The Canadian atmospheric transport model for simulating greenhouse gas evolution on regional scales: GEM-MACH-GHG v.137-reg

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Abstract. In this study, we present the development of a regional atmospheric transport model for greenhouse gas (GHG) simulation based on an operational weather forecast model and a chemical transport model at Environment and Climate Change Canada (ECCC), with the goal of improving our understanding of the high spatio-temporal resolution interaction between the atmosphere and surface GHG fluxes over Canada and the United States. The regional model uses 10 km x 10 km horizontal grid spacing and 80 vertical levels spanning the ground to 0.1 hPa. The lateral boundary conditions of meteorology and tracers are provided by the global transport model used for GHG simulation at ECCC. The performance of the regional model and added benefit of the regional model over our lower resolution global models is investigated in terms of modelled CO₂ concentration and meteorological forecast quality for multiple seasons in 2015. We find that our regional model has the capability to simulate high spatial (horizontal and vertical) and temporal scales of atmospheric CO₂ concentrations, based on comparisons to surface and aircraft observations. In addition, reduced bias and standard deviation of forecast error in boreal summer are obtained by the regional model. Better representation of model topography in the regional model results in improved simulation of the CO₂ diurnal cycle compared to the global model at Walnut Grove, California. The new regional model will form the basis of a flux inversion system that estimates regional scale fluxes of GHGs over Canada.

20 1 Introduction

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The global mean atmospheric carbon dioxide (CO₂) concentration or mixing ratio (in mole fractions of dry air) has been increasing since the industrial revolution mainly due to anthropogenic emissions into the atmosphere, while terrestrial and oceanic uptake moderate the increase of CO₂ in the atmosphere (Canadell et al., 2007; Le Quéré et al., 2009). Apart from this global increase, information about each component affecting the global carbon budget and its uncertainties are estimated and updated regularly at the global scale, using a wide range of methods and data (Le Quéré et al., 2009; 2018). Since the ocean CO₂ sink have been increasing constantly in line with the increased CO₂ in the atmosphere (Le Quéré et al., 2018), the interannual variability of the CO₂ growth rate reached a record high mainly due to the impact of El Niño Southern Oscillation on the interannual variability of biospheric fluxes (Buchwitz et al., 2018) and increased net biospheric respiration in the tropics

(Liu et al., 2017). Globally, increased CO₂ and temperature are positively or negatively associated with terrestrial uptake by enhancing photosynthesis or respiration (Fernández-Martínez et al., 2018). Regionally, however, the carbon balance of Canadian boreal forest and its impact on global carbon budget is highly uncertain and the ecosystems of Canada are vulnerable to a changing climate (Kurz et al., 2013; Bush and Lemmen, 2019). Therefore, correctly accounting for biospheric fluxes over Canada is important for understanding both the global and regional carbon cycles.

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Surface sources and sinks of CO₂ can be estimated through inverse modelling using atmospheric CO₂ concentrations as a constraint to adjust prior fluxes so as to minimize the difference between the modelled CO₂ concentrations and observed values (Ciais et al., 2010). Many atmospheric inversion studies have been conducted to quantify surface CO₂ fluxes on both global and regional scales (Tans et al., 1990; Gurney et al., 2002; Peters et al., 2007; Lauvaux et al., 2008, 2012b). Although there is a consensus of estimated fluxes at the global scale, significant discrepancies among different inversion system results still exist, especially in partitioning terrestrial fluxes at continental scales (Peylin et al., 2013; Crowell et al., 2019) due to the contribution of atmospheric transport model errors and prescribed fossil fuel emissions (Peylin et al., 2011; Gaubert et al., 2019).

In an atmospheric inversion of CO₂, the transport model plays a key role in transforming the surface CO₂ flux information into atmospheric CO2 concentrations and can be used as a verification tool for estimated surface CO2 fluxes (Ciais et al., 2010; Nisbet and Weiss, 2010; Bergamaschi et al., 2018). The errors caused by an imperfect transport model can introduce biases and uncertainties into estimated fluxes during the inversion process (Law et al., 1996; Gloor et al., 1999; Engelen et al., 2002; Houweling et al., 2010; Chevallier et al., 2010, 2014; Locatelli et al., 2013). Such errors may arise from a variety of sources: model formulation, meteorological fields and representativeness errors. Model formulation errors may arise from processes associated with parametrizations of vertical mixing within the planetary boundary layer (PBL) (Lauvaux and Davis, 2014), vertical mixing between the PBL and the free troposphere (Stephen et al., 2007), isentropic transport (Parazoo et al., 2012; Barnes et al., 2016), synoptic scale variations due to advection and convection (Parazoo et al., 2008) and mid-latitude storm tracks (Parazoo et al., 2011). In fact, the impact of synoptic and mesoscale transport on the variability of CO₂ is comparable with that of surface fluxes (Chan et al., 2004). Since an atmospheric transport model is driven by meteorology, uncertainties in meteorological model and observations is another important source of error in the transport of tracers (Liu et al., 2011; Miller et al., 2015; Polavarapu et al., 2016). Finally, representation error is also a source of errors in inversions. The mismatch between coarse resolution transport model simulations and observations from real CO₂ field impacts the ability to resolve sub-grid scale variability of CO2. In particular, unresolved synoptic and mesoscale processes increase representation error (Engelen et al., 2002).

The sparseness of the CO₂ observation network used in inversions is another major contributing factor to the uncertainty in estimated fluxes. Increasing the density of the surface observation network is beneficial for reducing uncertainty and improving the accuracy of retrieved fluxes in the context of both global (Bruhwiler et al., 2011) and regional inverse modelling (Lauvaux et al., 2012a; Schuh et al., 2013). Since a number of new measurement sites have been established over Canada and the US in recent decades (e.g. Worthy et al., 2005; Andrews et al., 2014; Bush et al., 2019), it should now be

possible to obtain optimized fluxes on finer spatial scales and with reduced uncertainties. However, in order to better interpret information from spatially dense observation networks which contain information on strongly varying biospheric fluxes and strong sources of anthropogenic emissions, a high resolution atmospheric transport model capable of capturing these signals is needed.

Resolving the fine scale spatial and temporal variability of CO₂ generated by heterogeneous land surface and complex topography, which is not resolved by typical grid sizes of global models, is the primary motivation for regional scale inverse modelling. Increased horizontal resolution could alleviate transport and representation errors and thus improve simulations of synoptic variations of CO₂ concentrations (Patra et al., 2008; Remaud et al., 2018). Indeed, Gerbig et al. (2003) suggested that in order to resolve spatial variations of CO₂ in the PBL over a continent, a horizontal grid spacing no larger than 30 km is required. In addition, Pillai et al. (2011) showed that a maximum horizontal resolution of 12 km is required to represent the variability of CO₂ concentrations especially over mountainous or complex terrain. To this end, several studies focusing on forward CO₂ simulation at regional scales were carried out, using different models and configurations over various regions of interest. One approach to simulate atmospheric CO₂ concentration at finer spatial and temporal resolution is using zooming or nested domains within a global model (Krol et al., 2005; Lin et al., 2018). Another option is to use a regional atmospheric transport model. Various kinds of regional scale modelling studies have been conducted for the mid continental region of North America (Díaz-Isaac et al., 2014), south west France (Ahmadodv et al., 2007, 2009), western Europe (Kretschmer et al., 2014) and East Asia (Ballav et al., 2012). By increasing horizontal and vertical resolutions, regional models have an advantage over global models in terms of simulating CO₂ concentrations as shown by intercomparison experiments (Geels et al., 2007; Pillai et al., 2010; Díaz-Isaac et al., 2014).

At Environment and Climate Change Canada (ECCC), a carbon assimilation system (EC-CAS) is under development in order to estimate surface greenhouse gas (GHG) states and fluxes. To this end, a GHG forward modelling system which includes coupled meteorology and tracer transport model with full model physics, namely GEM-MACH-GHG (Polavarapu et al., 2016), has been developed. GEM-MACH-GHG is based on an operational weather forecast model, Global Environmental Multiscale model (GEM) at Canadian Meteorological Centre (CMC) (Côté et al. 1998a, b; Girard et al., 2014), and a chemical transport model with complete tropospheric chemistry, GEM-Modelling Air quality and Chemistry (GEM-MACH) model (Moran et al., 2010; Robichaud and Ménard, 2014; Makar et al., 2015), although the tropospheric chemistry module is not used in GEM-MACH-GHG simulations. GEM-MACH-GHG with 0.9° horizontal grid spacing is capable of simulating CO₂ concentrations over the globe acceptably well in comparison with in-situ and surface-based column averaged CO₂ observations. GEM-MACH-GHG was also used to investigate the uncertainty of CO₂ transport across different global transport models (Polavarapu et al., 2018), and was tested with the Canadian Land Surface Scheme and Canadian Terrestrial Ecosystem Model (CLASS-CTEM) in order to be able eventually to consistently simulate atmosphere-land exchange of CO₂ over the globe (Badawy et al., 2018). While a limited area version of the GEM model exists for operational weather and air quality forecasting, the ability to simulate GHGs on a regional model domain over a continental region had not been developed before now.

In this paper, in order to obtain a better understanding of the variability of atmospheric CO₂ concentration at finer spatio-temporal scales over a continental region during a relatively long time period, a regional scale atmospheric transport model for GHG simulation based on GEM-MACH-GHG is developed and tested. As a first step, CO₂ simulations with a 10 km grid spacing are performed for the year 2015, on a domain covering most of Canada and the US. The performance of the new model is investigated using meteorological and CO₂ concentration observations. In addition, the added benefit of the regional model over the global model in terms of CO₂ simulation as well as weather forecasts is investigated. The article is organized as follows. A description of the model, data and methodologies used in this study is provided in Sect. 2. In Sect. 3 the performance of the regional model is assessed in terms of its meteorological forecast and CO₂ simulation capability through comparisons with global model results. The benefit of higher horizontal resolution is investigated in section 4, followed by a discussion of the results and a conclusion in Section 5.

