The Resolvable Scales of Regional-Scale CO₂ Transport in the Context of Imperfect Meteorology: The Predictability of CO₂ in a Limited-Area Model

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Abstract

Transport model error is an important source of uncertainty when estimating surface CO₂ fluxes via an atmospheric model inversion. In this study, the transport error due to uncertainty of meteorological fields is investigated with a high resolution, limited-area model. We characterize the extent to which errors in meteorological initial conditions (ICs) and lateral boundary conditions (LBCs) impact the quality of atmospheric CO₂ transport across spatial scales. A series of experiments is conducted using different meteorological ICs and LBCs that possess varying levels of accuracy. We find that the transport error of CO₂ is more sensitive to errors in meteorology at smaller scales O(10 km) than at larger scales O(1,000 km), and that surface CO₂ fluxes can explain the predictability of CO₂ at the largest scales. We also determine the spatial scales resolvable in the context of uncertain meteorology. These findings have implications for the development of regional-scale inverse modeling systems. When assimilating CO₂ observations near the surface, using accurate meteorological ICs is important for resolving fine-scale spatial variability of CO₂ because CO₂ transport at lower levels is more sensitive to meteorological ICs and surface CO₂ fluxes than to meteorological LBCs. However, when assimilating aircraft CO₂ measurements or XCO₂ satellite retrievals which contain information at higher altitudes, using accurate meteorological LBCs is also important. Improvement in meteorological inputs through a data assimilation system could be helpful in further resolving finer spatial scales of CO₂ at regional scales.

1. Introduction

CO₂ is one of the most significant trace gases in the atmosphere due to its role in global climate change (IPCC, 2013). Thus, estimating accurate carbon budgets is an important task in understanding the carbon cycle (Friedlingstein et al., 2019). In an atmospheric inversion of CO₂, a transport model is used as a forward model with prescribed or forecasted meteorology to transform surface CO₂ flux information into atmospheric CO₂ concentrations (e.g., Ciais et al., 2010). The recent availability of high horizontal resolution transport models at global and regional scales (e.g., Agustí-Panareda et al., 2019; Feng et al., 2016; Kim et al., 2020) now permits the variability of atmospheric CO₂ to be simulated at higher spatial and temporal resolutions than before. At the same time, the CO₂ observation network has expanded considerably so that high resolution models constrained by a dense network of surface, aircraft, and satellite-based measurements which cover broad and high temporal frequencies may be able to retrieve surface CO₂ fluxes at higher spatio-temporal scales with an inverse model. However, systematic differences in transport models continue to play an important role in errors and uncertainties of estimated surface CO₂ fluxes obtained through atmospheric inversions (Gloor et al., 1999; Houweling et al., 2010; Schuh et al., 2019).

In particular, capturing mesoscale variations correctly is important for reducing representation error in an inverse model (Corbin et al., 2008) and thus to retrieving CO₂ fluxes on higher spatial resolutions. The quality of a constituent transport model can be assessed through the quantification of transport error. Transport error refers to the departure of a model predicted CO₂ field from a true unknowable CO₂ distribution and is mathematically described in Polavarapu et al. (2016). Transport error arises due to incorrect prior surface fluxes, model formulation errors, representation errors, initial meteorological and CO₂ errors, and meteorological analysis errors. Much is already known about the CO₂ transport error that arises due to model formulation errors. For example, sources of such errors include the modeling of the planetary...
boundary layer (PBL) (Denning et al., 1995), vertical mixing in the free troposphere (Stephens et al., 2007; Yang et al., 2007), synoptic scale and frontal motions (Parazoo et al., 2008), and convective transport (Ott et al., 2011; Parazoo et al., 2008). Here, we focus on the component of transport error that arises due to uncertainty in meteorological fields.

An accurate depiction of atmospheric CO2 state evolution by a transport model depends on the quality of meteorological forcing (Deng et al., 2017) as well as the meteorological model itself. However, an explicit representation of the uncertainty in the driving meteorology is generally neglected in an inversion using offline transport model. At the global scale, the impact of meteorological uncertainty on the quality of CO2 transport was investigated by Liu et al. (2011), Kang et al. (2011, 2012), Miller et al. (2015), and McNorton et al. (2020) via experiments using ensemble of meteorological forecasts. At regional scales, similar studies using a limited-area model (LAM) have been conducted as well (Chen et al., 2019; Feng, Lauvaux, Davis, et al., 2019; Lauvaux et al., 2009). Unlike global models, a LAM must also contend with an additional source of transport error, namely, that due to uncertain the lateral boundary conditions (LBCs) (Díaz-Isaac et al., 2018). These studies have measured the uncertainty of CO2 transport due to uncertainties in atmospheric conditions with a focus on domain-averaged concentrations over specific regions. Recently, Lauvaux et al. (2019) have applied filters to small ensembles to relate spatial structures of CO2 transport error to meteorological errors. However, a systematic exploration of the limitations (or lack thereof) of meteorological uncertainty on spatial scales of transport error has yet to be performed.

