of Skamarock et al. (2012).

$$\frac{\partial(\tilde{\rho}\, q_{co_2})}{\partial t} = -(\nabla \cdot \tilde{\rho}\, q_{co_2}\, \mathbf{V})_\zeta + F_{bl} + F_{cu} \tag{1}$$

where $\tilde{\rho} = \rho_d/(\partial\zeta/\partial z)$, ρ_d is dry air density, ζ is the vertical coordinate, z is geometric height, t is time, and $\mathbf{V}$ is the velocity vector. The left hand side of the equation is the total CO_2 time tendency ($\partial(\tilde{\rho}\ q_{co_2})/\partial t$), and the ~~three~~ first, second, and third terms on the right hand side represent the contributions from advection, vertical mixing, and convective transport respectively. CO_2 tendency from advection is modeled in flux form (Section 2.1), while tendency from vertical mixing (F_{bl}) and convective transport (F_{cu}) are modeled in uncoupled form ($\partial q_{co_2}/\partial t$) which are coupled to $\tilde{\rho}$ before being added to the total tendency. We choose to implement CO_2 vertical mixing in the Yonsei University (YSU) PBL scheme (Hong et al., 2006), and CO_2 convective transport in Kain-Fritsch (KF) scheme (Kain, 2004) because they are widely used in CTM and have been validated using observations (Borge et al., 2008; Hu et al., 2010; Kretschmer et al., 2012; Polavarapu et al., 2016). Details of the three terms on the right hand side of Eq. 1 are described in the following sections. We note that because the monotonicity constraint in the third-order scalar horizontal advection scheme (Skamarock and Gassmann, 2011) introduces dissipation ~~MPAS~~ MPAS-A does not use any explicit horizontal diffusion for scalar~~, and~~. Accordingly we did not include horizontal diffusion for CO_2~~accordingly~~.

2.1 CO_2 advection

Advection is the most significant component of CO_2 atmospheric transport. Following the example of other scalars in ~~MPAS~~ MPAS-A (Skamarock and Gassmann, 2011), we model CO_2 advection as:

$$(\nabla \cdot \tilde{\rho}\, q_{co_2}\, \mathbf{V})_\zeta = \Big[\frac{\partial(\tilde{\rho} u q_{co_2})}{\partial x} + \frac{\partial(\tilde{\rho} v q_{co_2})}{\partial y}\Big]_\zeta + \frac{\partial(\tilde{\rho} w q_{co_2})}{\partial \zeta} \tag{2}$$

where $\mathbf{V} = (u, v, w)$, and u, v, and w is the zonal, meridional, and vertical wind respectively. The first item on the right hand side enclosed in the square bracket is the CO_2 horizontal flux divergence, and second item is the vertical flux divergence. The horizontal flux divergence is transformed via the divergence theorem into an integral of flux over each control volume, which is modeled as:

$$\Big[\frac{\partial(\tilde{\rho} u q_{co_2})}{\partial x} + \frac{\partial(\tilde{\rho} v q_{co_2})}{\partial y}\Big]_\zeta = \frac{1}{A_i}\sum_e^{n_e} l_e\, F_e(\mathbf{V}_H, \tilde{\rho}\, q_{co_2}) \tag{3}$$

where e indexes the edges of a cell and n_e represents the number of edges the cell has, l_e is the length of an edge, A_i is the cell's areal size, $F_e(\mathbf{v}_H, \tilde{\rho} q_{co_2})$ is the instantaneous horizontal CO_2 flux that crosses the cell edge e, and $\mathbf{V}_H = (u, v)$ is the horizontal wind vector. The details of ~~MPAS~~ MPAS-A instantaneous horizontal flux ~~calculattion~~ calculation can found in Skamarock and Gassmann (2011). The vertical CO_2 flux divergence in Eq. 2 is calculated using finite difference

$$\frac{\partial(\tilde{\rho} w q_{co_2})}{\partial \zeta} = \frac{1}{\Delta \zeta}\left[F(w, \tilde{\rho} q_{co_2})_{k+\frac{1}{2}} - F(w, \tilde{\rho} q_{co_2})_{k-\frac{1}{2}}\right] \tag{4}$$

where $F(w, \tilde{\rho} q_{co_2})$ is the vertical CO_2 flux that crosses a cell's vertical face, and k indexes the vertical coordinate.

2.2 CO_2 vertical mixing

Like in WRF (Skamarock et al., 2008), a ~~planetary boundary layer (PBL) parameterization in MPAS~~ PBL parameterization in MPAS-A treats the vertical mixing of momentum and scalars not only in the boundary layer (BL) but in the entire atmospheric column. ~~Yonsei University (YSU) PBL scheme~~ YSU (Hong et al., 2006) is one of the PBL schemes available in ~~MPAS~~ MPAS-A 6.3. The present YSU scheme treats vertical mixing of momentum, potential temperature, and water species, but not atmospheric tracers. We modified the scheme to treat CO_2 vertical mixing.

In the YSU scheme, after the ~~boudary layer (BL) top~~ top of BL is determined, the vertical mixing of momentum, potential temperature, and water vapor are treated separately: above BL, local K-profile approach (Louis, 1979) ~~)~~ is used for vertical diffusion of momentum and scalars (Noh et al., 2003; Hong et al., 2006). Within BL, an entrainment flux at the inversion layer is included for momentum and scalars diffusion. In addition, a countergradient mixing term is included for the diffusion of momentum and potential temperature to account for the convective-driven mixing (γ_c of Eq. 4 in Hong et al. (2006)), but this term is not used for water vapor.

Following the treatment of water vapor, we parameterize CO_2 vertical mixing in BL as

$$\frac{\partial q_{co_2}}{\partial t} = \frac{\partial}{\partial z}\left[K_h\left(\frac{\partial q_{co_2}}{\partial z}\right) - \overline{(w' q'_{co_2})_h}\left(\frac{z}{h}\right)^3\right] \tag{5}$$

where z is the vertical distance to surface, h is BL top height, K_h is vertical eddy diffusivity. Note that this formulation does not include a countergradient mixing term following the treatment of water vapor in the original YSU (Hong et al., 2006). The second term in the square bracket of Eq. 5 represents the contribution from CO_2 entrainment flux at the inversion layer, which is parameterized as:

$$\overline{(w' q'_{co_2})_h} = w_e \Delta q_{co_2}|_h \tag{6}$$

where $\Delta q_{co_2}|_h$ is the CO_2 mixing ratio difference across the inversion layer, and w_e is the entrainment rate at the inversion layer calculated by Eq. A11 of Hong et al. (2006). Above BL top, vertical mixing of CO_2 is parameterized as:

$$\frac{\partial q_{co_2}}{\partial t} = \frac{\partial}{\partial z}\left[K_h(\frac{\partial q_{co_2}}{\partial z})\right] \tag{7}$$

We use the same value for CO_2 ~~eddy~~ vertical diffusivity as water vapor. The details of K_h calculation can be found in the appendix of Hong et al. (2006), and its value is limited between 0.01 and 1000 m^2s^{-1} to prevent too weak or strong vertical mixing. The term $\partial q_{co_2}/\partial t$ from Eqs. 5 is coupled with dry air density before being applied to the continuity equation (Eq. 1).

2.3 CO_2 convective transport

For convective transport, we modified the Kain-Fritsch scheme (hereafter KF) ~~(?)~~ (Kain, 2004) to include the CO_2 treatment. KF is a mass-flux convection scheme which rearranges mass in an air column using convective updrafts, downdrafts, and environmental mass fluxes. Both the updraft and downdraft entrain from and detrain to the environment, thus altering the vertical profile of an air column's thermodynamic properties. We added the CO_2 convective transport as:

$$\frac{\partial q_{co_2}}{\partial t} = \frac{(M_u + M_d)}{\rho A}\frac{\partial q_{co_2}}{\partial z} + \frac{M_{ud}}{M}(q^u_{co_2} - q_{co_2}) + \frac{M_{dd}}{M}(q^d_{co_2} - q_{co_2}) \tag{8}$$

where q_{co_2}, $q^u_{co_2}$, and $q^d_{co_2}$ are the CO_2 mixing ratio in the environment, updraft, and downdraft respectively, M_u and M_d are the updraft and downdraft mass respectively, ρ is the environment air density, A is the horizontal area of a cell, $M = \rho A \delta z$ is the mass of environmental air in a grid box, and M_{ud} and M_{dd} are the detrainment from the updraft and downdraft respectively.