2 Methods and data

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2.1 Model description

2.1.1 GEM-MACH-GHG

GEM-MACH-GHG (Polavarapu et al., 2016) is a global GHG transport model, coupled with the meteorological model, wherein tracers are transported every time step. The horizontal resolution of the model is 0.9° using a global uniform latitude-longitude grid (400 × 200 grid points) and there are 80 vertical levels, spanning the surface to 0.1 hPa. For meteorology and tracer transport, a semi-Lagrangian advection scheme is used. Additionally, a global mass fixer was implemented for the transport of tracers in order to conserve the global mass of CO₂ during model forecasts. Kain and Fritsch (Kain and Fritsch, 1990; Kain, 2004) was implemented for convective transport of tracers through deep convection. More details about the model can be found in Polavarapu et al. (2016). While CO₂ is regarded as an inert trace gas in the model, methane (CH₄) and carbon monoxide (CO) utilise a simple parameterized climate chemistry. Specifically, the full troposphere chemistry package employed in GEM-MACH is replaced by simple hydroxide reactions related to oxidations of CH₄ and CO in the atmosphere, along with the conversion of CH₄ to CO.

As the operational version of GEM is updated periodically, model parameters are invariably tuned to optimize the performance of the model. In the previous configuration of GEM-MACH-GHG used in Polavarapu et al. (2016), thermal eddy diffusivity values within the PBL calculated by GEM were overridden to enhance vertical mixing of CO₂ concentration. A minimum value of 10 m² s⁻¹ was imposed within the PBL to prevent too little vertical mixing of CO₂ in boreal summer because low values resulted in spuriously low CO₂ concentrations on model levels near the surface in daytime when the magnitude of biospheric fluxes sinks is great (Polavarapu et al., 2016). In contrast, the lower limit imposed in the operational version of GEM-MACH is 0.1 m² s⁻¹ which was also empirically chosen for air quality applications. In this study, we use a more recent version of GEM which has better vertical mixing within PBL than the version used in the previous study. This improvement

allowed us to revise the thermal eddy diffusivity minimum imposed with the PBL to 1 m² s⁻¹ from 10 m² s⁻¹ for all simulations (with both global and regional models) conducted in this study because the previous value resulted in too low CO₂ concentrations at model levels near the surface over snow covered regions, e.g. Alberta and Saskatchewan, in boreal winter. The impact of the revised value in summer daytime is minimal, and some improvements are found in night time, making the diurnal cycle of modelled CO₂ concentrations more realistic overall.

Results from the global model with 0.9° horizontal grid spacing are used as the reference experiment for the verification of the newly developed regional model. However, to provide lateral boundary conditions (LBC) of CO₂ concentration and meteorology to the regional model, a higher horizontal resolution of 0.45° (800 × 400 grid points) is needed to avoid numerical instability in meteorological forecasts caused by drastic change of spatial resolution at the lateral boundary of the regional model domain. The original configuration is somewhat coarse to be used as LBCs for our regional model with 10 km horizontal resolution. Therefore, we also run the global model with a 0.45° horizontal grid spacing. All other configurations except horizontal grid spacing are the same with those used in coarse (0.9°) resolution global model.

2.1.2 Extension to regional domain

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For the regional model simulation, a rotated latitude-longitude map projection with approximately 10 km horizontal grid spacing and a hybrid vertical coordinate is used. The domain of the regional model covers most of Canada and the US, as shown in Fig. 1, and consists of 528 by 708 grid points. The number of vertical levels is the same as in the global model as described in section 2.1.1, namely, 80 levels spanning the atmosphere from the surface to 0.1 hPa. Since the number of grid points is also almost 5 times greater than that used in the 0.9° global model, the new regional model is more expensive to run. The physics packages used in the regional model are similar to those of the global model and GEM-MACH, and include radiation (Li and Barker, 2005), boundary layer mixing (Bélair et al., 1999), shallow (Bélair et al., 2005) and deep convection (Kain and Fritsch, 1990; Kain, 2004), orographic gravity wave drag (McFarlane, 1987) and nonorographic gravity wave drag (Hines, 1997a, b) schemes. More details are provided in Mailhot et al. (1998).

For a simulation with tagged tracers to distinguish each component of CO₂, e.g. those associated with biospheric, ocean and fossil fuel fluxes etc., a transport model should have the ability to simulate consistent masses across different components. In other words, the mass of the total CO₂ field should exactly equal the sum of the tagged CO₂ species, both globally and locally. This property is also required for estimating surface fluxes through Bayesian synthesis inversion (e.g. Enting, 2002). As already described in Polavarapu et al. (2016), the semi-Lagrangian advection scheme implemented in GEM alters mass slightly during model integration. The magnitude of the change for short range forecasts is negligible but this is not the case for the lengthy simulations of inert trace gases such as CO₂. To compensate for mass losses of tracers, a mass conservation scheme (Bermejo and Conde, 2002) and a shape preserving locally mass conserving scheme (Sørensen et al., 2013) were applied to tracer fields. At the lateral boundaries of the regional model domain, a mass restoration scheme (Aranami et al. (2015) scheme) is applied. These schemes, however, can make mixing ratios across multiple tracers inconsistent since they correct for global mass changes in local regions where tracer gradients are large. Since each tagged component has rather

different spatial structure and gradients from the total CO₂ field, the mass fixes made to the individual tagged variables need not be consistent with that made to the total field. As a result, the sum of each component may not equal the total CO₂ concentration field. To address this issue, the monotonicity and mass conservation schemes applied during the advection step are turned off in the regional model. The impact of the configuration on total mass of CO₂ within the regional model domain was compared with total mass of CO₂ from an experiment using mass related schemes turned on. The results show that there is no significant difference between two configurations in terms of total mass of CO₂ in the whole model domain as well as modelled CO₂ concentrations at the lowest model level in which most surface-based in-situ observation sites are located (not shown). This occurs because the majority of tracers' mass are injected into the regional model domain through its lateral boundaries. The mass of CO₂ from the surface flux is small compared to the total mass of CO₂ in the atmosphere of the regional model domain and the signal of surface fluxes exits the lateral boundaries during model integration before they reach the upper levels of the atmosphere (e.g. upper troposphere and stratosphere). Therefore, in the regional model, we obtain perfect "additivity" of the tagged components with negligible loss of mass conservation.

2.2 Surface flux

In this study, the optimised CO₂ fluxes from NOAA's CarbonTracker, version CT2016 (Peters et al., 2007, with updates documented at http://carbontracker.noaa.gov) were used as surface CO₂ fluxes for CO₂ simulations. The temporal resolution of the surface flux is 3 h. Because of their ready availability and careful validation, many studies aimed at global to regional to urban scales have used optimized fluxes from CarbonTracker for forward CO₂ simulations (Houweling et al., 2010; Ballav et al., 2012; Díaz-Isaac et al., 2014; Polavarapu et al., 2016; Li et al., 2017; Wu et al., 2018).

The original CT2016 flux product is available on 1° by 1° horizontal grid spacing. However, the global and regional models have different horizontal grid spacing. Thus, fluxes are re-gridded to GEM's grids with 0.9°, 0.45° and 10 km horizontal spacing grid, respectively, in a mass-conservative way. In addition, one more redistribution method is applied to re-gridded fluxes on the 10 km grid. This process applies a land-sea mask to the regional model grid in order to avoid unphysical modelled CO₂ concentrations caused by the different behaviour of vertical mixing over land and water grid cells. Because coarse resolution fluxes do not contain all the information needed for high resolution grid cells, considering only the size of a grid cell in re-gridding is insufficient because it would lead to too low or high modelled CO₂ concentrations relative to observed CO₂ concentrations in regions of strong surface CO₂ fluxes. With respect to fossil fuel emissions, for example, dynamic consistency is one of the important factors in regional scale CO₂ concentration simulation, in particular along coastal margins (Zhang et al., 2014). Hence, biospheric and fossil fuel flux components on water grid cells where the fraction of land is less than 30%, including lakes and oceans, are redistributed into land grid cells (within a radius of 30 grid points) in order to simulate realistic CO₂ concentrations along coastlines in the regional model domain while minimizing the impact of redistributed surface fluxes on CO₂ simulation and while conserving the total mass of surface fluxes within the regional model domain.

2.3 Observations

Modelled CO₂ concentrations are verified against observations from ObsPack (Masarie et al., 2014) which is maintained and provided by NOAA. For surface measurement sites, not all observations available in the regional model domain are used in the evaluation. The following selection criteria are applied: (1) sites were used to infer optimized CO₂ fluxes in CT2016. Thus, we can expect that optimized surface fluxes from CT2016 provide information about sources and sinks consistent with observed CO₂ concentrations at those sites so that differences in model simulation results may be attributable to model error, (2) sites have continuous measurements, (e.g. hourly data) as we want verify the results for all forecast hours, (3) sites have no periods of missing data longer than one month (except for ESP (See Table 1 for full list of station abbreviations) which has no data in January 2015) so results can be obtained for all seasons during experimental period. As a result, 19 measurement sites (11 sites in Canada and 8 tower sites in the US) are selected as shown in Fig 1 and listed in Table 1. For aircraft profiles of CO₂, measurement sites available over Canada and the US in the year 2015 were selected.

2.4 Experiment design

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Three experiments are performed as listed in Table 2. GLB90 is the reference experiment using the global model with 0.9° horizontal grid spacing. GLB45 which uses the global model with 0.45° horizontal grid spacing is carried out to provide LBCs to the regional model. LAM is the regional model run with 10 km horizontal grid spacing. The simulation period is one year for 2015. In the analysis, the first 10-days of simulations are regarded as a spin-up period and discarded.