The usefulness of weather forecasts is limited due to the chaotic nature of atmospheric flow (Lorenz, 1963, 1969). That is, the error in the initial condition (IC) grows with time and saturates at some point when the forecast is not differentiable from a random forecast (i.e., loss of predictability) (Lorenz, 1969). Error in the IC cascades upscale from smaller to larger scales (Lorenz, 1969; Morss et al., 2009). Then, error grows spontaneously at all scales without saturating at smaller scales (Durran & Gingrich, 2014; Mapes et al., 2008). Atmospheric predictability is an important factor in CO2 transport error since atmospheric CO2 transport is driven by weather patterns. Despite its importance on the fidelity of the CO2 simulation, less attention had been paid to the relationship between forecast sensitivity to the meteorological IC and CO2 evolution until Polavarapu et al. (2016) first investigated the predictability of CO2 using the global version of GEM-MACH-GHG, an online greenhouse gas (GHG) transport model at Environment and Climate Change Canada (ECCC). Online models enable investigating the predictability of CO2 because GHG transport therein is calculated every time step along with forecasted meteorological fields. The loss of predictability of CO2 was found to closely follow that of the wind components. Predictability was lost more quickly in the middle troposphere than in the lower troposphere or in the stratosphere. However, long after sensitivity to meteorological ICs has been lost, CO2 retains predictability months later due to surface flux information. In boreal summer, there is also a weak sensitivity to land and ocean surface fields.

The predictability limit of weather forecasts in a LAM is somewhat different from that in global models due to information of analyses or forecasts from global models provided at lateral boundaries of a LAM (Laprise et al., 2000). Larger-scale flows (e.g., O(1,000 km)) may remain predictable in a LAM for a longer period of time beyond 2–3 days. Understanding the limits imposed by the predictability of weather on the predictability of CO2 is necessary for understanding the potential for retrieving fluxes on regional scales with a LAM. Recently, a regional version of GEM-MACH-GHG has been developed, showing the capability of simulating atmospheric CO2 concentrations well throughout its model domain (Kim et al., 2020) and providing an opportunity to explore these limits.

In this paper, we identify the spatial scales of CO2 (e.g., from O(10 km) to O(1,000 km)) that can be resolved in the context of imperfect meteorological conditions such as ICs and LBCs. We also explore the limits imposed by the loss of predictability of weather on CO2 in the context of a limited area model and we use predictability limits to define upper limits to the component of transport error due to imperfect meteorology. This work is an extension of the Polavarapu et al. (2016) study in which global scales of CO2 motions were investigated. Although we focus on CO2 in this work, the experimental framework presented here can be used to explore the predictability of other "long-lived" chemical constituents (e.g., CO and CH4). The paper is organized as follows. The model and the experiments are described in Section 2 while the errors in meteorological conditions and the results are presented and discussed in Sections 3 and 4, respectively. Section 5 summarizes the conclusions.
2. Materials and Methods

2.1. Model Description

For CO$_2$ simulations, we use GEM-MACH-GHG, a coupled weather and greenhouse gas transport model at ECCC, for both global and regional-scale simulations. The global model (Polavarapu et al., 2016) is run with 0.45° (approximately 45 km) grid spacing to provide LBCs of CO$_2$ and meteorology to the regional model (Kim et al., 2020) every hour. The regional model runs with 10 km horizontal grid spacing in a domain which covers most of Canada and the United States (Figure 1). Both global and regional models have 80 vertical levels from the surface to 0.1 hPa. The global and regional models are initiated every 24 h. After each 24 h forecast, the meteorological variables are replaced by new meteorological ICs. CO$_2$ fields are not refreshed during simulations regardless of the meteorological IC. Detailed model descriptions and the forward modeling framework of GEM-MACH-GHG are available in Polavarapu et al. (2016) and Kim et al. (2020).