In KF, the updraft and downdraft mass and the rates for the entrainment and detrainment are determined by a steady-state plume model and a convective available potential energy (CAPE) closure assumption: 90% of the existing CAPE should be removed by the convection parameterization (Kain and Fritsch, 1990; Fritsch and Chappell, 1980; Kain, 2004). Because the calculation of the updraft and downdraft mass fluxes is related to a cell's horizontal area, the KF scheme may behave differently at different areas of MPAS-A's variable-resolution grid. The updraft source layers are determined by a search from the model's lowest vertical level for a group of consecutive layers that is buoyant and at least 50 hPa deep (Kain, 2004). The initial value of CO_2 mixing ratio in the updraft is modeled as a pressure weighted average of the source layers:

$$q^u_{co_2} = \sum_k \frac{\delta q_{co_2,k}\,\delta p_k}{\delta p_k} \tag{9}$$

where δp_k is layer's pressure depth, and $q_{co_2,k}$ is the layer's CO_2 mixing ratio. ~~Carbon dioxide~~ CO_2 mixing ratio of the updraft is modified by the entrainment of the environmental air through its ascent from its starting level to the cloud top.

$$q^u_{co_2} = \frac{q^u_{co_2} M_u + q_{co_2} M_{ue}}{M_u + M_{ue}} \tag{10}$$

where M_{ue} is the updraft entrainment. The initial CO_2 mixing ratio of a downdraft (q^d_{co2}) is the same as that of the environment (q_{co2}) at the downdraft starting level and it is modified by entrainment through the downdraft descent:

$$q^d_{co_2} = \frac{q^d_{co_2} M_d + q_{co_2} M_{de}}{M_d + M_{de}} \tag{11}$$

where M_{de} is the downdraft entrainment.

3 Model evaluation

In this section we evaluate the newly developed ~~MPAS~~ MPAS-A CO_2 transport model through simulation experiments~~using airborne and near-surface observations~~. After describing the simulation configuration (Sect. ~~3.1~~3.1), we assess the model's global mass conservation property (Sect. ~~3.2), then~~ 3.2). Then we evaluate the model~~accuracy by comparing MPAS simulated with high-resolution airborne measurements (Sect. 3.4) and~~ 's CO_2 transport accuracy at the global scale using hourly near-surface ~~hourly measurements (Sect. 3.3). Two sets of MPAS simulations are conducted: the first set simulations covers four ACT campaign seasons (2016-2018) for evaluation~~ CO_2 observations from 50 in situ stations and column-averaged CO_2 dry air mole fraction (XCO_2) measurements from 18 Total Carbon Column Observing Network (TCCON) stations (Sect. 3.3). Finally, we evaluate MPAS-A at the regional scale using high-resolution airborne measurements ~~; the second set covers January and July of 2014 for evaluation using near-surface measurements and comparison with the IFS simulation results reported in Agusti-Panareda et al. (2019)~~from ACT over the eastern United States (Sect. 3.4). MPAS-A CO_2 transport are also compared with three established CTMs: NOAA CT2019 (Jacobson et al., 2020), ECMWF IFS (Agusti-Panareda et al., 2019) , and WRF-Chem (Skamarock et al., 2008). In the following model evaluation, we use root mean square error (RMSE), bias (μ), and random error (STDE) as the model accuracy metrics:

$$\text{RMSE} = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(m_i - o_i)^2} \tag{12}$$

$$\mu = \frac{1}{N}\sum_{i=1}^{N}(m_i - o_i) \tag{13}$$

$$\text{STDE} = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(m_i - o_i - \mu)^2} \tag{14}$$

where o_i and m_i represent the observed and modeled values respectively.

For model-data intercomparison, MPAS-A model data need to be interpolated to the observation space. Following Patra et al. (2008), the model is sampled in the horizontal by taking the nearest cell overland. MPAS-A uses a height-based terrrain-following vertical coordinate (Skamarock et al., 2012). At a given cell, the height of the k^{th} vertical layer boundary is denoted as z_k^h. The height of the layer center is $z_k = 0.5 \times (z_k^h + z_{k+1}^h)$. In MPAS-A, horizontal wind fields are defined at the vertical layer boundaries and CO_2 fields are defined at layer centers. For horizontal wind fields validation using radiosonde data (Sect. 3.3.1), the column profile of air pressure and horizontal wind fields defined at layer boundaries are used to interpolate to the measurements' pressure levels. For comparison with near-surface CO_2 observations from in-situ stations (Sect. 3.3.3) and aircraft observations (Sect. 3.4), model CO_2 defined at layer centers are interpolated to the measurement heights. Vertical interpolation and integration for the comparison with TCCON XCO_2 are described in Sect. 3.3.4. MPAS-A simulation outputs are saved at 1-hour intervals. For comparison with radiosonde observations and near-surface CO_2 observations, no temporal interpolations are applied: observations are paired with the closest hourly MPAS-A output. For comparison with aircraft observations, the hourly model outputs that bracket an observation's time stamp are used for the temporal interpolation.

3.1 Simulation experiment configuration

For all subsequent simulations, ~~MPAS~~ MPAS-A uses a 60-15km variable-resolution global mesh. Fig. **??** shows the cell size (in km^2) of the simulation domain, where the highest resolution (15 km) over North America has cell size smaller than 250 km^2 which gradually increases to about 3,600 km^2 for the rest of the global domain. On the vertical direction, there are 55 levels spanning from surface to 30 km above the mean sea level. Model time step is 90 seconds in accordance with the highest (15km) horizontal resolution. For physical parameterizations, in addition to the modified YSU PBL (Hong et al., 2004) and Kain-Fristch cumulus schemes ~~(?) described in Sect.~~ (Kain, 2004) described in Section 2, we use RRTMG for longwave and shortwave radiation ~~(?)~~(Iacono et al., 2008), Noah land scheme (Chen and Dudhia, 2001), Monin-Obukhov surface layer scheme, and WRF single-moment 6-class microphysics scheme (Hong and Lim, 2006). The third-order accuracy advection is used for all scalars and CO_2 tracer. A summary of the physics parameterizations used in the simulations is given in Table ~~??~~.1.

~~Meteorology initial conditions~~ Initial meteorological fields are generated from the ERA-Interim ~~analysis (Dee et al., 2011).~~ reanalysis (Dee et al., 2011). To keep model meteorological fields close to the ~~analysis, MPAS~~ reanalysis, MPAS-A meteorological fields are re-initialized using the analysis at 00:00 UTC each day throughout a simulation period. ~~Carbon dioxide~~ CO_2 mixing ratio is kept unchanged during the meteorology re-initializations, thus a free-running simulation. This configuration is the same as that used by Agusti-Panareda et al. (2014, 2019) in their IFS global CO_2 simulations. The first CO_2 initial condition for a simulation is from CT2019 $3^\circ \times 2^\circ$ posterior dry mole fraction product and surface CO_2 fluxes are prepared by interpolating the CT2019 3-hourly $1^\circ \times 1^\circ$ posterior flux product (Jacobson et al., 2020). The four ~~fluxes from~~ CT2019 fluxes (biosphere, ocean, fossil fuel, and fire) are interpolated to ~~MPAS~~ MPAS-A model grid and ingested at 3-hour intervals throughout a simulation.

3.2 CO_2 mass conservation

For ~~a transport model~~CTM, it is very important to maintain the global CO_2 mass conservation (Agusti-Panareda et al., 2017; Polavarapu et al., 2016). ~~To evaluate the massconservation property of MPAS, we run a 120-hour continuous simulation and examine the change of total mass of CO_2 in the model domain. The simulation starts at 2016-08-01 00:00 UTC and ends at 2016-08-06 00:00 UTC. It uses the physics parameterizations and domain setup described~~ Because meteorological re-initializations introduce changes in dry air mass, they impact MPAS-A's global CO_2 mass conservation. We first examine MPAS-A's inherent mass conservation property through a simulation without the meteorological re-initializations in Sect. 3.2.1. Then we examine and treat the impacts of the meteorological re-initializations in Sect. ~~3.1. The CO_2 dry air mixing ratio field in the model is initialized using CT2019 dry mole fraction , but fluxes ingestion is turned off. Because the meteorology re-initiation will introduce dry air mass change which in turn will impact the total CO_2 mass, we run the simulation without applying the meteorology re-initialization. The impact of meteorological re-initialization and its treatment are considered in Section ??.~~ 3.2.2.