Figure 2 depicts the global and regional model cycles. In each forecast, the weather forecast and CO₂ transport by forecasted wind fields are performed simultaneously in every time step. For the initial condition (IC) of meteorological fields for the two global models at the beginning of every cycle, the operational global analysis products from the global deterministic prediction system (GDPS; Buehner et al., 2015), whose horizontal resolution is roughly 25 km on a regular lat-long grid or, as of 15 December 2015, a vin-yang grid (Oaddouri and Lee, 2011) are used. The archived data are interpolated to our lowresolution grids and topographies of GLB90 and GLB45, separately. For the IC of meteorological fields for the regional model, the operational regional analysis products from the regional deterministic prediction system (RDPS; Fillion et al., 2010; Caron et al., 2015) are used. The regional model grid is a subset of the model domain of the operational RDPS with the same horizontal resolution, sharing grid points on the same latitudes and longitudes. Therefore, it is not necessary to perform a horizontal interpolation at the start of every cycle. Also, a spin-up period for the meteorological forecast is unnecessary. Both operational global and regional meteorological analyses are produced 4 times per day with a 6 h assimilation window centred on the analysis time. We only use analyses produced on 00:00 UTC every day as an IC of meteorology. Thus, a 24-hour weather forecast is produced during each 24 h CO₂ cycle and these forecasts are replaced by new analyses every 00:00 UTC, with the exception of microphysics tracers which are retained to allow a hot start for hydrometeor fields (Milbrandt et al., 2016). On the other hand, the mass of CO₂ in a model grid volume is kept during cycles without replacements. The 24 h forecast of CO₂ from the previous cycle is used as the IC of the CO₂ field for the next cycle at 00:00 UTC. This is combined with the updated meteorological analysis for a complete initial state for the coupled model. Such 24 h forecast cycles are also used in other global model systems (e.g. Agustí-Panareda et al., 2014; Ott et al., 2015).

The IC of 3-D atmospheric CO₂ concentrations at the beginning of all three experiments is taken from CT2016 CO₂ concentrations at 00:00 UTC 1 January 2015. The LBC of CO₂ concentrations for the regional model are obtained from GLB45 and include hourly meteorological and CO₂ fields.

One more possible configuration is that of using operational RDPS forecasts as meteorological LBCs for the regional model, which is similar to the configuration of the operational regional GEM-MACH (Moran et al., 2010). We tested this configuration and compared modelled CO₂ concentrations with the LAM experiment's modelled CO₂ concentrations. Negligible difference was found (not shown), therefore we decided not to include that configuration in this study because our purpose is in developing an integrated global/regional forward modelling framework for GHG simulation as shown in Fig. 2.

2.5 Sampling method and metrics

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In order to evaluate the performance of CO₂ simulations and meteorological forecasts, a series of metrics are used as described below. Modelled or forecasted values are sampled at the observed location by applying horizontal and vertical interpolation to model fields rather than selecting the nearest grid point to measurement locations, and selecting the time step closest to observed time. To sample modelled CO₂ concentrations, the sampling height above the ground level (or the model surface level) is considered to determine the altitude for vertical interpolation instead of using the actual sampling height above sea level (i.e. the sum of the altitude of an observation site plus intake height). Coarse horizontal resolution models cannot resolve well the complex topography (e.g. mountain regions) around some measurement sites. As a result, the altitude of model topography may be far above or below the actual height of a measurement site. If the height of a model-sampled observation is erroneously placed in the PBL (free troposphere) as a result of coarse model topography, this can result in an unphysical, too strong (too weak) diurnal cycle of CO₂ compared to observed values (see Agustí-Panareda et al., 2019). From a comparison of the two sampling methods, it was found that the LAM experiment is not sensitive to the vertical sampling method, as expected, because it can resolve actual topography well thanks to the higher spatial resolution, but the GLB90 experiment is sensitive to the method at a number of measurement sites (not shown). Thus, in order to reduce the topography mismatch problem in the coarse resolution global model and investigate the impact of higher horizontal resolution without this problem, the method using intake height is used to help the global model capture the behaviour of the PBL variations with time and height. A detailed discussion of the vertical sampling methods in connection with horizontal resolution is found in Agustí-Panareda et al. (2019).

To analyse our results, including CO_2 and meteorology, bias and standard deviation of forecast error (STDE) are used. The bias is defined as

Bias =
$$\bar{X} = \frac{1}{N} \sum_{i=1}^{N} (M_i - O_i),$$
 (1)

where N indicates the number of observations, M_i indicates modelled CO₂ concentration or meteorological forecast and O_i indicates the corresponding observation.

The STDE is defined as

STDE =
$$\sqrt{\frac{1}{N} \sum_{i=1}^{N} (X_i - \bar{X})^2}$$
, (2)

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 $X_i = M_i - O_i$ and the overbar refers to the bias of the quantity.

To calculate the amplitude of CO₂ diurnal cycle (or other frequencies) for a measurement site or grid point, we use a Discrete Fourier Transform (DFT) technique. The linear trend in hourly CO₂ time series from a specific location is removed first, then the DFT technique is applied to the detrended CO₂ time series to extract the amplitude of CO₂ variability across temporal scales, from synoptic to diurnal to sub-daily scales, as discussed in Section 4.4.

3 Model evaluation

3.1 Evaluation of meteorological fields

Before considering CO₂ simulation results, weather forecasts from the three experiments are verified against observations over the regional model domain and are compared with each other. The motivation for doing this is two-fold: (1) to check that the meteorological forecasts from our regional model have not drifted away from the operational forecasts which have been produced and maintained by the CMC for many decades and (2) to compare the regional model results with the global model results. The first check is necessary because the configuration for weather prediction and the GEM model version used in this study are different from what was used to produce the operational forecast in 2015. For example, LBC in RDPS were obtained from a global model forecast using a 33 km horizontal grid spacing (Caron et al., 2015) (or with a 25 km horizontal grid spacing as of 15 December 2015), with a different model domain extent and vertical coordinate. As shown by Polavarapu et al. (2016), the performance of the weather forecast by the global model is already well evaluated. The uncertainty of 24 h weather forecasts in the global model corresponding to GLB90 experiment in this study is comparable with those of reanalyses provided by three operational centres; monthly and zonal means of fields in 2009 and 2010 are within acceptable range on global scales. Thus, in this section, we focus on the regional model's results and on differences between experiments on the regional domain.

Figure 3 shows the bias and STDE of 24 h forecast error of the three experiments for vertical levels from 1000 hPa to 10 hPa in July 2015. The same numbers of North American radiosonde observations are used in each of the three sets of verifications and these are indicated in the right of each panel. Statistical significance of the differences using a T-test for the means and an F-test for the standard deviations with the 95% confidence level were computed but not shown explicitly. However, the discussion below uses this information in that we only mention results that are statistically significant. The three experiments show good agreement with observations in terms of bias and STDE. For zonal wind, there are quite small

differences among experiments and the scores remain within the range of operational forecasts, except at 925 and 850 hPa where biases in the GLB90 and GLB45 experiments are slightly better than those in the LAM experiment (Fig. 3a). For wind speed, unlike the zonal wind, forecasts in LAM experiment are better than those from the GLB90 experiment for levels from 925 hPa to 50 hPa and better than those in the GLB45 experiment for levels from 300 hPa to 70 hPa and at 700 hPa (Fig. 3b). For geopotential height, the forecasts in the LAM experiment are better than those in the GLB90 and GLB45 experiments from 400 hPa to 10 hPa where relatively large positive biases in GLB90 and GLB45 experiments exist. In addition, the STDE in the LAM experiment is better at all vertical levels except a few levels (Fig. 3c). For temperature, the forecasts in the LAM experiment are better than those in the GLB90 and GLB45 experiments from 1000 hPa to 70 hPa with the exception of 150 hPa (Fig. 3d).

The scores for December 2015 are shown in Fig. 4. The differences in bias and STDE among experiments are smaller than those in July. In addition, patterns in the reduction of bias and STDE from coarse horizontal resolution to higher horizontal resolution can be seen much more clearly than in July, which means that the values in the GLB45 experiment are located between those of the GLB90 and LAM experiments. For zonal wind, there are quite small differences in bias and STDE among experiments as was the case in July (Fig. 4a). For wind speed, forecasts in the LAM experiment are better than those in the GLB90 experiment from 850 hPa to 150 hPa and those in the GLB 45 experiment at 500 hPa and 400 hPa, but not better at 925 hPa (Fig. 4b). For geopotential height, both bias and STDE in the LAM experiment are better than those in the GLB90 experiment at most pressure levels except from 700 hPa to 400 hPa, while the LAM experiment is better than GLB45 from 1000 hPa and 925 hPa (Fig. 4c). For temperature, the bias in the LAM experiment is better than that in the GLB90 and GLB45 experiments from 925 hPa to 250 hPa (Fig. 4d).

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It is also worth considering how our meteorological forecasts compare to those of other systems. Agustí-Panareda et al. (2019) show RMSEs of vector wind for January and July 2014 from 1 d forecasts from the Copernicus Atmosphere Monitoring Service (CAMS). Our RMSE scores computed using the data from Figs. 3 and 4 for wind speed are shown in Table S1 and these can be compared to their Fig. 4. Our LAM scores are lower than those of the 9 km CAMS at all heights in January and July. However, this is not a fair comparison since their scores are for global domain whereas we consider the North American domain, and their values are for 2014 but ours are for 2015. Nevertheless, the comparability of the scores further suggests that our LAM is performing well in terms of 24 h meteorological forecasts.