2.2. Experiment Design

Three sets of experiments were conducted for the year of 2015, one set for the whole year and the other two sets for 1 month (July and December), with different choices of meteorological IC and LBC, as listed in Table 1. Each experiment has transport error relative to the reference simulation due to the errors imposed in meteorological IC or LBC or both.

2.2.1. Initial Conditions for Meteorological Fields

ICs are specified on 0 UTC January 1, 2015. There are three choices for meteorological ICs for the LAM. The first is the Canadian Meteorological Centre’s (CMC’s) operational regional analysis from the regional deterministic prediction system (RDPS) which is obtained through a 4DEnVar data assimilation algorithm (Caron et al., 2015). This represents the best quality of meteorology available with our system. The second is the climatological run wherein weather forecasts are not replaced by analyses after the initial time. The 24 h forecast from the previous cycle serves as the IC for next cycle. Therefore, meteorological fields do not contain any information from atmospheric meteorological observations (through updated analyses) after the first 24 h of the run. In this scenario, CO$_2$ transport is expected to diverge rapidly from the reference.
CO₂ state due to the erroneous weather forecasts. This choice of IC represents the worst case scenario and provides an upper limit to meteorological errors. The last choice is the perturbation run and it uses the operational analyses but with added perturbations as ICs. The perturbations represent a realistic level of meteorological error. To get a realistic sample of actual meteorological analysis error, we take a realization of an analysis error from ECCC's operational regional ensemble prediction system (REPS; Baek et al., 2012) that provides flow-dependent error structures of atmospheric analyses via the operational Ensemble Kalman Filter (EnKF) data assimilation system. Since the operational deterministic analysis does not come with an error estimate, we make use of EnKF products, following the same approach used in Polavarapu et al. (2018). A perturbation is calculated by subtracting the ensemble mean from one ensemble member which is selected randomly from among 256 ensemble members. Then the calculated perturbation is added into the operational deterministic analysis.

### 2.2.2. Lateral Boundary Conditions for Meteorological Fields

The global model provides meteorological and CO₂ LBCs to the LAM every hour. In order to provide different levels of errors in meteorological conditions through the lateral boundaries of the LAM domain, three kinds of LBCs are prepared by running the global model with different ICs (Table 1). First, the GLBref experiment uses the global operational analyses from the CMC’s global deterministic prediction system (GDPS: Buehner et al., 2015) as the IC for every cycle in the simulation. As a result, the LBCs provided by the GLBref experiment would provide a realistic constraint to the LAM. Second, the GLBclim experiment provides climatological LBCs to the LAM. In GLBclim, meteorological forecasts are not replaced by updated analyses. Instead a 24 h forecast from the previous cycle is used as the IC for the next cycle. As a result, the quality of meteorological LBCs from the GLBclim experiment becomes very poor with respect to those from the GLBref experiment (a few days after the simulation start date). This choice of LBC represents the case where no realistic observations are used and provides an upper bound on the errors due to LBCs. Third, the GLBpert experiment is initiated by perturbed analyses every 24 h. The method for producing global perturbed analyses is identical with that used to produce regional perturbed analyses, except that an ensemble member from the global ensemble prediction system (Houtekamer et al., 2014) rather than the REPS is used. It is also the method that was used in Polavarapu et al. (2018).

### 2.2.3. CO₂ Surface Fluxes and CO₂ Initial and Lateral Boundary Conditions

For CO₂ surface flux, we take optimized surface CO₂ fluxes from CarbonTracker, version CT2016 (Peters et al., 2007; with updates documented at https://carbontracker.noaa.gov). Surface CO₂ fluxes on a 1° by 1° grid are available every 3 h and are interpolated onto the global and regional model domains in a
mass-conservative way. The optimized CO₂ field on 0 UTC January 1, 2015 from CT2016 is used as the CO₂ IC for both the global and regional models. For CO₂ LBCs, the GLBref experiment provides hourly CO₂ LBCs to the regional model. Unlike the meteorological fields, the CO₂ IC, CO₂ LBC, and surface CO₂ fluxes are identical for all experiments. This assumes that they do not act as error sources in the simulation, and it allows us to focus on the impact of meteorological errors on CO₂ transport error.