3.2.1 Mass conservation without meteorology re-initialization

~~Through the 120-hour simulation ,~~ To examine MPAS-A's mass conservation property, we conducted a MPAS-A simulation that lasts from January 1 to December 31 2014. The simulation is initialized with the CT2019 CO_2 mole fraction and is driven with 3-hourly CT2019 surface CO_2 fluxes. Meteorological re-initializations are not applied during the simulation and the model outputs are saved ~~at 1-hour intervals~~ using double-precision. ~~The total mass of dry air (m_{air}) and CO_2 (m_{co_2})~~ MPAS-A's global dry air mass (M_{air}) is then calculated ~~from these hourly outputs using Eqs. 15and 16 respectively.~~

$$m_{air} = \sum_{k}^{L}(\sum_{i}^{N} A_i \, h_{i,k} \, \rho_{i,k})$$

at 00:00 UTC each day through the one-year simulation using Eq. 15,

$$\mathrm{M_{air}} = \sum_{k}^{L}(\sum_{i}^{N} A_i \, h_{i,k} \, \rho_{i,k}) \tag{15}$$

$$m_{co_2} = \sum_{k}^{L}(\sum_{i}^{N} A_i \, h_{i,k} \, \rho_{i,k} \, q_{i,k})$$

~~In Eqs. 15 and 16,~~ where subscript i indexes the horizontal cell, ~~ranging from 1 to N=535,554; subscript j~~ subscript k indexes the vertical level, ~~ranging from 1 to L=55. MPAS hexagon cell base areal size is~~ A_i ~~(cell sizeis constant within a given column at different vertical levels), and the height of the cell is represented by $h_{i,j}$ (heights at different columns of a same~~

~~vertical level maybe different due to the terrain-following vertical coordinate). The volume of a grid box is $V_{i,k} = A_i\, h_{i,k}$, its~~ is cell size, $h_{i,k}$ is cell height, and $\rho_{i,k}$ is dry air density (~~kg/m^3) is $\rho_{i,j}$, and its CO_2 dry air mixing ratio (kg/kg) is $q_{i,j}$.~~

~~The calculation shows that the total volume of the MPAS model domain is $1.5184682921961 \times 10^{19}$ m^3, which is a constant through the simulation because of the model's height based vertical coordinate. At the start of the simulation~~ $kg\,m^{-3}$). After the model's global dry air mass is calculated at 00:00 UTC each day of the simulation period, its variation is quantified as a ratio $E^t_{air} = (M^t_{air} - M^0_{air})/M^0_{air}$, where M^0_{air} and M^t_{air} are the model's global dry air mass at the simulation start (00:00 UTC January 1 2014) and the current time step respectively. The top panel of Fig. 2 shows E^t_{air} at 00:00 UTC of each day through the one-year simulation period. The figure shows that the maximal magnitude of E^t_{air} is less than 4×10^{-12} during the one-year simulation. In comparison, the total dry air mass ~~is $5.053906341880670208 \times 10^{18}$ kg and total~~ of ECMWF IFS increases about 0.01% of its initial value in a 10-day forecast (Diamantakis and Flemming, 2014). Similarly, the Environment and Climate Change Canada (ECCC) Global Environmental Multiscale (GEM) model loses about 0.01% of its initial total dry air mass in a 10-day forecast (Polavarapu et al., 2016). MPAS-A has a significantly lower global dry air mass variation than the two global models because its explicit grid point advection scheme conserves mass (Skamarock and Gassmann, 2011) while the semi-Lagrangian advection scheme used by IFS and GEM does not conserves mass (Williamson, 1990). Thus, no mass fixer (Diamantakis and Flemming, 2014; Polavarapu et al., 2016) is used in MPAS-A.

MPAS-A's global CO_2 mass (M_{co_2}) is calculated using Eq. 16,

$$M_{co_2} = \sum_{k}^{L} (\sum_{i}^{N} A_i\, h_{i,k}\, \rho_{i,k}\, q_{i,k}) \tag{16}$$

where $q_{i,k}$ is CO_2 dry air mixing ratio (kg/kg) and the rest of the terms are the same as in Eq. 15. To assess the global CO_2 mass ~~is $3.079178060337270 \times 10^{15}$ kg. The total mass of dry air and~~ conservation, M_{co_2} calculated using Eq. 16 is adjusted for the CO_2 ~~at each subsequent hour is normalized by their respective starting values and resulting variations are shown in Fig. **??**. The maximal variation of the total dry air mass from its starting value during the 120-hour simulation is $29,660,160$ kg, which when divided by the total air volume represents a variation of mean dry air density approximately 1.95310^{-12} kg/m^3. The maximal variation of~~ mass introduced through the ingestion of the 3-hourly surface CO_2 fluxes. For a 3-hour period, total CO_2 mass ~~from its starting value is $18,980$ kg, which represents a variation in mean~~ introduced through the surface CO_2 fluxes is $\sum_i^N A_i F_i \Delta t$, where F_i is the combined biosphere, ocean, fossil fuel, and fire CO_2 ~~mixing ratio of 1.0416×10^{-15} kg/kg (equivalent to 6.839×10^{-10} ppm) for a mean dry air density of 1.2 kg/m^3.~~

~~In MPAS dry air density and dry air mixing ratioare prognostic variables and their precision is limited by the round-off error of the~~ fluxes ($kg\,m^{-2}\,s^{-1}$) at a surface cell, A_i is the cell's areal size, N is number of surface cell, and Δt=3 hours. After the adjustment, the variation of global mass of CO_2 is quantified as a ratio, $E^t_{co_2} = (M^t_{co_2} - M^0_{co_2})/M^0_{co_2}$, where $M^0_{co_2}$ and M^t_{co2} are the model's ~~Fortran double-precision code. Fig.**??**~~ initial and current time step global CO_2 mass. $E^t_{co_2}$ at 00:00 UTC of each day of the simulation period is shown in the lower panel of Fig. 2. The figure shows that the maximal ~~variation in dry~~

~~air density is 1.95310^{-12} kg/m^3) and maximal variation in dry air mixing ratio is 1.0416×10^{-15} kg/kg during the simulation period. Given that the double precision float number calculation of MPAS Fortran code is capable of having 14 decimal places of precision, we consider the MPAS~~ magnitude of $E^t_{co_2}$ is about 10^{-5}. This is much higher compared to E^t_{air} and it is due to the strong gradients caused by surface CO_2 ~~transport maintains the global mass conservation to the level allowed by the machine precision~~flux which challenge the model's numerical scheme.

3.2.2 CO_2 mass conservation during meteorology re-initialization

~~The last section has demonstrated that MPAS is capable of maintaining global CO_2 mass conservation. However when meteorology~~ When meteorological re-initialization is applied ~~,~~ during a simulation, the values of dry air density ~~fields in the model~~ in MPAS-A are replaced by values from the initialization files generated from the ERA-Interim ~~analysis (Dee et al., 2011)~~ reanalysis. In most cases, this will cause dry air density change which in turn will introduce CO_2 mass change ~~because~~ if CO_2 dry air mixing ratios are kept unchanged during the ~~re-initializations.~~

re-initialization. To assess this possible change in global CO_2 mass, we conducted ~~a 48-day MPAS simulation starting 00:00 UTC 15 July 2016 with meteorology~~ another one-year long MPAS-A simulation identical to that used in Section 3.2.1 except that meteorological re-initialization is applied at 24-hour ~~interval but without CO_2 fluxes ingestion. The~~ intervals during the simulation. The variation of global CO_2 mass ~~is calculated using Eq. 16 at the end of each 24-hour period which is then compared with its initial value at the start of the simulation. The resulting global mass variation is shown in the~~ caused by a meteorological re-initialization is quantified as a ratio $E = (M'_{co_2} - M_{co_2})/M_{co_2}$, where M_{co_2} and M'_{co_2} are the global CO_2 mass before and after a meteorological re-initialization. The top panel of Fig. **??**. ~~The figure shows that during the 48-day simulation, the maximal variation of total~~ 3 shows the value of E at each meteorological re-initialization. The figure indicates that a meteorological re-initialization could cause a change of more than 0.01% of the global CO_2 mass~~can reach approximately $\pm 0.05\%$, which is 7 to 8 magnitudes larger than that without re-initialization (Fig.**??**).~~ .

To restore the CO_2 mass conservation ~~, we calculate the total~~ after a meteorological re-initialization, we adjust MPA-A's CO_2 ~~mass difference caused by a meteorology re-initialization using~~ fields by a spatially uniform scaling factor: $q'_{i,k} = r \times q_{i,k}$, where $q_{i,k}$ and $q'_{i,k}$ are the CO_2 dry air mixing ratio, before and after the adjustment, respectively. The scaling factor r is calculated as,

$$r = \frac{\sum_k^L (\sum_i^N A_i \, h_{i,k} \, \rho_{i,k} \, q_{i,k})}{\sum_k^L (\sum_i^N A_i \, h_{i,k} \, \rho'_{i,k} \, q_{i,k})} \tag{17}$$

where the notations are the same as in Eq. 16 except that $\rho'_{i,k}$ is the dry air density after a meteorology re-initialization and $\rho_{i,k}$ is the value before the re-initialization. ~~Then CO_2 mixing ratio at each grid box is scaled by~~

~~$q'_{i,k} = r \times q_{i,k}$~~

where $q'_{i,k}$ is the mixing ratio that will be used as the initial value for the next 24-hour simulation period.

To test the effectiveness of this scaling method, the 48-day simulation is one-year MPAS-A simulation with meteorological re-initialization was conducted again but with the mixing ratio scaling after each this time with the CO_2 dry air mixing ratio adjustment applied after each meteorological re-initializationusing Eqs. 17 and **??**. . The resulting variation in total CO_2 mass is plotted at in the lower panel of Fig . **??**. 3. The figure shows that maximal variation is less than $\pm 0.001\%$, which is small enough to be acceptable for most the maximal magnitude of the variation caused by a meteorological re-initialization has been reduced from $\sim 10^{-4}$ to $\sim 10^{-6}$ of the global CO_2 transport simulations. We note that an alternative approach mass. Note the different scales in the y-axis used in the top and bottom panels of Fig. 3.