The number of available observations at 1000 hPa is much smaller than that of other pressure levels (see numbers in right side of each panel in Fig. 3 and 4) because typical altitudes of many sites are above the level corresponding to 1000 hPa and surface pressures may be below 1000 hPa depending on the synoptic situation, there is little confidence in the verification at this level by means of radiosondes. A better approach to rigorously investigate the performance of weather forecasts at lower levels is to use surface observations because of their much greater numbers (in both space and time). Therefore, weather forecasts in the three experiments are also verified against observations near the surface. Figure 5 shows bias and STDE of sea level pressure, 2-m temperature and 10-m wind speed as well as the Heidke Skill Score (HSS) (Wilks, 2006) of 10-m wind speed for July 2015. The STDE of sea level pressure in the LAM experiment is lower than those in the GLB90 and GLB45

experiments, while the bias of sea level pressure in the LAM experiment is slightly lower than those of the GLB90 and GLB45 experiments. However, the difference between the LAM and the GLB90 and GLB45 biases does not exceed 0.5 hPa (Fig. 5a). The STDEs of 2-m temperature and 10-m wind speed in the LAM experiment are smaller than those in GLB90 and GLB45 experiments for all forecast hours (Fig. 5b and c), which implies that the error of forecasts from the LAM experiment fluctuates less than those of the GLB90 and GLB45 experiments. Also, the better results of the LAM experiment in 10-m wind direction is evident in the higher HSS of the LAM experiment (Fig 5d). Higher HSS means a better forecast of wind direction. In addition, root-mean squared errors (RMSEs) of variables in the LAM experiment are lower than those of the GLB90 and GLB45 experiments (not shown).

In December 2015, the better forecasts in the LAM experiment compared to those of the GLB90 and GLB45 experiments can be seen more clearly (Fig. 6). The bias and STDE of each variable in the LAM experiment are lower than those of GLB90 and GLB45 experiments at most forecast hours (Fig. 6a-c), and higher HSS of 10-m wind direction are evident at all forecast hours (Fig. 6d).

In summary, the LAM experiment produces reasonable meteorological forecasts in comparison with meteorological observations and better results relative to the GLB90 and GLB45 experiments, in particular at surface levels which are important for correctly capturing the flow of CO₂ affected by surface fluxes and boundary layer mixing, with reductions in both bias and STDE. Since forecasted meteorological fields are used to transport CO₂ in each simulation individually, better CO₂ simulations in the LAM experiment can be expected in the verification of modelled CO₂ concentrations.

3.2 Evaluation of CO₂ fields

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The CO₂ fields in the LAM and other experiments are investigated in terms of monthly bias and STDE of daily afternoon (12:00-16:00 LST) modelled CO₂ concentrations at the measurement sites shown in Fig. 1 and listed in Table 1 (Fig. 7). Daily afternoon time was selected because this is what CT2016 used to estimate surface CO₂ fluxes. Also, CT2016 results are included as a reference since this is what all our model experiments used as input fluxes. In general, bias and STDE in summer are larger than in other seasons at most sites except BAO, SCT, WGC and WKT. Better results in CT2016 than in all three experiments at many sites can be seen, especially in June to October. Surface CO₂ fluxes from CT2016 were inferred by minimizing the difference between observations and forecasts of TM5 (Krol et al., 2005) which is the transport model used in CT2016, reflecting the signature of TM5's transport which may not match with GEM-MACH-GHG's transport (Polavarapu et al., 2016). As a result, larger biases in the three experiments relative to CT2016 at the above mentioned sites may be expected due to the discrepancies of modelled CO₂ concentrations between CT2016 and GEM-MACH-GHG. In contrast, the three models show similar biases to each other, except at ESP and WGC especially in boreal summer. Since the LAM experiment uses LBCs of CO₂ concentrations from the GLB45 experiment, information about large scale transport of CO₂ concentration is reflected in LAM experiment.

Now we consider the seasonal variation of the performance of the LAM model over the regional (North American) domain. The seasonal bias and STDE of modelled CO₂ concentrations in the LAM experiment are shown in Fig. 8, based on

daily afternoon CO₂ concentrations. In boreal winter (DJF) and spring (MAM), there are mainly positive biases at most Canadian sites, while STDE are small relative to other sites in the US. The magnitude of surface CO₂ fluxes in those seasons over Canada is quite small (not shown), and thus bias at the Canadian sites contributes little to the overestimation of CO₂ concentrations. This result suggests that biases included implicitly in the LBC of CO₂ concentrations provided by the GLB45 experiment is more important in determining the biases in the regional model domain in those seasons. Four sites (BRA, EST, ETL and LLB) in Alberta and Saskatchewan provinces show relatively large STDE in DJF due to local influences of surface fluxes trapped within a shallow boundary layer by low temperatures. On the other hand, in boreal summer (JJA), large STDE can be seen with negative biases at most sites. The large biases and STDE in JJA may be attributed to errors in terrestrial CO₂ fluxes within the regional model domain. As shown in Fig. 7, the GLB90 and GLB45 experiments underestimate CO₂ concentrations over northern sites in JJA. That is also reflected in the underestimation of CO₂ concentrations in LAM experiment because the same surface fluxes are used in the simulations. Finally, in boreal autumn (SON), both biases and STDE show moderate values between MAM and JJA. Negative biases at northern sites in LAM experiment is partially due to biases in LBCs obtained from the GLB45 experiment. In summary, the performance of the regional model partially depends on biases in the global model which provides the LBC, and the relative importance of these biases varies with season. In this regard, the use of CT2016 posterior fluxes to drive our global models exacerbates such biases. However, in the future when our global models provide their own flux estimates, such biases may be reduced. Furthermore, the need for a better understanding of the relative role of initial and boundary conditions and surface fluxes in controlling CO₂ distributions within the regional model's domain is evident. This is the subject of our future work.

4 The impact of horizontal resolution on CO₂ simulation

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Since our regional model requires more computational resources (due to the greater number of grid points and shorter time step) than our global (GLB90) model, it is important to consider the added benefit of the higher horizontal resolution on CO₂ simulations. In this section, modelled CO₂ concentrations from the three experiments are analysed from the perspective of spatial patterns and vertical profiles of CO₂ concentrations and the reproducibility of temporal patterns against atmospheric CO₂ observations.

25 4.1 Spatial patterns of surface CO₂ concentrations

Biases between two experiments are compared pairwise for four seasons (Fig. 9). Three comparisons are shown because three experiments are conducted. Since bias can have both positive and negative values, absolute bias is used in the calculation. Blue (red) colour means the higher horizontal resolution model simulated smaller (larger) absolute bias compared to coarser one. In DJF, the GLB45 and LAM experiments are better than the GLB90 experiment. However, the LAM experiment is not better than GLB45 experiment at U.S. sites except WGC and BAO. In MAM, the differences among the experiments are the smallest, except at ESP near the west coast of North America. Differences between the LAM and GLB45 experiments at northern

Canadian sites are quite small in DJF and MAM, which is associated with weak surface CO₂ fluxes in those seasons. In JJA, the reduction in bias resulting from the higher horizontal resolution model can be seen clearly and the magnitude of reduction is higher probably due to better weather simulation (less transport errors) as shown in Figs. 3, 4, 5 and 6. The LAM experiment shows better results than both the GLB90 and GLB45 experiments at most sites. On the other hand, the GLB45 experiment is not better than the GLB90 experiment except at a few sites. This nonlinearity of the improvement with increased resolution is consistent with the results of Agustí-Panareda et al. (2019) although their conclusions are based on RMSE rather than absolute bias. In SON, the results are similar to those in JJA, namely, the LAM experiment is better than both the GLB90 and GLB45 experiments at most sites except ESP. At many sites, the benefit of finer grid spacing is evident, but higher horizontal grid spacing does not always guarantee a lower magnitude of bias for all sites. Part of the reason that improvement is not clear at certain sites in Figure 9 may be due to the focus on afternoon mean values. For example, if we consider higher temporal frequency output (i.e. hourly residuals), the LAM is better than GLB90 and GLB45 even at ESP in November (Fig. S1).

Figure 10 shows the differences of STDE between two experiments. The spatial pattern of differences of STDE is different from that of bias. More blue dots are evident indicating that the STDE of higher horizontal resolution model is smaller than that of the coarser horizontal resolution model at most sites. Specifically, the LAM experiment shows better results than both the GLB90 and GLB45 experiments in DJF. As shown in Fig. 6c and d, better forecasts of 10-m wind speed and direction at screen level in the LAM experiment should help to reduce STDE in LAM experiment relative to the two global model experiments. In MAM, the impact of horizontal resolution is very small at most sites except near the southern boundary of the regional model domain due to the weak magnitude of surface CO_2 flux in this season. The ratio between CO_2 concentrations resulting from surface CO_2 fluxes within the regional model domain and background CO_2 concentrations from the GLB45 experiment shows that the contribution of surface CO_2 flux is least in MAM due to the small magnitude of surface CO_2 flux (not shown). In JJA, on the other hand, the magnitude of the difference is larger than in other seasons in both positive and negative directions. Finally, in SON, the improvement due to finer grid spacing can be seen.

In summary, the difference of bias and STDE between experiments provide evidence of improvement in CO₂ simulations due to finer horizontal resolution and better wind forecasts near the surface in the LAM experiment. The pattern of the differences are strongly associated with the spatial and seasonal patterns of the magnitude of surface CO₂ fluxes used in simulations.

4.2 Vertical profile of CO₂ concentrations

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We now consider the quality of modelled CO₂ concentrations in the free troposphere. Observed profiles of CO₂ can reveal the signatures of vertical mixing, so they can be used to measure the performance of transport models (Lin et al., 2006). The seasonal bias and STDE of vertical profiles of modelled CO₂ concentrations against NOAA aircraft profiles (Sweeney et al., 2015) over sites in Canada and the US inside the regional model domain are shown in Fig. 11. Modelled CO₂ concentrations are sampled at the exact location and height of observations by applying vertical and horizontal interpolation to 3-D model fields at a time step close to observed time. Then, averages over all profiles of modelled and observed values for a season are

binned into 1 km thick layers. The three experiments generally overestimate CO₂ concentrations in DJF (at 1000 m) and MAM and underestimate them in JJA and SON, which is consistent with the comparisons against surface CO₂ measurement sites shown in Fig. 8. The magnitude of the bias does not exceed about 2 ppm in any altitude or season, and it decreases with altitude. Profiles of CO₂ concentrations near the ground are difficult to simulate due to the strong influence of surface fluxes (Geels et al., 2007). The range of the biases in the three experiments are similar to that seen in our previous study (Polavarapu et al., 2016) as is the direction of the biases. Specifically, the bias changes sign with height, with positive biases at low altitudes and negative biases in DJF and MAM. In JJA and SON, the bias remains negative at almost all heights. The LAM experiment generally has the smallest biases for all seasons and altitudes, in particular, below 4000 m in JJA when the influence of surface CO₂ fluxes is significant through active vertical mixing. Lower wind speeds in boreal summer compared to other seasons causes accumulation of surface fluxes over North America in the lower 4000 m (Sweeney et al., 2015). Reduced bias and STDE of forecasted temperature profiles in the LAM experiment in July (Fig. 3d) may help to improve vertical advection in the LAM simulations relative to the global model experiments through improved buoyancy calculations. This may explain why the LAM experiment has a better ability to simulate vertical profiles of CO₂.