2.2.4. The Simulation Experiments

Seven experiments, as listed in Table 1, were conducted for 1 year (2015) using different permutations of the available ICs and LBCs. The naming convention is as follows. The first three characters determine the model: GLB or LAM (limited area model) for the global and regional models, respectively. The next three or four characters indicate the choice of ICs whether ref (reference or truth), clim (no updates of meteorological fields after the initial time), or pert (24 h updates of meteorology perturbed by realistic analysis errors). The final three or four characters indicate the LBCs used: ref (from GLBref), clim (from GLBclim), or pert (from GLBpert). The global model is used only to generate LBCs and thus only results for the regional model are discussed here. The LAMref-ref is the reference experiment using ICs from the regional operational analyses and LBCs provided by the GLBref experiment constrained by global operational analyses. The simulation error incurred by the remaining experiments is defined with respect to the LAMref-ref experiment. The LAMclim-clim, LAMref-clim, and LAMclim-ref are “climate cycle” experiments. The LAMclim-clim experiment starts from the reference IC at the start time with the reference LBCs for the first 24 h. After that, the ICs and LBCs are never updated with meteorological observations so that the meteorology will rapidly drift from the reference run. However, errors do not continue to grow forever. Instead they will remain around or below some finite value, which is determined by the size of the basin of attraction of the system. This is referred to as the error being saturated around some climatological level. This extreme case is interesting because the CO₂ field may remain predictable long after the weather predictability is lost due to knowledge of the correct surface fluxes and some influence of the surface geophysical fields (Polavarapu et al., 2016). The LAMclim-ref and LAMref-clim experiments are used to dissect this experiment and learn about the relative importance of IC and LBC errors in this case. The LAMpert-pert, LAMref-pert, and LAMpert-ref are “perturbation cycle” experiments. In these experiments, atmospheric ICs and LBCs contain flow-dependent error structures and the magnitude of meteorological uncertainties is realistic as explained in Sections 2.2.1 and 2.2.2. The LAMpert-pert experiment reveals how much improvement is possible relative to the worst-case scenario when realistic meteorological uncertainty is simulated. The LAMpert-pert and LAMpert-ref experiments are used to reveal the relative roles of IC and LBC in the LAMpert-pert experiment.

Because atmospheric CO₂ is advected by weather patterns, the growth of error in weather forecasts strongly affects the error of CO₂ transport. The practical limit of atmospheric predictability for midlatitude weather due to the growth of error in the IC is known to be about 10 days (Zhang et al., 2019). The predictability of CO₂ on weather scale seems to follow that limit as well (Polavarapu et al., 2016). However, large spatial scales of CO₂ also remain predictable on climate timescales due to the surface fluxes and to geophysical fields (Polavarapu et al., 2016). Because the time scales of interest here are seasonal time scales, we first demonstrate that the loss of predictability of CO₂ due to forecast sensitivity to meteorological ICs, saturates within a short period of time (2–3 days). After this point in time when error saturation has occurred, monthly statistics of saturated errors can be computed. For the demonstration of error saturation in two different seasons, two sets of six additional experiments were run for 31 days only. One set started on 0 UTC July 1, 2015 and the other set started on 0 UTC December 1, 2015. Within each set, all six experiments use the same CO₂ IC as the corresponding LAMref-ref experiment on 0 UTC June 30, 2015 or 0 UTC November 30, 2015, while meteorological ICs and LBCs are provided, as explained in Sections 2.2.1 and 2.2.2, according to the second and final three or four characters in each experiment name. Thus, differences of the remaining six experiments with respect to LAMref-ref appear starting on July 1 or December 1, 2015.

2.3. Diagnostics

The predictability of CO₂ is investigated for two timescales: weather timescales and seasonal timescales. Thus, two different diagnostics are used, as explained in the next sections.
2.3.1. Time Series of Simulation Error on Weather Timescales

In order to determine when CO\textsubscript{2} transport error has saturated, we measure the simulation error which is defined as the difference of the CO\textsubscript{2} field with respect to the reference CO\textsubscript{2} field (the LAMref-ref experiments from the additional experiments conducted for July and December 2015). The standard deviation (SD) of the difference field is calculated every hour for 10 pressure levels. Then, in order to get a sense of the relative magnitude of the error, the SDs of the error fields on a given pressure level are normalized by the monthly mean (July or December 2015) of the SD of the reference CO\textsubscript{2} field on the corresponding pressure level. This normalization represents the natural variability of CO\textsubscript{2} in a given month. When normalized in this way, simulation errors typically range from zero to one. When the variability in the simulation error is equal to the variability of the reference CO\textsubscript{2} state itself, the normalized simulation error (NSE) approaches 1 (Polavarapu et al., 2016). Thus, values less than 1 ensure that some degree of predictability exists. However, values can occasionally exceed 1 since there is nothing to constrain errors to the variability of the reference state. The use of a relative error diagnostic follows the approach of Laprise et al. (2000) and it also allows for comparisons of error growth at different model heights. In Polavarapu et al. (2016), a relative error was also used to compare error growth between different meteorological variables.