An alternative approach to restore mass conservation is to scale CO_2 mixing ratio at each grid box individually by

$$q'_{i,k} = \frac{\rho_{i,k}}{\rho'_{i,k}} \times q_{i,k} \tag{18}$$

where notation is the same as Eq. 17. This scaling approach can maintain total global CO_2 mass conservation as allowed by machine precision but it will introduce artificial spatial variations in CO_2 mixing ratio. In the simulations in the following sections, we chose to use the first scaling approach to avoid the artificial CO_2 mixing ratio variation by accepting the small change in total changes in global CO_2 mass.

3.3 Model valuation using airborne measurementsevaluation at global scale

In this section, we evaluate the MPAS CO_2 simulation accuracy using an extensive high resolution CO_2 observation data acquired through the Atmospheric Carbon and Transport-America project (ACT-America, henceforth, referred to as ACT). ACT is a National Aeronautics and Space Administration (NASA) Earth Venture Suborbital 2 (EVS-2) mission, and its goal is to improve atmospheric inversion estimates of CO_2 and CH_4 through extensive airborne measurements over the eastern United Stated during multiple seasons (Davis et al., 2018a). Through four campaign seasons from Summer 2016 to Spring 2018 with two research aircraft (C130 and B200), the ACT project has collected an extensive dataset of highly resolved CO_2 measurements in both boundary layer and free troposphere. To use ACT airborne measurements for MPAS-A CO2 transport at the global scale. For the model evaluation, we ran four sets of MPAS simulations each covering one ACT campaign season. The duration of the ACT campaign seasons and the corresponding MPAS simulation periods is given in Table **??**. All simulations use the same domain and configurations MPAS-A was initialized at 00:00 UTC July 1 2013 and ran till December 31 2014. The model configuration for this simulation is as described in Sect. 3.1, and model outputs are saved at 1-hourly interval for subsequent evaluation. 3.1. With the first six-month as model spin-up, we use the one-year simulation of 2014 for the model evaluation. First MPAS-A simulated horizontal wind fields are evaluated using radiosonde measurements from 457 stations. Then the model's CO_2 fields are compared with CT2019, near-surface $CO2_2$ measurements from 50 stations, and XCO2 retrievals from 18 TCCON stations.

3.3.1 Evaluation of ~~modeled~~ horizontal wind fields

Accurate meteorological fields are critical for an accurate CO_2 transport simulation. ~~Michaelis et al. (2019) has assessed MPAS simulated meteorology accuracy using extensive observation data.~~ Before evaluating the simulated CO_2, we first evaluate the ~~MPAS~~ MPAS-A simulated horizontal wind fields considering their importance in CO_2 advection. We compare ~~MPAS~~ MPAS-A simulated horizontal wind fields at ~~00~~12:00 and ~~12~~00:00 UTC each day of the simulation period with raidosonde observations from 457 stations located around the globe at four pressure levels~~(1, 000, :~~1000, 850, 500, 850, and 200 hPa~~).~~ . Note that because of the 24-hourly meteorological re-initialization, the 00:00 and 12:00 UTC simulation results are 12-hour and 24-hour forecasts respectively. The locations of the 457 radiosonde stations are shown in Fig. ~~S3~~ S1 of the supplement~~material. The~~ .

To compare with the similar validation results reported in Agusti-Panareda et al. (2019), the horizontal wind fields evaluation results for ~~each of the four ACT seasons are summarized in Table **??**. For instance, the RMSE of vector wind at the~~ January and July of 2014 are listed in Table **??**. The table shows that while the mean difference in wind direction decreases with altitude, the mean RMSE vector wind generally increases with altitude, which agree with the IFS validation results (Agusti-Panareda et al., 2019). At 1000 ~~, 850, 500, and 200 hPa are 3.31, 3.87, 3.78,~~ hPa level, MPAS-A has a slightly lower accuracy than IFS during the same time period. For instance, MPAS-A's mean RMSE vector wind at 1000 hPa is 3.83 m/s for January 2014, and IFS results range from 3.2 m/s to 3.75 m/s for its 9 km and 80 km horizontal resolution simulations. For July 2014, the mean RMSE vector wind at 1000 hPa is 3.47 m/s from MPAS-A and ~~5.46~~ 3.0 m/s to 3.6 m/s for the IFS 9 km and 80 km simulations. At upper level, MPAS-A has a slightly higher accuracy than IFS: at 500 hPa, MPAS-A mean RMSE vector wind is 3.72 m/s and 3.39 m/s for January and July of 2014 respectively, while IFS results in 4.0-4.1 m/s and 3.5-3.6 m/s for the ~~Summer 2016 season. These values are comparable to but higher than the vector wind RMSE of the month of July~~ same time period.

An important finding of Agusti-Panareda et al. (2019) is that higher horizontal resolution generally lead to higher meteorological and CO_2 simulation accuracy. To examine the influence of horizontal resolution on MPAS-A's meteorological simulation accuracy, we conducted an additional set of simulation using the identical configuration except that it uses a global 60 km uniform-resolution grid instead of the 60-15 km variable-resolution grid (Fig. 1). Out of the 475 radiosonde stations, 131 are located at 15 km cells in the 60-15 km variable-resolution simulation. These 131 radiosonde stations are all located at 60 km cells in the 60 km uniform-resolution simulation. In Table 3, we calculated and compared horizontal wind accuracy at these 131 radiosonde stations between the 60 km uniform-resolution simulation (labeled as 60 km) and the 60-15 km variable-resolution simulation (labeled as 15 km). The table shows that the horizontal wind fields at these 131 stations are simulated with considerably higher accuracy on the 15 km grid than its 60 km grid counterpart. For instance at 1000 hPa, the mean RMSE wind vector for January 2014 ~~from the IFS 9km and 80km horizontal resolution global simulations reported in Agusti-Panareda et al. (2019). For the winter 2017 season, RMSE of vector wind is 3.70, 4.44, 4.56, and 5.40~~ is 3.46 m/~~s respectively~~s and 3.98 m/s at the 15 km and 60 km grids respectively. The values are 3.10 m/s and 3.64 m/s for July 2014.

Table 3 also shows that the difference in the mean RMSE wind vector between the 15 km and 60 km grids is larger near the surface at 850 and 1000 hPa than in the middle and upper troposphere (500 and 200 hPa), which is consistent with the findings of Agusti-Panareda et al. (2019). For both January and July at the four pressure levels, the mean RMSE wind vector at the 131 radiosonde stations at MPAS-A's 15 km grid is either similar to or slightly lower than the mean RMSE wind vector of the around 400 stations from the IFS 9 km resolution simulation (Agusti-Panareda et al., 2019).

3.3.2 Comparison of CO_2 fields with CarbonTracker

Having established that the horizontal wind fields simulated by MPAS-A are sufficiently accurate, the CO_2 fields can be evaluated. First we directly compare XCO_2 from MPAS-A and CT2019 field at the grid scale. First, XCO_2 are calculated at the native grid for MPAS-A (60-15km) and CT2019 ($3^\circ \times 2^\circ$). XCO_2 at a given model cell is calculated as the pressure weighted CO_2 dry air mixing ratio.

$$XCO_2 = (\sum_{k=1}^{N} p_k q_k^{co_2})/(\sum_{k=1}^{N} p_k) \tag{19}$$

where p_k is modeled air pressure at layer k corrected for water vapor, $q_k^{co_2}$ is CO_2 dry air mole fraction at the same level. N is the number of vertical levels in a model. Then, XCO_2 from MPAS-A and CT2019 are regridded from their respective grids an identical $1 \times 1^\circ$ grid for a direct comparison. Figure 4 shows the comparison of XCO_2 from MPAS-A (top) and CT2019 (middle) and their difference (bottom) for July 1 and December 1 2014 at ~~the four pressure levels,~~ 00:00 UTC. The figure shows that XCO_2 from MPAS-A and CT2019 are generally consistent at the large scales, but differences exist at small spatial scales. The higher horizontal resolution of MPAS-A is evident particularly in July over the northeast and southern China. In December, MPAS-A has higher XCO_2 than CT2019 within the Arctic Circle and southern China. Overall the differences between MPAS-A and ~~they are slightly higher than the~~ CT2019 are evident. The magnitude of differences are mostly within 3 ppm, which is similar to the magnitude reported in Polavarapu et al. (2016) for the GEM model. The differences between MPAS-A and CT2019 are expected due to the differences in the two models' horizontal resolution, dynamics, and physical parameterizations. Because no CTM can be expected to have perfect transport, the acceptability of transport is generally judged through comparisons of model simulation with measurements.

3.3.3 Comparison with near-surface CO_2 measurements

This section compares MPAS-A simulated CO_2 with hourly measurements from 50 stations that were used for the IFS model evaluation in Agusti-Panareda et al. (2019). The information of the 50 stations, including location, elevation, intake height, reference, and type is listed Table **??**. Like in Agusti-Panareda et al. (2019), only the highest intake level is used at towers that have multiple intake heights. When multiple observations within an hour are available (such as those with 30-min or shorter time interval), they are averaged to yield a single hourly value. For a given station this results in 744 (24×31) hourly

measurements per month at the maximum.