4.3 Temporal patterns

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We evaluate modelled CO₂ concentrations at various temporal scales including synoptic variability and the diurnal cycle. First, synoptic variability of modelled CO₂ concentrations is analysed. Figure 12 shows Taylor diagrams (Taylor, 2011) of modelled CO₂ concentrations in the afternoon compared with observations. Since the domain of the LAM experiment covers a variety of geographic regions across Canada and the US, including mountain, continental and coastal sites, the synoptic variability of CO₂ is not expected to be captured well at all sites. In DJF, the variability of modelled CO₂ concentrations in the LAM experiment is closer to the observed variability than that captured in the GLB90 and GLB45 experiments in accordance with decreased STDE seen in Fig. 10 (Fig. 12a). In MAM, the variability of modelled CO₂ is scattered with relatively lower correlations than other seasons (Fig. 12b). In general, due to the onset of growing season in MAM, transport models tend to produce lower correlations with observed CO₂ (Geels et al., 2004; Pillai et al., 2011; Agustí-Panareda et al., 2014). In JJA, despite having larger biases than in other seasons (Fig. 7), correlations are quite reasonable lying mostly between 0.6 and 0.95 (Fig. 12c). However, the variability in the CO₂ concentrations tend to be overestimated. This could be mainly due to the large uncertainty in biospheric fluxes (Patra et al., 2008). Also, the range of correlations is the biggest -- between approximately 0 and 0.95. In SON, the synoptic variability of CO₂ is well captured by all experiments (Fig. 12d). Many sites have correlations higher than 0.9, standard deviations similar to observed variability, and the least normalised RMSE (the distance from the reference point on the x axis) relative to other seasons. We expect the LAM experiment to produce higher correlations and smaller normalised RMSEs, and normalised standard deviations approaching 1. Indeed, the LAM experiment tends to simulate well the observed variability of CO2 and it produces smaller normalised RMSE relative to the GLB90 and GLB45 experiments to some extent although the results vary according to site, and each experiment shows similar seasonal patterns which are driven by the weather forecasts and surface fluxes.

Thus far, modelled CO₂ concentrations in the afternoon time (12:00-16:00 LST) have been analysed. Henceforth, data at all times of the day and night are retained. Figure 13 shows the mean diurnal cycle of modelled and observed CO2 concentrations for July and December at WGC where the most significant differences among the three experiments are observed. In general, the three experiments simulate similar CO₂ diurnal cycles for other sites (not shown). Three sampling levels data are available at WGC in 2015. CT2016 is included as well for comparison purposes, but only results at the highest sampling level are shown because only observations at this level were used in the inversion in CT2016. The LAM experiment captures the CO₂ diurnal cycle well, but the GLB90 and GLB45 experiments and CT2016 do not, especially in July (Fig. 13a, c and e). At the sampling level of 483 m, the GLB90 experiment overestimates morning time CO₂ concentrations and CT2016 overestimates night time CO₂ concentrations in July, while the GLB45 and LAM experiments capture the diurnal cycle (Fig. 13a) relatively well. This level (483 m) has a comparatively weak diurnal cycle because it is mostly decoupled from the surface at night and daytime enhancements are significantly diluted relative to lower levels. At lower sampling levels, 91 m and 30 m, both the GLB90 and GLB45 experiments overestimate night time CO₂ concentrations in July, whereas the LAM experiment captures both day and night time CO₂ concentrations well (Fig. 13 c and e). This greater sensitivity to model resolution at night was also seen by Agustí-Panareda et al. (2019). WGC is located in a valley between two mountain ranges. The model topographies of GLB90 and GLB45 do not resolve this geography well due to their coarse horizontal resolutions. In contrast, the LAM experiment resolves the actual topography around the WGC site well relative to the two global models. In daytime, CO₂ concentrations are well simulated in the LAM and GLB45 experiments due to the strong vertical mixing (Fig. 13a, c and e). In contrast, accumulated CO₂ in night time still remains in the afternoon time in the GLB90 experiment, leading to an overestimation of CO₂ in the afternoon at all sampling levels. In December, the LAM experiment simulates slightly better CO₂ concentrations and its standard deviation at all sampling levels, while the GLB90 and GLB45 experiments underestimate CO2 concentrations (Fig. 13b, d and f).

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In order to analyse CO₂ time series across various temporal scales beyond the diurnal cycle, the DFT method explained in Section 2.5 is applied to hourly CO₂ time series. Figure 14 shows the amplitude of hourly CO₂ concentration time series across different temporal scales from 2 h to 92 days for the period from June to August 2015 at the LEF and WGC sites. Unfortunately, not all sites have hourly observations without missing values for the year 2015. These two sites have hourly data available for three months from June to August 2015 without missing values and, fortunately, reveal different properties. Thus, they were selected to illustrate the impact of increased horizontal resolution on CO₂ simulations on the time scales captured by the models. At LEF, one sampling level, 396 m, satisfies our constraint of no missing data, and, at WGC, two sampling levels, 483 m and 91m, meet this constraint. At the LEF site, the three experiments capture well the signals across all temporal scales in observed CO₂ time series, including synoptic and diurnal variations (Fig. 14a and b). The topography mismatch of the GLB90 and GLB45 experiments are relatively small around at the LEF site. The intake height of measurements at LEF is 396 m above the ground at which laminar flow is more dominant than turbulent flow in night time so that the respiration signal from surface does not reach the free troposphere and synoptic variability is more dominant (Davis et al., 2003; Wang et al., 2007). As a result, the differences of amplitude in the three experiments are less than about 0.8 ppm

for all temporal periods (Fig. 14b). At the WGC site, on the other hand, as already shown in Fig. 13, the GLB90 experiment simulates too strong diurnal cycles of CO₂ at lower sampling heights in July, which can be seen clearly as well in Fig 14c-f, with largest overestimation at the lowest sampling level (Fig. 14 e and f). However, it is not just the diurnal cycle of CO₂ from June to August that is overestimated in the GLB90 experiment. Periods from sub-diurnal to longer day periods are also overestimated (Fig. 14c-f). Furthermore, while GLB45 performs better than GBL90, it also overestimates the diurnal cycle amplitudes and longer time scales at 483 m and 91m. Hence, the larger mismatch of topography results not only in inaccurate daily time scales but also other scales such as synoptic scales longer than 4-days. A similar result was also found for time periods for 92 to 300 days (Fig. S2). The amplitude of the diurnal cycle can also be computed in model space to illustrate its spatial variability as a function of model resolution (Fig. S3). With the same prescribed fluxes, greater spatial heterogeneity of diurnal cycle amplitude occurs with increased resolution. However, the validation of these finer spatial scales requires a dense observation network and is not possible at present.

5 Discussion and conclusions

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We have developed a regional atmospheric transport model for GHG gas simulation, as an extension of GEM-MACH-GHG which is ECCC's global atmospheric transport model for GHG simulation. The regional model shares much of the configuration of the global model, while its model domain is focused on Canada and the U.S. One gain from using the same vertical coordinate in both the regional and global models is that there is consistency at lateral boundaries of the regional model domain. CO₂ simulations using the same surface CO₂ fluxes from CT2016 are performed with three configurations of 2 global models and 1 regional model in order to assess whether the newly developed regional model is working properly and to assess the benefit of the regional model over the global model in terms of weather forecasts and CO₂ simulations. In a given experiment, a series of 24 h forecasts are replaced by operational analyses every cycle and used to transport CO₂ every time step, whereas transported CO₂ fields are not replaced but are kept during each 1-year simulations.

Meteorological forecasts in three experiments are verified against North American radiosondes and surface observations at screen level. All experiments show acceptable ranges of bias and STDE compared to observations. Overall, meteorological forecasts in the regional model show better results than both global models, especially in wind speed and direction at screen level which are of particular importance for CO₂ transport near the surface. We demonstrate the improvement of weather forecasts with increasing of horizontal resolution, which is most apparent in boreal winter. In addition, good quality meteorological forecasts in the global model are also required for providing meteorological LBCs to the regional model with reduced errors at large scales. Indeed, the GLB45 experiment can provide good quality of meteorological LBCs to the regional model every hour which is more frequent than when using reanalyses that are available at 3 h or 6 h intervals.