2.3.2. Diagnosis of Spatial Scales

Once the time elapsed to reach error saturation is determined, then the CO\textsubscript{2} predictability error can be computed for different months of the year. The difference of 24 h forecasted CO\textsubscript{2} fields from each 1-year simulation experiment was calculated with respect to the reference CO\textsubscript{2} field (from LAMref-ref). Then, power spectra of the variance of the differenced CO\textsubscript{2} fields on each model level were computed using a discrete cosine transform (DCT) method (Denis et al., 2002). This method is used to avoid the problem of aliasing caused by the aperiodic structures of planetary waves in a limited-area domain. Once the power spectra have been calculated, they are averaged in time (for a month) and over 12 model levels to obtain robust signals and filter out noise (Polavarapu et al., 2016). Here, we show results for only July and December to contrast the predictability of CO\textsubscript{2} in boreal summer versus that in boreal winter. Note that the same diagnostic (but without temporal averaging) is also applied to the LAMclim-clim and LAMpert-pert experiments from the additional experiments for July 2015 (in Section 4.1) to decompose the error growth on weather time scales into spatial scales.

3. Errors in Meteorological ICs and LBCs

Before investigating the predictability of CO\textsubscript{2}, errors in meteorological ICs and LBCs are analyzed to better understand the CO\textsubscript{2} transport error results in subsequent sections. Five variables, namely, zonal wind, meridional wind, air temperature, specific humidify, and surface pressure, are evaluated.

Three different ICs used in climate and perturbation cycle experiments are verified against the operational regional analyses used in the LAMref-ref experiment. Figures 2 and 3 show monthly averages of the bias and SD of the differenced fields for July and December 2015, separately. Biases in all experiments relative to the reference field are small compared to the SDs. In particular, the perturbed IC has the smallest error since the perturbation field is generated with respect to the ensemble mean of the analysis fields obtained from an operational atmospheric data assimilation system. Indeed, the magnitude of the SD is the largest in the LAMclim-clim experiment, followed by the LAMclim-ref then the LAMpert-ref experiment. The SD in the LAMclim-clim experiment is about 5–10 times larger than that of the LAMpert-ref experiment. For wind components and air temperature, the magnitude of SD is larger at higher altitudes, while that of specific humidity is larger near surface since the upper atmosphere is dry. The increase of SD with height for climatological wind field errors is due to the increase of wind speeds in the troposphere which maximize in the jet stream. When measurements are assimilated, analysis error SDs are reduced so that errors are nearly uniform with height (LAMpert-ref curve). Similarly, temperature errors increase at the tropopause if climatological ICs are used, but assimilating observations removes this pattern. In December, a similar pattern can be seen (Figure 3). However, the magnitude of the SD in the LAMclim-clim experiment in December is greater than that in July. The variability of errors is relatively low in the LAMpert-ref experiment (which also has the least error), while those in the LAMclim-clim and LAMclim-ref experiments vary in time (Figures S1–S3 in Supporting Information S1). This temporal variability of errors is due to the different
weather patterns that develop in the climate runs (LAMclim-clim and LAMclim-ref) relative to the reference simulation. When atmospheric data assimilation is used to obtain the IC (LAMpert-ref), such errors are greatly reduced and are more temporally consistent since the correct weather patterns are inferred from the atmospheric observations.

Figure 2. Monthly averages of the bias (solid line) and standard deviation (dashed line) of errors in the IC of (a) zonal wind, (b) meridional wind, (c) air temperature, and (d) specific humidity over the regional model domain for July 2015 for three experiments: LAMclim-clim, LAMclim-ref, and LAMpert-ref, based on a comparison against the RDPS (the IC used in the LAMref-ref experiment). Note that the errors in the IC between the LAMpert-ref and LAMpert-pert experiments are identical.

Figure 3. Same as Figure 2, but for December 2015.
To analyze errors in LBCs, hourly weather forecasts (including ICs at 0 UTC) from two global model experiments (GLBclim and GLBpert) are verified against the GLBref experiment. To consider inflow errors into the LAM and outflow errors out of the LAM, global model grid cells which overlap with the lateral boundaries of the regional model domain are selected. Figures 4 and 5 show the bias and SD of the differenced fields for July and December 2015, separately. The patterns are similar to those seen for errors in ICs. That

Figure 4. Monthly averages of the bias (solid line) and SD (dashed line) of errors in the LBCs of (a) zonal wind, (b) meridional wind, (c) air temperature, and (d) specific humidity for July 2015 for two experiments: GLBclim and GLBpert, based on a comparison against the weather forecasts in the GLBref experiment. Only the grid cells in the global model which overlap with the lateral boundaries of the regional model domain are used in the calculation.