The MPAS-A hourly CO_2 statistics, including RMSE, STDE, and bias at the 50 stations are listed in Tables S1 and S2 of the supplement for January and July of 2014 respectively. For comparison, Tables S1 and S2 also include the statistics from the IFS 9 km and 80 km resolution simulations (Agusti-Panareda et al., 2019) at the same sites for the same time periods. Table S1 shows that RMSE of the MPAS-A simulated hourly CO_2 ranges from 0.17 ppm at the SPO station to 16.65 ppm at the KAS station. In comparison, the IFS simulations also resulted in a much lower RMSE at the SPO than KAS, the latter of which has a RMSE of 4.44 ppm from the 9 km resolution simulation and 10.71 ppm from the 80 km simulation.

The comparison of RMSE and STDE from MPAS-A and IFS are show in Figs **??** and **??** for January and July of 2014, respectively. Table **??** provides a quantitative summary of the hourly CO_2 RMSE between MPAS-A and the IFS 9 km and 80 km simulations. The table shows that for January 2014, the mean RMSE at the 50 stations is 4.20 ppm from MPAS-A, which is higher than IFS 9 km simulation (3.12 ppm, $p = 0.01$) and similar to the IFS 80 km simulation (4.94 ppm, $p = 0.25$). For July 2014, the mean RMSE at the 50 stations is 8.09 ppm from MPAS-A, which is similar to IFS 9 km simulation (8.04, $p = 0.95$) and lower than the IFS 80 km simulation (11.77 ppm, $p = 0.04$). The above comparisons indicate that the 60-15 km MPAS-A simulation has a accuracy level between the IFS 9 km and 80 km simulations.

Agusti-Panareda et al. (2019) found that atmospheric CO_2 transport is generally better represented at higher horizontal resolutions, and mountain stations display the largest improvement at higher resolution as they directly benefit from the more realistic orography. There are 12 mountain stations of the 50 stations used for the model validation. Table **??** lists the 12 mountain stations in two groups: the first group includes the six mountain stations located at the 15 km cells of the MPAS-A's 60-15 km variable-resolution grid, and the second group includes the other six stations that are located at the 60 km cells of the grid. The table lists the hourly CO_2 RMSE for each of the 12 stations from MPAS-A and IFS 9 km and 80 km simulations are listed for January and July 2014. The table shows that at each of the six mountain stations located at 15 km cells, MPAS-A has lower hourly CO_2 RMSE than the IFS 9 km simulation for July 2014. For January 2014~~vector wind RMSE reported in Agusti-Panareda et al. (2019). The error statistics from the other two seasons are of similar magnitude as the summer 2016 and winter 2017 seasons .~~, MPAS-A has lower RMSE than IFS 9 km simulation at five out the six stations (the exception is NWR). In comparison, at the six mountain stations located at its 60 km cells, MPAS-A has higher hourly CO_2 RMSE than IFS 9 km simulation for both January and July of 2014 with the exception of JFJ for July 2014.

3.3.4 ~~Overall accuracy of MPAS modeled~~

3.3.4 Comparison with TCCON XCO_2 measurements

After the comparison with the near-surface CO_2 in the last section, we evaluate MPAS-A CO_2 fields using XCO_2 measurements from 18 TCCON sites listed in Table 7. To compare with TCCON retrieved XCO_2, smoothed MPAS-A XCO_2 is calculated following Wunch et al. (2010):

$$X_{CO_2}^{model} = c_a + \mathbf{h}^T \mathbf{a}^T (\mathbf{x}_m - \mathbf{x}_a) \tag{20}$$

where $X_{CO_2}^{model}$ is the smoothed MPAS-A XCO_2, c_a is the a priori total column, $\mathbf{a}^T$ is TCCON column averaging kernel, $\mathbf{h}^T$ is a dry-pressure weighting function, $\mathbf{x}_m$ is MPAS-A CO_2 dry mole fraction profile, $\mathbf{x}_a$ is the a priori CO_2 dry mole fraction profile. The column profile of CO_2, air pressure, and water vapor mixing ratio extracted from MPAS-A hourly output are interpolated to the same vertical grid as $\mathbf{x}_a$, and dry-pressure weighting function $\mathbf{h}^T$ is calculated following O'Dell et al. (2012) and Eq. A7 of Agusti-Panareda et al. (2014).

~~We~~ At a given TCCON site, averaged hourly XCO_2 (denoted as $X_{CO_2}^{TCCON}$) is calculated as the mean value of all valid XCO_2 retrievals within the hour. $X_{CO_2}^{TCCON}$ are then matched with the calculated hourly XCO2 from MPAS-A (denoted as $X_{CO_2}^{model}$). The comparisons of $X_{CO_2}^{model}$ and $X_{CO_2}^{TCCON}$ at the 18 TCCON sites for the year of 2014 are shown in Fig. 7. The results indicate that the observed seasonal variation in TCCON XCO_2 are in general well represented by MPAS-A. The hourly average XCO_2 comparison between MPAS-A and TCCON are summarized in Table **??**. In the table N is the number of data pairs used for calculating the statistics, including RMSE, bias, and correlation coefficient R. The mean RMSE of the 18 sites is 1.35 ppm, which is comparable to the IFS simulations (1.02 to 1.25 ppm) Agusti-Panareda et al. (2019). We then calculated the average daily XCO_2 as the mean value of all the hourly XCO_2 within a given day. The statistics of comparison of daily XCO_2 between MPAS-A and TCCON are also included in Table **??**. In the table N is the number of average daily XCO_2 used for calculating the statistics. Compared to their hourly counterparts, the average daily XCO_2 has lower RMSE and higher correlation coefficient. The mean value of the average daily XCO_2 RMSE of the 18 TCCON sites is 1.23 ppm, which is comparable to IFS simulations (0.97 to 1.25 ppm) reported in Agusti-Panareda et al. (2019).

3.4 Model evaluation at regional scale

In this section, we present an evaluation of the MPAS-A CO_2 simulation accuracy using an extensive high resolution CO_2 observation data acquired through the ACT aircraft campaigns. ACT is a National Aeronautics and Space Administration (NASA) Earth Venture Suborbital 2 (EVS-2) mission, and its goal is to improve atmospheric inversion estimates of CO_2 and CH_4 through extensive airborne measurements over the eastern United Stated during multiple seasons (Davis et al., 2018a) . Through four campaign seasons from Summer 2016 to Spring 2018 with two research aircraft (C130 and B200), the ACT project has collected an extensive dataset of highly resolved CO_2 measurements in both BL and FT. The duration of the ACT campaign seasons is given in Table **??**. To use ACT airborne CO_2 measurements for model evaluation, we conducted a MPAS-A simulation lasts from January 1 2016 to May 31 2018. The first 6 months is for the model spin-up. The simulation

uses the domain and configurations as described in Section. 3.1, and model outputs are saved at 1-hour intervals.

First we compare MPAS-A simulated horizontal wind fields during the ACT campaign seasons using the same procedure described in Section 3.3.1. Table 10 lists the statistics of horizontal wind fields evaluation at the four ACT campaign seasons. The table indicates the same pattern as in 2014 (Table 2): mean RMSE vector wind increases with altitude and mean difference of wind direction decreases with altitude. The magnitude of the statistics of the four ACT campaign seasons are comparable to that of 2014 (Table 2).

Next we use the ACT airborne measurements to evaluate ~~MPAS~~ MPAS-A CO_2 simulation regarding its overall accuracy and its performance measured by three model evaluation metrics proposed by Pal et al. (2020). To provide an objective reference, we also compare ~~MPAS~~ MPAS-A performance with two established CO_2 model systems: WRF-Chem (Skamarock et al., 2008) and CT2019 (Jacobson et al., 2020) using the same set of airborne measurements.

WRF-Chem is an online ~~chemistry transport model~~ CTM based on the regional ~~NWP~~ model WRF (Grell et al., 2011; Skamarock et al., 2008). WRF-Chem simulations have been carried out at 27 km horizontal grid (Fig. S2) over North America as a part of the ACT project (Feng et al., 2020). The WRF-Chem simulations use ERA5 reanalysis (Hersbach et al., 2020) for meteorological initial and lateral boundary conditions, CarbonTracker ~~(Jacobson et al., 2007)~~ (Jacobson et al., 2020) posterior mole fraction for CO_2 initial and boundary conditions, and CarbonTracker posterior fluxes for surface CO_2 fluxes. The WRF-Chem simulations use meteorological nudging and 120-hour meteorological re-initialization to keep meteorological fields close to the analysis. ~~CarbonTracker (Jacobson et al., 2007, 2020)~~

CarbonTracker (Jacobson et al., 2020) is an operational carbon data-assimilation system which uses Transport Model 5 (TM5) (Krol et al., 2005) for ~~atmosphere~~ atmospheric transport. TM5 is an offline global ~~chemical transport model~~ CTM which includes CO_2 advection, deep and shallow convection, and vertical diffusion in both ~~the planetary boundary layer~~ PBL and free troposphere (Krol et al., 2005). In producing CT2019 CO_2 mole fraction (Jacobson et al., 2020), TM5 simulation ran over a $3^\circ \times 2^\circ$ global domain and an nested $1^\circ \times 1^\circ$ North America domain driven by ERA-interim reanalysis (Dee et al., 2011).