While the meteorological forecasts from the higher resolution region model are demonstrably better than those of the coarser resolution global models, demonstrating improved CO₂ simulations with higher resolution is more challenging. For example, the impact of biases in the LBCs provided by the GLB45 experiment on CO₂ simulations near the Arctic region in

the regional model is large, especially in boreal spring. In a regional scale inverse modelling system, estimated fluxes within the regional model domain are strongly influenced by the inflow of CO2 from the global transport model through lateral boundary (Schuh et al., 2010). Because LBCs of CO₂ include information of sources and sinks outside of the regional model domain, correct information at the lateral boundary is important to determine the sources and sinks in the regional model domain (Gerbig et al., 2003). As discussed in Polavarapu et al. (2016), GEM has different transport behaviour from the transport model used in CarbonTracker, in particular over the Arctic region, as seen in time series of CO2 concentrations and column-averaged CO₂. Thus, our models are not expected to perform better than CT2016 because we use surface CO₂ fluxes inferred by an inversion framework using a different transport model which has different transport behaviour. That is why our focus in this work is in the comparison of our regional and global models. We are able to find some benefits of our regional model over our global model when looking at the diurnal cycle of CO2 concentrations at particular sites in which large topography mismatches exist, e.g., WGC. Our global models did not capture diurnal cycles well, while our regional model did. This is a promising result because it suggests that using night time data in an inversion to estimate night time fluxes (e.g. Lauvaux et al., 2008) may be beneficial if a high resolution model is used. Currently, a GHG state estimation system using GEM-MACH-GHG and ECCC's operational Ensemble Kalman filter data assimilation system (Houtekamer et al., 2014) is under development. When posterior fluxes become available from our global model, this will alleviate the issue of model transport error mismatches with CarbonTracker. However, we will still have transport error, which is one of the biggest sources of posterior uncertainties in an inversion (Schuh et al., 2019). To address this issue, we plan to use multiple sources of meteorology to better account for transport error in posterior flux and uncertainty estimates.

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The regional model produces lower STDEs of CO₂ at surface measurement sites, in line with its lower STDE of meteorological forecasts. With respect to aircraft CO₂ profile comparisons, clear improvement of profiles of modelled CO₂ in the LAM experiment occurs at altitudes lower than 4000 m in boreal summer. Although the regional model domain is vast enough to include most of Canada and the U.S. so as to be able to estimate national to provincial scale surface GHG fluxes at finer spatial resolution via inverse modelling in the future, it is not easy to obtain better results everywhere. For example, at the ESP site located on the coastline of Vancouver Island, British Columbia, the LAM experiment does not have a lower bias of modelled CO₂ than the GLB90 and GLB45 experiments in MAM and SON. Nonetheless, the overall performance of CO₂ simulations by the regional model is better than our global models. It is well known that only afternoon time CO₂ concentrations are typically used in inversions due to the difficulty in capturing boundary layer evolution in most global transport models (Law et al., 2008; Patra et al., 2008). Noticeable improvement in reproducing the CO₂ diurnal cycle by the regional model can be seen at WGC which is located in complex terrain. Reduced topographic mismatch in the finer horizontal resolution model is the major driving force behind reduced sampling and representation error. This effect is not limited to just the diurnal cycle but also occurs for synoptic variability of CO₂ at the level where large scale motions are dominant, and even more so at lower sampling levels near the surface. In addition, the potential benefit in reproducing detailed diurnal cycles over regions with complex terrains hypothesized here is consistent with the findings of Agustí-Panareda et al. (2019).

Previous studies comparing high and low horizontal resolution transport models for CO₂ simulations concluded that some advantages can be attained by using higher horizontal resolution (Geels et al., 2007; Pillai et al., 2010; Díaz-Isaac et al., 2014). For example, better resolved amplitude and phase of short-term variability of CO₂ (Geels et al., 2007), reduced representation errors (Pillai et al., 2010) and smaller-scale structures of modelled CO₂ that are more sensitive to the distribution of CO₂ fluxes (Díaz-Isaac et al., 2014) were attained by using higher spatial resolution of transport model. Indeed, we also find similar results as mentioned above, but these advantages from the regional model experiment are not obtained at every observation site (Fig. 9 and 10). Basically, increasing horizontal resolution gives some positive impact to some extent but it generally has a mixed impact in this study. Part of the reason may be due to the fact that our models are variants of the same model but with different grid spacing and/or domain. Furthermore, the same coarse resolution surface fluxes were used with all models and this limits the potential for improvement (Remaud et al., 2018). In addition, the global model configurations used in this study already have relatively higher horizontal resolutions (0.9° and 0.45°) compared to other coarse resolution global transport models (e.g. Geels et al., 2007) and they all use the same number of (80) vertical levels as the regional model. Another major difference is that our global model is not an offline transport model which generally uses reanalyses as a meteorological driver for transport. Instead we take advantage of operational analyses to initialize weather forecasts every day and produce weather forecasts at every model time step. A major limitation in validating the overall improved ability to capture fine spatial scales may simply be due to the current sparsity of verifying observations of CO₂. With vastly greater numbers of verifying observations, the meteorological simulations are demonstrably better with increased resolution. Since the regional model can better simulate the spatial heterogeneity of the diurnal cycle of CO₂ in model space (Fig. S3), better observational density is needed to validate the performance of CO₂ simulations in the regional model in more detail.

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While this work has focused on the benefit of our higher resolution regional model over our global model for CO₂ simulation, both models are "online" in that the meteorology is coupled to the tracer transport every time step. An interesting question that was not addressed here is the impact of increased horizontal resolution in the context of an "offline" transport model which ingests meteorological analyses or reanalyses from another model (e.g. Kjellström et al., 2002; Geels et al., 2004, 2007). Additional errors arise due to spatial and temporal interpolation from another model's grid to the offline model's grid then arise.

A limitation of this study is the use of coarse resolution surface CO₂ fluxes in conjunction with the fine horizontal grid spacing of the regional model. For better simulation of CO₂, not only high-quality meteorological forcing but also high resolution prescribed surface fluxes are demanded (Locatelli et al., 2015). Higher spatial and temporal resolution fluxes could lead to better simulation of CO₂ concentrations (Feng et al., 2016; Lin et al., 2018) if the fluxes have correct space and time information about the distribution of sources and sinks of CO₂ fluxes. The challenge is in obtaining high spatial and temporal resolution surface fluxes that are accurate. One way to deal with this issue is to model biogenic fluxes explicitly at the same horizontal resolution as the transport model (e.g. Agustí-Panareda et al., 2019). Indeed, this is an avenue we plan to investigate in the future. Preliminary investigations with a high resolution anthropogenic flux product revealed improved comparisons to observations at some sites but degradation at other sites. For that reason, we chose to start of investigation of the regional

model by using fluxes with the same resolution as the global model and limiting the potential benefit of high resolution to improved meteorological depictions.

The LBCs of CO₂ from the global model plays an important role, as shown in Fig. 8, dominating the bias in the regional model when the magnitude of surface flux is weak. In addition, the LBCs of meteorology also play an important role in CO₂ simulations. For example, the meteorological IC and LBC contribute to the variability of daytime CO₂ in the PBL (Díaz-Isaac et al., 2018). Thus there is a need to better understand the relative importance of initial conditions, boundary conditions and surface fluxes on the performance of the regional model in order to better characterize these components of CO₂ model error within the regional domain. Indeed, the predictability of CO₂ on the regional domain and the relative role of initial and boundary conditions and surface fluxes on model error is a topic that is currently under investigation.

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There are a number of extensions to this work that are envisioned. For example, the newly developed regional model is not limited to CO₂ simulations but also includes other greenhouse gases such as CH₄. Thus, a separate validation of the regional model's ability to simulate CH₄ is planned. The regional model can also be utilized to provide information (e.g. IC and LBC) to urban scale forward or inverse modelling systems (e.g. Feng et al., 2016; Pugliese et al., 2018; Ishizawa et al., 2019). Lastly, and most importantly, an inverse modelling system for estimating surface CO₂ fluxes is being developed using the new regional GHG transport model to better understand the carbon cycle in Canada at finer spatial and temporal scales.

Code and data availability. The GEM-MACH-GHG model source code is publicly available at https://zenodo.org/record/3246556 (Neish et al., 2019) under the GNU Lesser General Public License version 2.1 (LGPL v2.1) or ECCC's Atmospheric Science and Technology license version 3. The model output data are available at http://crd-data-donnees-

rdc.ec.gc.ca/CCMR/pub/2019_Kim_GMD_Canadian_atmospheric_transport_model_for_simulating_greenhouse_gas_evolution_on_regional_scales/.

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Table 1: Information of surface in-situ measurement sites used in this study.

	Code	Station name	Latitude (°)	Longitude (°)	Altitude (m a.s.l.)
1	AMT	Argyle, Maine	45.0345	-68.6821	53
2	BAO	Boulder Atmospheric Observatory	45.03	-105.004	1584
3	BCK	Behchoko	62.8	-115.92	160
4	BRA	Bratts Lake	50.2	-104.71	595
5	CBY	Cambridge Bay	69.13	-105.06	35
6	CPS	Chapais	49.82	-74.98	381
7	EGB	Egbert	44.23	-79.78	251
8	ESP	Estevan Point	49.38	-126.54	7
9	EST	Esther	51.67	-110.21	707
10	ETL	East Trout Lake	54.35	-104.99	493
11	FSD	Fraserdale	49.88	-81.57	210
12	INU	Inuvik	68.32	-133.53	113
13	LEF	Park Falls, Wisconsin	45.95	-90.27	472
14	LLB	Lac La Biche	54.95	-112.47	540
15	SCT	Beech Island	33.41	-81.83	115
16	SNP	Shenandoah National Park	38.62	-78.35	1008
17	WBI	West Branch	41.72	-91.35	242
18	WGC	Walnut Grove	38.27	-121.49	0
19	WKT	Moody	31.31	-97.33	251

Table 2. Experiment design

Experiment name	Horizontal grid	Lateral boundary	Initial condition of meteorological	Time step
	spacing	condition	fields	
GLB90	0.9° (~ 90 km)	N/A	Global operational analysis	15 min
GLB45	0.45° (~ 45 km)	N/A	Global operational analysis	15 min
LAM	0.09° (~10 km)	GLB45 experiment	Regional operational analysis	5 min

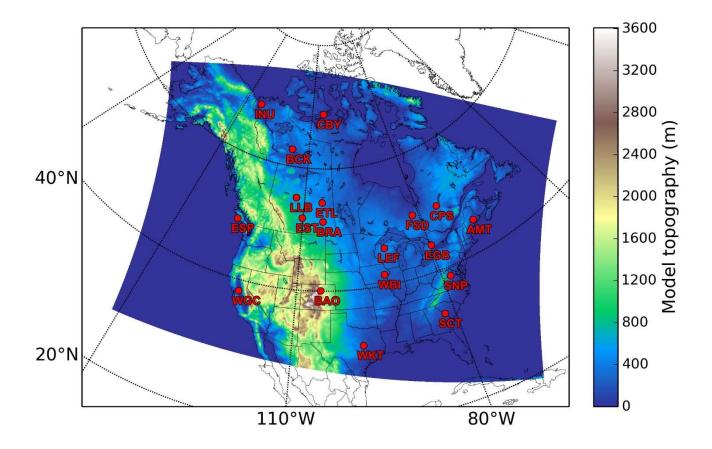


Figure 1: Model topography of the regional model with 10 km horizontal grid spacing and CO_2 measurement sites used in this study (red dot with site code).