Figure 5. Same as Figure 4, but for December 2015.
is, the GLBclim experiment has much greater meteorological error than the GLBpert experiment. Interestingly, the ratio of the magnitude of SD between the GLBclim and GLBpert experiments are similar to that between the LAMclim-clim and LAMpert experiments. The weather forecast error in the LAMclim-clim experiment is strongly influenced by meteorological errors in the LBCs provided by the GLBclim experiment. The temporal variability of SDs differs for the GLBclim and GLBpert experiments (Figures S4 and S5 in Supporting Information S1). In general, the errors in the GLBpert experiment drop suddenly at every 0 UTC when new perturbed ICs are provided. Although the perturbed ICs are not perfect, their errors are small. On the other hand, the variability of errors in GLBclim does not have discontinuities because no data assimilation is done so no error reduction at hour 0 UTC can occur. Figure S6 in Supporting Information S1 shows the temporal variation of error biases and SDs for surface pressure. Biases are much smaller than SDs and GLBclim has large temporal variability as well.

In conclusion, ICs and LBCs for climate cycle experiments have greater errors than those for perturbation cycle experiments. Therefore, it can be anticipated that CO₂ transport errors in climate cycle experiments are greater than those in perturbation cycle experiments.

4. Results and Discussions

4.1. Weather Timescales

The time series of the SD of the simulation error for July and December 2015 is shown in Figure 6 for the climate cycle experiments and in Figure 7 for the perturbation cycle experiments. The SD of simulation error grows rapidly for the first day. Since the CO₂ IC is provided by the LAMref-ref experiment, the SD of simulation error starts from zero at the initial time in all experiments. As expected, the largest SD of simulation error is near the surface where surface CO₂ fluxes generate large gradients in CO₂, in combination with the imperfect meteorology. As the surface CO₂ flux signal gets lofted upward into the free troposphere, it experiences atmospheric mixing and the signal diffuses. Also apparent is a diurnal cycle at 925 hPa. The amplitude of the diurnal cycle diminishes with altitude and is absent by 500 hPa as in Olsen and Randerson (2004).
The amplitude of the diurnal cycle is much larger in summer than in winter as biospheric fluxes are much larger in summer. Also, the magnitude at which the error with respect to the reference state saturates is larger in summer than in winter, in the troposphere. Because of the large differences in error magnitudes for different altitudes and for different seasons, the pattern of error growth cannot be contrasted among the different altitudes or seasons. Thus, the need for normalization of the error is apparent.

The NSEs from the climate cycle experiments for July and December 2015 are shown in Figure 8. The NSEs also grow rapidly for the first day as the SD of simulation error does. The growth of NSE is generally faster at 500 hPa than at 925 or 200 hPa. NSEs of CO₂ for short forecasts are strongly associated with the error growth in wind fields (Polavarapu et al., 2016). Once the NSE stops growing rapidly, the magnitude of the NSE depends on the choice of IC and LBC. In the LAMclim-clim experiment, the extreme case, predictability is mostly lost within a few days in both July and December, as expected (Figures 8a–8d). That is, the NSE approaches 1 which means that the error is as large as variations in the CO₂ in the reference state. Because the meteorological fields are not constrained by atmospheric observations, the simulations diverge completely from the reference field within a few days. Without correct meteorology, the predictability of CO₂ on weather scales is very short (around 2–3 days). This result is consistent with those obtained with our global model (Polavarapu et al., 2016). However, it slightly differs from the case of meteorology in a LAM shown in Laprise et al. (2000) where larger error in the x-component of the horizontal wind was found near the surface than at 500 hPa. For meteorological fields, the magnitudes of SDs for wind components are larger at higher altitude than near the surface in climate cycle experiments (Figures 2 and 3) and this is consistent with Laprise et al. (2000). However, relatively similar or smaller NSEs occur at 925 hPa compared to 500 hPa due to the contribution of surface CO₂ fluxes to transport of CO₂, combined with active vertical mixing within PBL. Compared to the global case, the regional model has better predictability (due to ICs and LBCs), in fact, extended predictability, as was seen for the case of regional weather and climate forecasts (Laprise et al., 2000). Indeed, with either perfect meteorological IC or LBC, NSE remains below 1 for the entire month of July and December for some altitudes (Figures 8e, 8g, 8i, and 8k). The relative importance of ICs and LBCs changes as a function of month. In July, lower NSE at 925 hPa is obtained with perfect meteorological ICs (compare Figures 8e and 8i), while in December, perfect meteorological LBCs lead to lower values at all three altitudes (compare Figures 8g and 8k). In December, higher wind speeds
and weaker magnitudes of biospheric CO₂ fluxes make it important to have good LBCs. These climate cycle experiments show that some predictability of CO₂ exists on regional scales in a LAM even when not constrained by realistic meteorological fields. Although weak, this predictability exists because all experiments used the same CO₂ surface fluxes. To check the sensitivity of results to the meteorology of the start date, the experiments in Figure 8 were repeated starting 3 days later (not shown). Only results that were not sensitive to the choice of the initial date are reported here.