We use the ACT 5-second averaged CO_2 measurement dataset (Davis et al., 2018b), which has a horizontal resolution approximately 500 m given the average aircraft velocity. MPAS simulated CO_2 fields are interpolated in time and space to match each 5-second airborne data points. WRF-Chem simulated CO_2 fields are also interpolated to match the ACT 5-second data point using the same approach as ~~MPAS~~MPAS-A. CarbonTrack CO_2 used for the evaluation is obtained from CarbonTrack ObsPack ~~((Masarie et al., 2014))~~(Masarie et al., 2014), which is the CT2019 posterior mole fraction interpolated to the ACT 5-second data points.

For each ACT flight day, CO_2 measurements from the two aircraft are combined if both are available, and their corresponding modeled CO_2 values from ~~MPAS~~MPAS-A, WRF-Chem, and CT2019 are combined in the same way. With the four seasons

combined, there are a total of 97 flight days (Pal and Davis, 2020), each one presented by an observation-model dataset consisted of observed CO_2, modeled CO_2 from the three models, along with the time, latitude, longitude, and altitude of each observation data point. Using the ACT maneuver flag dataset Pal et al. (2020), we further divide each flight day's data into two groups: one for ~~the boundary layer (BL)~~ BL and another for ~~he~~ the free troposphere (FT). ~~RMSE and bias of the modeled from the three models (MPAS, WRF-Chem, and CT2019) are then calculated for each flight day, with the calculations for~~ For each ACT campaign season, all the BL data-model pairs are combined for each of the three models for model comparison. Figure 8 shows the Taylor diagram of the model comparison in BL for the four campaign seasons. N in the title of each figure is the ~~BL and FT carried out separately. In addition to the RMSE and bias, standard deviation is also calculated for the observed and modeled for assessing each model's representation of spatial variability.~~

~~Using RMSE as an accuracy metric, we compare MPAS simulation with WRF-Chem~~ number of model-data pairs used for plotting the diagram. Similarly the model comparison in FT is summarized in the Taylor diagrams of Fig. 9. A comparison of Figs. 8 and ~~CT2019 in Fig. **??**. The figure shows~~ 9 show that all three models have higher accuracy (lower RMSE) in FT than BL, which ~~is most likely because flux errors impact simulated in BL more than in FT . The figure also shows that compared with WRF-Chem, MPAS has a similar magnitude of RMSE overall in both BL and FT, with some exception where substantial differences exist between the two models. Averaged over the 97 flight days, the mean RMSE is 4.49/1.85 ppm (BL/FT) for MPAS, and 4.91/1.71 ppm for WRF-Chem, indicating MPAS achieved a slightly higher accuracy than WRF-Chem in BLwhich maybe partially because the former has a higher horizontal resolution (15 km) than the latter (27 km). In free troposphere, WRF-Chem resulted in a slightly higher accuracy than MPAS maybe because that it applied meteorological nudging while MPAS did not. Compared with~~ could be attributed to the larger error in the weather forecast in BL than FT associated with the accuracy of PBL height in the model simulation. Figure 8 shows that in BL, MPAS-A has higher RMSE and higher standard deviation than CT2019~~, MPAS simulations resulted in larger RMSE in the majority of flight days, and the differences in RMSE between the two models are large in BL than FT. Averaged over the 97 flight days, the mean RMSE for~~ . MPAS-A has more accurate estimation of the observations' standard deviation than CT2019 ~~is 3.36 ppm and 1.42 ppm in BL and FT respectively, which are substantially lower than MPAS (4.49 ppm/1.85 ppm in BL/FT).~~

~~Next we examine how well each model represents the spatial variability (as measured by standard deviation). Fig. **??** shows the comparison of the standard deviation (σ) of the modeled and aircraft observed . The figure shows that the observed CO_2 has higher variability (σ) in BL than in FT(note that different scales are used for BL and FT plots). The figure also shows that the MPAS simulated has larger σ than the observations in most of the flight days (Fig. **??**(a). and (d)). In comparison, CT2019 CO_2 has substantially lower variability than the observations in the majority of the flight days (Fig. **??**(c). and (f)), especially in the boundary layer. Averaged over the 97 flight days, ACT airborne observationshas a mean σ of 3.53 ppm in BL, comparing to 4.27 ppm of MPAS, 4.63 ppm of~~ in all but summer 2016. Compared with WRF-Chem, MPAS-A has lower RMSE and more accurate estimation of the observations' standard deviation. Figure 9 shows that in FT, MPAS-A has higher RMSE than CT2019 in all four campaign seasons and but it has more accurate estimation of the observations' standard deviation than CT2019 in all but summer 2016 season. Compared to WRF-Chem, ~~and 2.23 ppm of CT2019.~~MPAS-A has lower RMSE and

more accurate estimation of observations' standard deviation in all but summer 2016.

~~The above model evaluation using airborne observationsshows that the MPAS transport model is capable of achieving a similar level of accuracy as 27 km resolution WRF-Chem simulation. Compared with the global inversion system CT2019, although MPAS resulted in higher RMSEs, it achieved better estimations of the observed spatial variability.~~

3.4.1 Model representation of CO_2 difference between warm and cold sectors

Through analyzing the ACT Summer 2016 campaign data, Pal et al. (2020) identified three consistent features in CO_2 mole fraction and proposed to use these features as transport model assessment metrics. The three features are the differences between the warm and cold sectors, the difference between the boundary layer and free troposphere, and the CO_2 enhancement bands in the vicinity of frontal boundaries. Here and in the next two sections, we evaluate how ~~MPAS~~ MPAS-A simulated CO_2 represents the three features.

Using the ACT maneuver flag dataset (Pal et al., 2020), we identified flights that crossed a weather front and their associated warm and cold sectors. The CO_2 mole fraction statistics for the warm and cold sectors are calculated from the aircraft measurements and the modeled CO_2 by ~~MPAS~~MPAS-A, WRF-Chem, and CT2019, respectively. The results are shown in Fig. ~~??~~10, which summarizes the statistics of CO_2 mole fraction differences between the warm and cold sectors measured by 15 front-crossing flights: 10 from the summer 2016 season and 5 from the winter 2017 season. The figure confirms that the warm sector has higher average CO_2 mole fraction in the boundary layer than the cold sector during summer 2016 as reported by Pal et al. (2020). The figure also shows that the average CO_2 mole fraction in the warm sectors are lower than than the colder sectors in winter 2017, opposite to the summer 2016.

~~Fig. ?? also shows that MPAS simulated well represents the difference between the warm and cold sectors well in all 15 cases. Table ??~~ Table 11 lists the mean ~~and standard deviation~~ CO_2 of the warm ~~and cold sectors~~sector, cold sectors, and their difference as calculated from the ACT measurements, ~~MPAS~~MPAS-A, WRF-Chem, and CT2019. The table shows that the ~~MPAS~~ MPAS-A simulations are similar to WRF-Chem, and both tend to have larger CO_2 differences between the warm and cold sectors than CT2019. For instance, the ~~2016-08-24~~ 2016-08-08 case where the observed mean CO_2 difference between warm and cold sector is ~~about 14 ppm, MPAS~~ 26.9 ppm, MPAS-A and WRF simulations resulted in ~~13.8 ppm and 7.8~~ 36.9 ppm and 21.2 ppm respectively, while CT2019 ~~results in a 3.3~~ resulted in a 15.3 ppm difference. The above evaluation indicates that ~~MPAS~~ MPAS-A CO_2 model is capable of well representing the observed CO_2 difference between the warm and cold sectors, and its accuracy in this respect is ~~similar~~ comparable to WRF-Chem and CT2019.

3.4.2 Model representation of CO_2 vertical difference

The second feature identified by Pal et al. (2020) is the vertical difference of CO_2 mole fraction between BL and FT. During ACT campaign season, two research aircraft (B200 and C130) took many vertical profile measurements during take off, landing, spiral up and down, and inline ascend and descend maneuvers (Pal, 2019). These profile observations characterize the vertical variation of the atmospheric CO_2 mole fraction. From the vertical profile measurements taken during the summer 2016 season, Pal et al. (2020) calculated the mean CO_2 mole fraction in ~~the boundary layer (BL) and free troposphere (FT)~~BL and FT, denoted as $[CO_2]_{BL}$ and $[CO_2]_{FT}$ respectively. They further defined BL-to-FT CO_2 difference as $\Delta[CO_2] = [CO_2]_{BL} - [CO_2]_{FT}$. They found that $\Delta[CO_2]$ tend to be positive in the warm sector and negative in the cold sector. In this section, we evaluate how well ~~MPAS represent~~ MPAS-A represents the BL-to-FT CO_2 difference and compare its performance with WRF-Chem and CT2019.