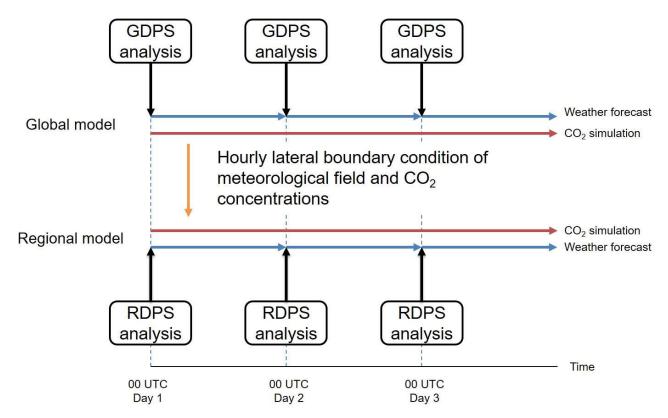


Figure 2: Schematic diagram of GEM-MACH-GHG global and regional forward model cycles. Meteorological analyses are from CMC's operational global deterministic prediction system (GDPS) and regional deterministic prediction system (RDPS). Global and regional 24 h weather forecasts start at 00:00 UTC of each day with operational analyses, while CO₂ concentrations are kept during cycles. The global forward model provides lateral boundary condition of meteorological and CO₂ fields to the regional forward model every hour.

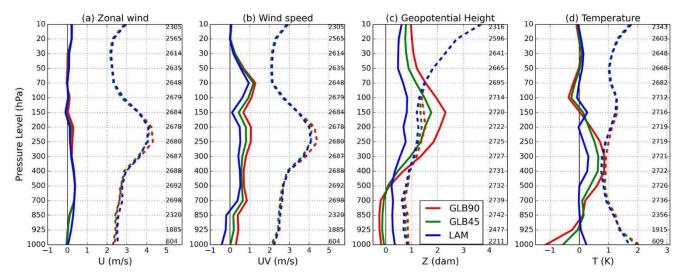


Figure 3: The bias (solid line) and standard error (dashed line) of (a) zonal wind (m s⁻¹), (b) wind speed (m s⁻¹), (c) geopotential height (dam) and (d) temperature (K) from GLB90 (red), GLB45 (green) and LAM (blue) experiments, based on comparison 24-h forecasts against North American radiosondes for July 2015. Numbers on the left side of each panel denote the pressure level. Numbers on the right side of each panel denote the number of observations used in statistics at each pressure level.

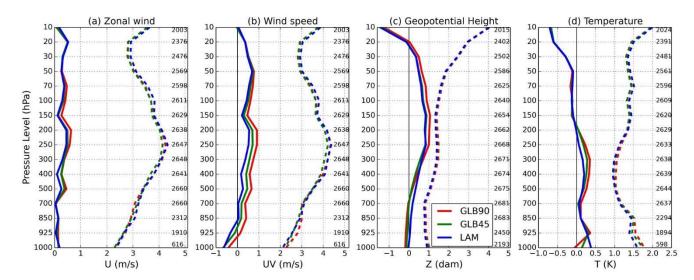


Figure 4: As in Fig. 3, but for December 2015.

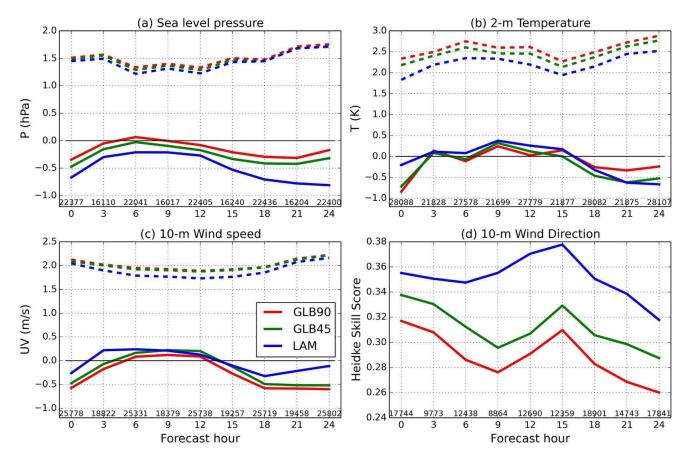


Figure 5: The Bias (solid line) and standard error (dashed line) of (a) sea level pressure (hPa), (b) 2-m temperature (K), (c) 10-m wind speed (m s⁻¹) and (d) Heidke skill score of 10-m wind direction from GLB90 (red), GLB45 (green) and LAM (blue) experiments, based on comparison forecasts against surface-based stations over North America for July 2015. Numbers on bottom of each panel denote the number of observations used in statistics at each forecast hour.

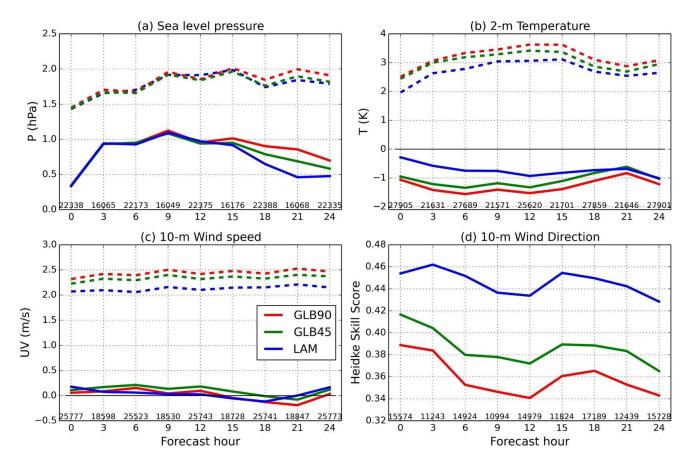


Figure 6: Same as Fig. 5, but for December 2015.

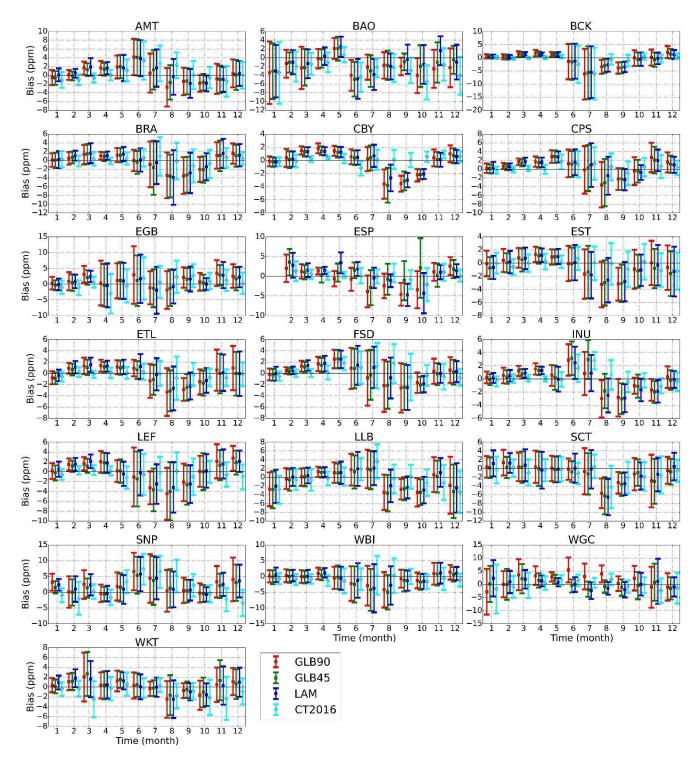


Figure 7: Monthly mean bias of daily afternoon averaged (12-16 LST) CO₂ concentrations from GLB90 (red), GLB45 (green), LAM (blue) experiments and CT2016 (cyan) at all measurement sites used in this study.

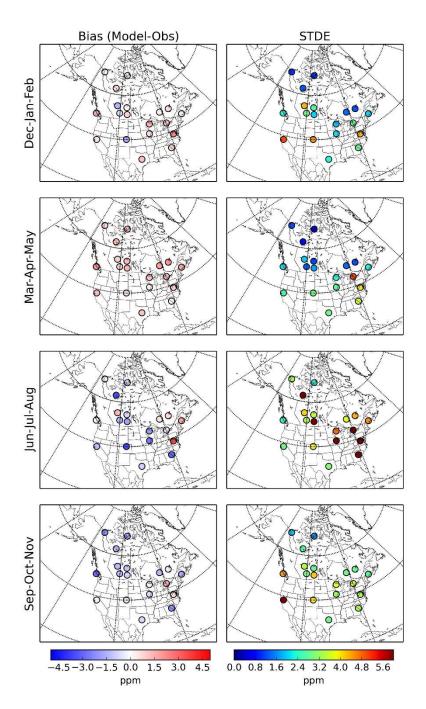


Figure 8: Mean (left column) and standard deviation (right column) of the residuals between modelled CO₂ from LAM experiment and observed CO₂ concentrations (modelled – observed) at each observations site over January to February and December 2015 (first row), March to May 2015 (second row), June to August 2015 (third row) and September to November 2015 (fourth row).