In the perturbation cycle experiments, the NSE is much smaller since the meteorological fields have realistic error magnitudes and extended predictability is evident (Figure 9). Errors are particularly low at 925 hPa for an entire month with perfect ICs (Figures 9e and 9g). With realistic meteorological errors, the growth of NSE is suppressed in the model domain or at the lateral boundaries. Thus, we can expect that some portion of the variability of CO₂ could be resolved on longer timescales (beyond two weeks) due to this extended predictability. As with the climate experiments, having correct ICs is paramount in July (compare Figures 9e and 9i). However, in December, correct ICs are also more important than correct LBCs with realistic meteorological uncertainty in the troposphere (Figures 9g and 9k). This is in contrast to the climate case where correct LBCs were more important in December (Figures 8g and 8k). Thus, if the LBCs are reasonably good, the critical factor becomes the quality of the IC.

At 925 hPa, a diurnal cycle in the NSE can be seen in July but is less apparent in December (compare second to fourth columns in both Figures 8 and 9). This is related to the fact that the variability of CO₂ is governed by the covariation of biospheric fluxes and PBL height (Denning et al., 1995; Law et al., 2008). The uncertainty of CO₂ transport reaches a peak at 12 UTC (morning in North America in general). This is consistent with Chen et al. (2019) who saw a similar diurnal variation of CO₂ transport uncertainties near the surface associated with the variation of PBL height and vertical mixing. At 500 hPa, on the other hand, a diurnal cycle in the NSE reaches a peak at 0 UTC in the LAMpert-pert and LAMref-pert experiments in both July and December (Figure 9). This is associated with the different magnitude of errors in perturbed meteorological LBCs. A discontinuity appears at 0 UTC because the GLBpert experiment that provides perturbed meteorological LBC to the LAM starts from the perturbed meteorological IC every day at 0 UTC (Figure S5)
in Supporting Information S1), while CO2 fields are not refreshed. The error in the perturbed meteorological LBC grows in time from 0 UTC to the end of a cycle (Figure S5 in Supporting Information S1). As a result, the NSE at 500 hPa decreases from 0 UTC when the error in the perturbed LBC is relatively low. Then the NSE at 500 hPa increases along with the increase in the error in the perturbed meteorological LBC until new perturbed meteorological IC for the global model replaces the forecast in the GLBpert experiment at 0 UTC.

In Figures 8a and 8d, predictability is lost (the NSE approaches unity) within 2 days. In Figures 9a and 9d, predictability is retained, but a rapid error growth is seen in the first 2 days. In order to see which spatial scales are responsible for this rapid error growth, a spectral decomposition of the variance of the error fields at 12, 24, and 48 h of the additional experiment started from 0 UTC July 1, is computed for the LAM-clim-clim and LAMpert-pert experiments (Figure 10). The spectra are normalized by that of the LAMref-ref experiment at corresponding times in order to identify threshold levels of error. For example, a cross-point with value of 1 (black dashed line) indicates the wavelength at which an experiment loses predictability and a cross-point with value of 2 (black solid line) indicates the wavelength at which the error variance of the error fields reaches twice that of the LAMref-ref experiment. The error variance of a difference field can be double that of either field being differenced when the two fields in the difference are uncorrelated and have the same variances (Polavarapu et al., 2016). In Figure 10, it is evident that small spatial scales lose predictability faster than larger scales in a LAM. Even for the largest scales, rapid error growth from 12 to 24–48 h can be seen (Figures 10b and 10c) in the middle and upper troposphere and stratosphere for the climate cycle experiment. The pattern is less obvious but still evident with realistic error levels (Figures 10e and 10f). This pattern of errors increasing fastest at small spatial scales is consistent with what is seen in meteorological fields in a LAM (Laprise et al., 2000).