Using the ACT maneuver flag dataset (Pal et al., 2020), we identified all vertical profiles taken during the four campaign seasons, from which we selected profiles that meet two criteria: (1) a vertical profile must include at least 20 5-second measurements in the boundary layer and 20 measurements in the free troposphere; and (2) a vertical profile must extend at least 2 km in the vertical direction. These two criteria are used to ensure that the resulting $[CO_2]_{BL}$ and ~~$[CO_2]_{BL}$~~ $[CO_2]_{FT}$ are statistically representative. A total of 199 qualified vertical profiles are identified from the four campaign seasons, including 72 from the summer 2016 season, 27 from winter 2017, ~~41~~ 40 from fall 2017, and ~~59~~ 60 from spring 2018. For each of the vertical profiles, $\Delta[CO_2]$ is calculated for the aircraft CO_2 measurements, and the simulated CO_2 by ~~MPAS~~MPAS-A, WRF-Chem, and CT2019. We compare $\Delta[CO_2]$ from the models with that from the ~~observation~~ observations to assess how each model represents the observed BL-to-FT CO_2 difference. ~~Fig. ??~~ Figure 11 shows the comparisons grouped by the campaign seasons. The figure indicates a clear distinction in $\Delta[CO_2]$ between the summer 2016 and the other three seasons: There are a substantial number of both positive and negative $\Delta[CO_2]$ in the summer 2016 season, but the vast majority of cases in the rest of the three campaign seasons have positive $\Delta[CO_2]$. The positive BL-to-FT CO_2 differences from the winter 2017 season measurements could be at least partially attributable to the lack of CO_2 draw-down during the non-growing season. In comparison, the fall 2017 and spring 2018 seasons have more mixed results probably because of their partial overlap with the growing season. For the summer 2016 season, vertical profiles with negative $\Delta[CO_2]$ (lower mean CO_2 in BL than FT) suggest photosynthesis during the growing season, but those with positive $\Delta[CO_2]$ values are probably caused by the interaction between photosynthesis and frontal passage (Pal et al., 2020).

~~The comparison of~~ To compare the three models' accuracy in representing the BL-TO-FT CO_2 difference, we calculated the mean absolute error (MAE) for each model at each season, where $AE = |\Delta[CO_2]_{model} - \Delta[CO_2]_{obs}|$ (the absolute difference in $\Delta[CO_2]$ ~~in Fig. ?? between the aircraft observations and the model simulations show how well the three modelsrepresent the BL-to-FT difference. For instance, the figure indicates that MPAS represents the BL-to-FT difference more accurately during the fall 2017 seasons than the summer 2016 season. In comparison CT2019 represents BL-to-FT difference more accurately~~

during the summer 2015 season than the Fall 2017 between a model and the ACT observations).

$$\mathrm{MAE} = \frac{1}{N}\sum_{i=1}^{N} \mathrm{AE}_i \tag{21}$$

Table 12 summarize the MAE of the three models for each season. The figure also show that while the comparisons between CT2019 and the aircraft observations have less scattering than MPAS and WRF-Chem, CT2019 also tend to underestimate the range of the BL-to-FT differences, particularly during the spring 2018 season. To provide a quantitative evaluation, we calculated RMSE for each of the model-observation comparison of Fig. **??**. In addition, the standard deviation of $\Delta[CO_2]$ is calculated for the observation and the three models for each season. The resulting statistics are summarized in Table **??** which shows that : MPAS has lower RMSEs than WRF-Chem in all but the summer 2016 season, and it has lower RMSEs than p values of paired t tests of AE between MPAS-A and the other two models are also listed in the table to provide the significance level of the model comparisons. Using $p = 0.1$ as the cut-off value the table shows that MPAS-A has smaller MAE than CT2019 for the winter and fall of in fall 2017 but higher in the and a larger MAE in summer 2016. The differences between the two models in the other two seasons . Regarding the standard deviations, MPAS compares with the observation better than are not significant. Compared to WRF-Chemin all four season, and better than CT2019 in all but the summer 2016 season, MPAS-A has smaller MAEs in winter 2017 and spring 2018 while the differences in the other two seasons are not significant. In summary, the above model evaluation and comparison demonstrate that MPAS MPAS-A CO_2 transport model is capable of representing the aircraft observed CO_2 difference between boundary layer and free troposphere BL and FT at least as accurately as WRF-Chem and CT2019.

3.4.3 Model representation of CO_2 enhancement at frontal boundaries

The third feature identified by Pal et al. (2020) in the summer 2016 aircraft measurements is the bands of enhanced CO_2 close to frontal boundaries in the boundary layerBL. They found these CO_2 enhancement bands are typically about 100 km wide and speculated that it would require a 20-km horizontal resolution model to effectively represent the feature. In this section, we identify the frontal boundary CO_2 enhancements in the four campaign seasons and examine how well they are represented by MPAS. For instance, Fig. **??** shows the MPAS simulated equivalent potential temperature (θ_e) and mole fraction at 18:00 UTC 4 August 2016. The sharp boundary in θ_e indicates a surface cold front extending from southern Colorado northeastward to Wisconsin. Abrupt horizontal wind direction changes shown in Fig. S3 (supplement material) also indicate the cold front and its southeastward movement. Meteorological measurements taken during the flight (not shown) confirm the cold front passage too. The B200 research aircraft crossed the cold front from southeast to northwest at about 400-500 meters above the ground between 17:15 UTC and 19:15 UTC, and its flight track and timing are marked on Fig. **??**. The aircraft measurements show an approximately 20 ppm enhancement along the front boundary, which can be clearly identified in the MPAS simulated mole

fraction (lower panel of Fig. ??). MPAS-A.

Using the same approach as Pal et al. (2020), a total of 48 front-crossing level-leg flight constant-altitude flight segments are identified from the four seasons (15 from Summer 2016, 5 from Winter 2017, 17 from Fall 2017, and 11 from Fall 2018). To evaluate how well MPAS MPAS-A represents the frontal boundary CO_2 enhancements and compare its performance with WRF-Chem and CT2019, CO_2 mole fraction measured by the aircraft and simulated by the three models are plotted together for each of the identified front-crossing level-leg flight constant-altitude flight segment. Figure 12 includes 8 of the front-crossing flight segments and the full set is included in Fig. ??S3 of supplement. For each flight in the figure, the vertical dashed line marks the approximate time when the aircraft crossed the frontal boundary, and the associated segment in Fig. 12, the pair of vertical dashed lines mark CO_2 enhancement observed by the aircraft along a frontal boundary. The warm and cold sectors are labeled on either side of the boundary. Examination of the aircraft observations (black lines) in the figure indicate associated with the frontal boundary in each flight are labeled as warm and cold respectively. The figure indicates that frontal boundary CO_2 enhancements can be identified in most of the flights flight segments but not all cases. For instance, there is not clearly identifiable CO_2 enhancement in the B200 flights on 2017-10-08 and 2018-04-23 .(Fig. S3).

Fig. ?? shows that MPAS (red lines in the figure) Figure 12 shows that MPAS-A has a varying degree of success in simulating the frontal boundary CO_2 enhancements: it represents both the timing and the magnitude of the enhancements very well in some cases (2017-10-28 and 2017-11-03 2016-08-04 and 2017-10-18 by B200), but results in substantial errors in either the timing (2016-08-12 2016-07-25 B200) or the magnitude (2017-03-10 C130) in other cases. The figure also shows that the MPAS MPAS-A simulated CO_2 is more similar to WRF-Chem than CT2019: CT2019 tends to substantially underestimate the magnitude of CO_2 enhancement while MPAS MPAS-A and WRF-Chem tend to overestimate.

To provide an quantitative comparison between the three models' accuracy regarding the frontal boundary Figure 13 shows the MPAS-A simulated equivalent potential temperature (θ_e) and CO_2 variation, we calculated RMSE and standard deviation for each of the 48 level-leg flights mole fraction at 18:00 UTC August 4, 2016. The sharp boundary in θ_e indicates a surface cold front extending from southern Colorado northeastward to Wisconsin. Abrupt horizontal wind direction changes shown in Fig. ??. For each flight, RMSE is calculated for each of the three models as compared with the aircraft observations, and standard deviation is calculated for both the models and aircraft observations. The resulting RMSEs from MPAS are compared with WRF-Chem and CT2019 in Fig. ??. The figure indicates that MPAS RMSEs in general are similar in magnitude to CT2019 and lower than WRF-Chem. Averaged over the 48 cases, S4 of the supplement also indicate the cold front and its southeastward movement. Meteorological measurements taken during the flight (not shown) also confirm the cold front passage. The B200 research aircraft crossed the cold front from southeast to northwest at about 400-500 meters above the ground between 17:15 UTC and 19:15 UTC, and its flight track and timing are marked on Fig. 13. The aircraft measurements show an approximately 20 ppm enhancement along the front boundary, which can be clearly identified in the MPAS-A simulated CO_2 mole fraction

(lower panel of Fig. 13).