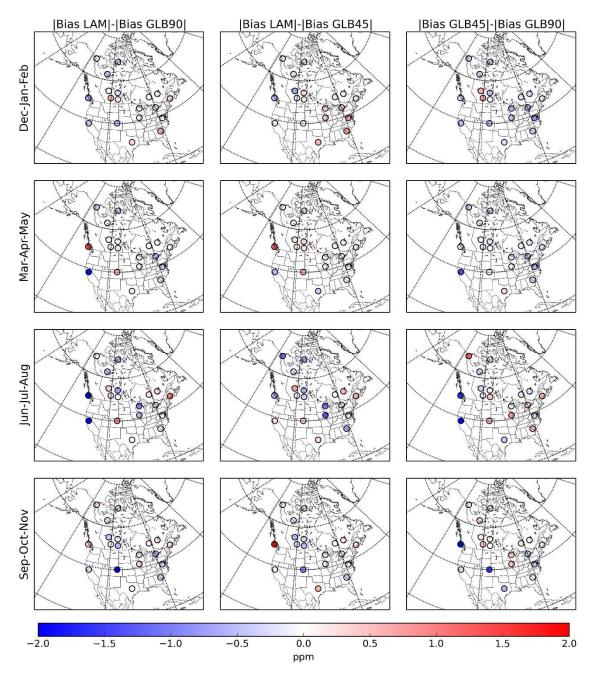


Figure 9: The difference of absolute mean bias between GLB90 and LAM (first column), GLB45 and LAM (second column) and GLB90 and GLB45 (third column) over January to February and December 2015 (first row), March to May 2015 (second row), June to August 2015 (third row) and September to November 2015 (fourth row).

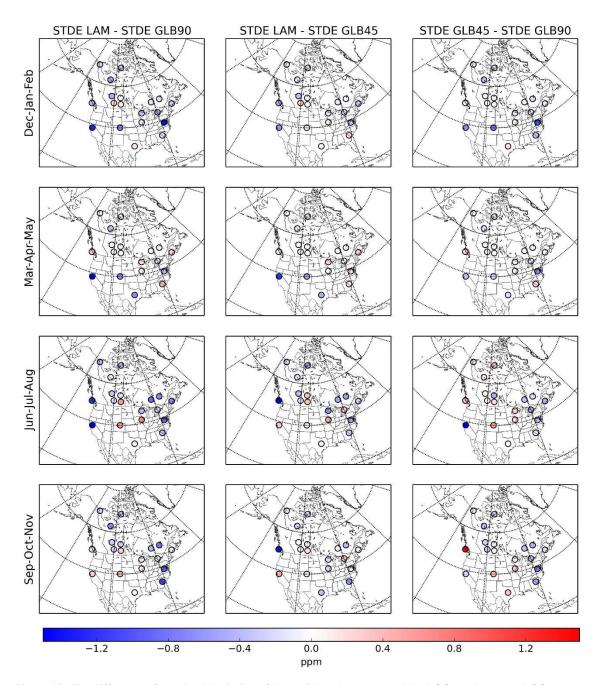


Figure 10: The difference of standard deviation of the residuals between modelled CO₂ and observed CO₂ concentrations (modelled – observed) between GLB90 and LAM (first column), GLB45 and LAM (second column) and GLB90 and GLB45 (third column) over January to February and December 2015 (first row), March to May 2015 (second row), June to August 2015 (third row) and September to November 2015 (fourth row).

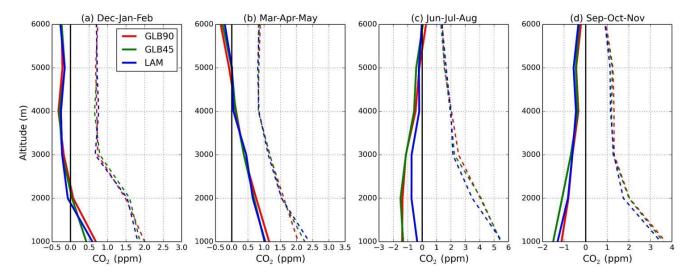


Figure 11: Comparison of profiles of modelled CO₂ concentrations from GLB90 (red), GLB45 (green) and LAM (blue) experiments to NOAA aircraft observations for (a) January to February and December 2015, (b) March to May 2015, (c) June to August 2015 and (d) September to November 2015. Solid line denotes mean bias and dashed line denotes standard error. Sites used are Briggs dale, Colorado; Cape May, New Jersey; Dahlen, North Dakota; Estevan Point, British Columbia; East Trout Lake, Saskatchewan; Homer, Illinois; Park Falls, Wisconsin; Worcester, Massachusetts; Poker Flat, Alaska; Charleston, South Carolina; Southern Great Plains, Oklahoma; Sinton, Texas; Trinidad Head, California; West Branch, Iowa.

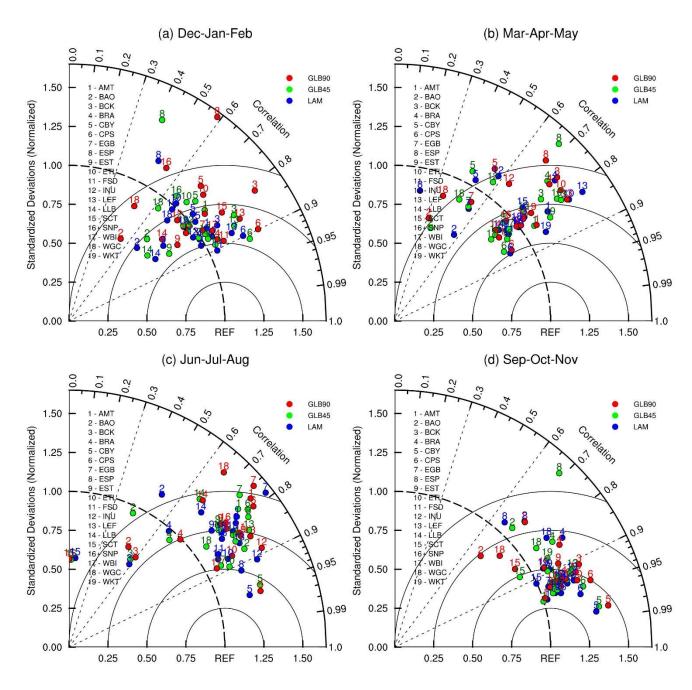


Figure 12: Taylor diagram showing correlations and normalised standard deviations between daily afternoon modelled CO₂ concentrations from GLB90 (red) and GLB45 (green) and LAM (blue) experiments and observed CO₂ concentrations over (a) January to February and December 2015, (b) March to May 2015, (c) June to August 2015 and (d) September to November 2015.

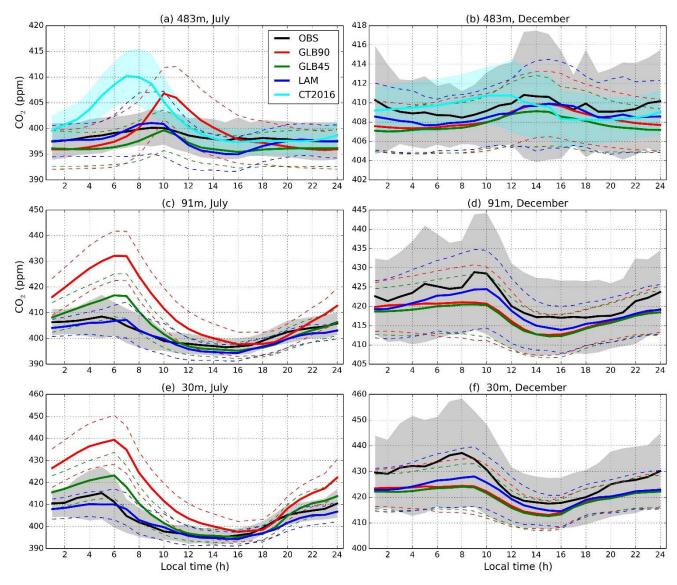


Figure 13: Mean diurnal cycle of observed CO₂ concentrations (black) and modelled CO₂ concentrations from the GLB90 (red), GLB45 (green) and LAM (blue) experiments at WGC (Walnut Grove, California) for the intake height at (a) 483 m for July 2015 and (b) 483 m for December 2015, (c) 91 m for July 2015 (d) 91 m for December 2015, (e) 30 m for July 2015 and (f) 30 m for December 2015. The grey (cyan) shaded region indicates 1 standard deviation above and below observed (CT2016) CO₂ concentrations while the dashed lines indicate the same for modelled CO₂ concentrations. Note that CT2016 results are only available at 483 m.

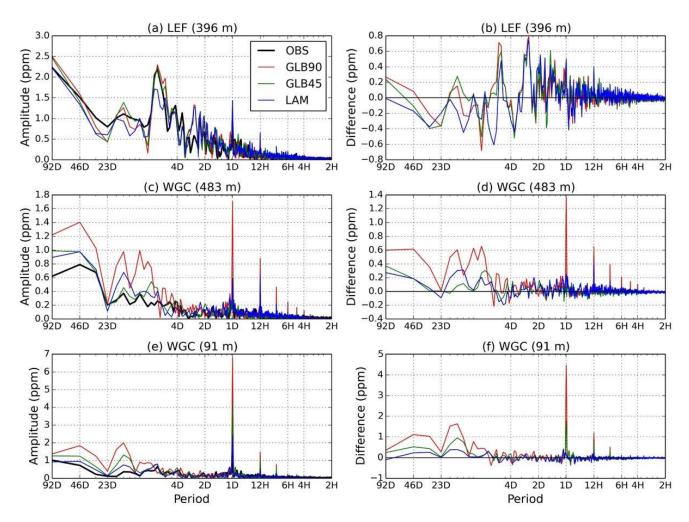


Figure 14: The amplitude of hourly time series of observed CO₂ (black) and modelled CO₂ concentrations from GLB90 (red), GLB45 (green) and LAM (blue) experiments across temporal scales from 2 h to 92 days at (a) LEF (the intake height at 396 m) and (b) their differences, (c) WGC (intake height at 483 m) and (d) their difference and (e) WGC (intake height at 91 m) and (f) their differences.