Figure 14 compares the ~~mean RMSE is 4.63 ppm for MPAS, 4.68 ppm for CT2019, and 5.86 ppm for WRF-Chem, indicating MPAS perform as well as the other two models as measured by the RMSEs. We also assess how well the~~ three models ~~estimate the spatial variability~~ in their representation of the frontal boundary ~~by comparing the standard deviation (σ) of the three models with the observations (Fig. **??**).~~ CO_2 variation. The figure shows that ~~MPAS and WRF-Chem represent the spatial variability in the 48 level-leg flights better than~~ except for summer 2016, MPAS-A has similar level of RMSE as CT2019 ~~which substantially underestimates the variability in the majority of the cases~~and it has more accurate estimation of the observations' standard deviation. As horizontal resolution impacts a model's ability to represent small scale spatial variability (Agusti-Panareda et al., 2019), the coarser resolution of CT2019 ($1^\circ \times 1^\circ$ over the North America) is likely the primary cause of its underestimation of the frontal boundary CO_2 variability.

3.5 ~~Model evaluation using near-surface hourly CO_2 observations~~

~~The MPAS transport model has been evaluated using the extensive high-resolution aircraft measurements from four ACT campaign seasons. Considering that these aircraft measurements were all acquired during the day time hours over the eastern United States, here we carry out further model evaluation using near-surface hourly observations acquired by surface and tower stations located across the globe. For this evaluation, we conducted another set of MPAS simulations which cover the month of January and July of 2014 using the same model domain and configurations as described in Sect. 3.1. As this is the same time period that Agusti-Panareda et al. (2019) used for their IFS global simulations, we can assess the MPAS model performance through a comparison with the IFS simulations.~~

~~As with the simulations for the 2016-2018 ACT campaign seasons, we first evaluate the MPAS simulated horizontal wind fields using radiosonde observations from 457 stations located around the globe (Fig. S3 of the supplement material). The resulting statistics, including the RMSE of vector wind, RMSE of wind speed and wind direction, and the mean wind direction difference are shown in Table **??**. Compared with the IFS simulations by Agusti-Panareda et al. (2019) (their Fig. 4), the MPAS simulated horizontal wind fields have larger errors in both January and July of 2014. For instance, the IFS 80 km resolution simulation has a vector wind RMSE about 4.5 m/s at the 200 hPa level in January while MPAS results in 5.16 m/s at the same pressure level.~~

~~We then compare MPAS simulated with hourly measurements from 50 stations that were used for the IFS model evaluation in Agusti-Panareda et al. (2019). The information of the 50 stations, including location, elevation, intake height, and reference is listed Table **??**. Like in Agusti-Panareda et al. (2019), only the highest intake level is used at towers that have multiple intake heights. When multiple observations within an hour are available (such as those with 30-min or shorter time interval), they are averaged to yield a single hourly value. For a given station this result in 744 (24×31) hourly measurements per month at the maximum. Following Patra et al. (2008) and Agusti-Panareda et al. (2019), MPAS hourly outputs are horizontally sampled by taking the nearest land cell to a given station. The resulting statistics (RMSE, bias, and STDE) for the MPAS simulation covering the month of January 2014, along with their counterparts from the IFS 9 km and 80 km resolution simulations~~

(Agusti-Panareda et al., 2019), are listed in Table S1 of the supplement material. The table shows that RMSEs of the MPAS simulated hourly ranges from 0.36 ppm at the spo to 25.96 ppm at ssl station. In comparison, the IFS simulations also resulted in a much lower RMSE at the spo than ssl, the latter of which has a RMSE of 5.83 ppm from 9 km resolution simulation and 23.99 ppm from the 80 km simulation. Agusti-Panareda et al. (2019) noted that the increasing horizontal resolution improves model simulation accuracy the most at mountain stations. The Schauinsland station (ssl), located at the southwest Germany with an elevation of 1205 meters above the means sea level, has considerable variability because its vicinity to biogenic and anthropogenic sources and sinks (Schmidt et al., 2003). This station is located in the 60-km resolution portion of the MPAS 60-15km global domain, and the MPAS RMSE (25.96 ppm) at the station is closer to the IFS 80 km simulation (23.99ppm) than MPAS-A has lower RMSE than WRF-Chem in winter 2017 and spring 2018, and similar RMSE as WRF-Chem in the 9km simulation (5.83 ppm), suggesting the influence of the model horizontal resolution. At this station, MPAS results in a positive bias of 21.34 ppm and IFS 80 km simulation has a positive bias of 18.72 ppm, meaning that both models substantially overestimate the hourly . An examination of the MPAS simulated hourly (not shown) indicates that the large bias is mostly caused by overestimation at the station during the nighttime hours. Such overestimation could be caused by a combination of errors in fluxes, vertical mixing, and large scale transport. other two seasons. In all but summer 2016, MPAS-A has more an accurate estimation of the observations' standard deviation than WRF-Chem.

Finally, we compare RMSE and random error (STDE) of the MPAS simulation at the 46 stations with the IFS simulations in Fig. **??**. The figure shows that MPAS has larger RMSE and STDE than the IFS 9 km simulation, but similar to the 80 km IFS simulation. Similar model evaluation and comparison carried out for the month of July 2014 are shown in Table S2 (supplement material) and Fig. **??**. Comparing with the January simulation, Fig. **??** shows that both MPAS and IFS have larger RMSEs in July, suggesting the impacts of large uncertainty in biospheric carbon fluxes during boreal summer. Fig. **??** also shows that the MPAS simulation has larger RMSE and STDE than the 9 km IFS simulation but similar to the 80 km IFS simulation, a similar pattern as in January.

4 Summary

This paper implemented transport in the global variable-resolution model MPAS-A v6.3. The atmospheric We implemented the CO_2 atmospheric transport processes, including advection, vertical mixing, and convective transport, are implemented in the modelin the global variable-resolution model MPAS-A. After the model development details are presented, simulation experiments designed for model evaluation are described. Two sets of simulations over a 60-15 km variable-resolution global domains domain are conducted for model accuracy evaluation using an extensive aircraft measurements over the eastern United States and near-surface hourly measurements from surface and tower stations distributed across the globe. Meteorology Meteorological initial conditions for these simulations are from the ERA-interim analysis (Dee et al., 2011), and CO_2 initial conditions and fluxes are from CT2019 posterior mole fraction and fluxes products (Jacobson et al., 2020). To keep model meteorological fields close to the analysis, meteorology re-initialization are applied at 24-hour interval throughout the simulation periods. Global CO_2 mass conservation property is assessed by a one-year continuous simulation without meteorology re-

initialization and fluxes, and the results show that ~~MPAS~~ MPAS-A is capable of maintaining total dry air mass conservation to the limit of machine precision. ~~A 48-days simulation~~ During the one-year simulation period, the total CO_2 mass change is about 10^{-5} of its initial value. The larger variation of CO_2 mass than the dry air is due to the complex and strong spatial gradient caused by the surface CO_2 fluxes. Another one-year simulation with meteorology re-initialization indicates that changes in dry air density during the re-initialization causes changes in global total CO_2 mass, and a scaling method applied after each re-initialization is able to reduce the change from $\sim 10^{-4}$ to $\sim 10^{-6}$ of the global CO_2 mass~~variation to less than $\pm 0.001\%$ of its initial value~~.

The accuracy of ~~MPAS~~ MPAS-A CO_2 transport is evaluated ~~using the~~ first at the global scale and then at the regional scale. At the global scale, MPAS-A simulation is evaluated using CT2019, near-surface hourly CO_2 measurements from 50 stations and XCO_2 measurements from 18 TCCON stations. The resulting statistics are compared with the ECWMF IFS 9km and 80 km resolution simulations over the same period conducted by Agusti-Panareda et al. (2019). The comparison indicates that RMSE of the MPAS-A simulation is similar to the 80 km IFS simulation, but larger than the 9 km IFS simulation.

At the regional scale, A MPAS-A simulation extending from January 1 2016 to June 1 2018 is evaluated using the extensive high-resolution aircraft measurements from four ACT campaign seasons~~during 2016-2018~~. Compared with a 27 km resolution WRF-Chem simulation and CT2019 posterior CO_2 mole fraction, ~~MPAS~~ MPAS-A simulated CO_2 achieves a comparable level of accuracy (as measured by RMSE). Further evaluation using three metrics proposed by Pal et al. (2020) shows that ~~MPAS~~ MPAS-A simulation is capable of representing the observed CO_2 features as accurately as the WRF-Chem simulation and CT2019.

~~A second set of MPAS simulations for the month of January and July of 2014 are evaluated using near-surface hourly measurements from surface and tower stations across the global. The resulting statistic, including RMSE, random error, and bias, are compared with the ECWMF IFS 9km and 80 km resolution simulations over the same period conducted by Agusti-Panareda et al. (2019). The comparison indicates that RMSE and random error of the MPAS simulation is similar to the 80 km IFS simulation, but larger than the 9 km IFS simulation.~~

The model evaluations using the airborne and near-surface measurements, indicates that the newly developed ~~MPAS~~ MPAS-A CO_2 transport model is capable of achieving a comparable level of accuracy with the more established CO_2 modeling systems, including the regional model system WRF-Chem, the operational assimilation system CarbonTracker, and the lower resolution (80 km) simulation of ECWMF IFS global CO_2 modeling ~~sytem~~system. Although further improvements are expected, the ~~MPAS~~ MPAS-A CO_2 transport model has the potential to contribute to improving our knowledge of ~~the~~ atmospheric CO_2 transport and fluxes.

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