4.2. Seasonal Timescales

Having demonstrated that simulation errors saturate within a few days, we can compute statistics of the simulation error (if we skip the first few days) and see how they differ between boreal summer and winter in 2015. Figure 11 shows the monthly mean spectral variance of CO2 as a function of spatial scale for July and December 2015 from the 1-year experiments. This is 6 and 11 months after the predictability of weather has
been lost. The black solid line in each panel depicts the monthly mean spectral variance of the LAMref-ref experiment. Note that it is calculated from the raw CO2 state, not from any differenced fields. The spectra at all layers and months peak at large scales because CO2 is a spatially smooth field. This shape was also seen with our global model (Polavarapu et al., 2016). The spectral variance of the reference CO2 state is higher in July than December and higher at lower layers than upper layers, due to the contribution of surface CO2 fluxes to the variability of atmospheric CO2. The remaining curves correspond to spectral decompositions of error fields. Solid lines represent climate cycle experiments and dashed lines represent perturbed cycle experiments. Note that when the error at a given wavelength equals the power of the reference state, the colored curve intersects the black curve. These cross-points with black solid line indicate the spatial scales which are not resolved simply because of uncertainty in the driving meteorological fields as this is the only source of uncertainty in our experimental design. For spatial scales to the right of the cross-point, the error exceeds the power of the reference state and there is simply no correlation between the CO2 from the experiment and the reference field on those scales. That is, the experiment cannot resolve spatial scales of CO2 finer than that corresponding to the cross-point. Beyond the cross-point, the power spectra asymptote to a value that is twice that of the reference state, as seen in Figure 10.

The spatial scales which can be resolved naturally depend on the level of error imposed on the ICs and LBCs. Table 2 compares the wavelengths corresponding to the cross-point and hence the smallest resolvable scale. The LAMclim-clim experiment can resolve only the largest scales, as expected, and has the largest simulation error as seen in Figure 10, since both ICs and LBCs have no information from atmospheric observations. This is true both in July and December. Nevertheless, a few spatial scales have some predictability at layer 1. This is due to the common surface CO2 fluxes, surface flux impact from outside the model’s domain through LBCs, and surface meteorological fields that provide common information to all experiments. In contrast, at layers 2 and 3, predictability is lost even at the largest scale (which is the size of the limited area domain) since the impact of surface variables is limited at higher altitudes. When either the IC or LBC is provided with the correct information, the resolvable scales can be extended. At layer 1, in

Figure 10. Spectra of error variances for 12, 24, and 48 forecast hours after the initial time from the LAMclim-clim (top row) and LAMpert-pert experiments (bottom row) from layer 1 (first column), layer 2 (second column), and layer 3 (third column). Layers 1–3 correspond to surface~790, 790–490, and 490–190 hPa, respectively. Relative error spectra are calculated by dividing the spectral variance of differenced CO2 field of the LAMclim-clim or LAMpert-pert experiments by the spectral variance from the LAMref-ref experiment for each layer and wavelength. Differenced CO2 fields are computed by subtracting the CO2 field from the LAMref-ref experiment. CO2 fields are taken from the additional experiments started from 0 UTC July 1, 2015.
Figure 11. Monthly averages of power spectra as a function of wavelength (km). Spectra are averaged over 1 month for July 2015 (top row) and December 2015 (bottom row) and over 12 model levels from the lowest model level. Layers 1–3 correspond to surface–790, 790–490, and 490–190 hPa, respectively. Vertical dashed (dashed dot) line indicates the wavelength where range possesses 90% (95%) of the total variance of the LAMref-ref experiment (black solid contour) counting from the largest wavelength. These wavelengths corresponding to the intersection of the two vertical lines and the solid black curve in each panel are presented in Table 3.

| Experiment       | Layer 1 (surface–790 hPa) | Layer 2 (790–490 hPa) | Layer 3 (490–190 hPa) |
|------------------|---------------------------|-----------------------|----------------------|
|                  | July          | December             | July          | December             | July          | December             |
| LAMclim-clim     | 1,760         | 2,640                 | 5,280         | 5,280                 | 5,280         | 5,280                 |
| LAMref-clim      | 406           | 1,320                 | 1,760         | 2,640                 | 1,760         | 5,280                 |
| LAMclim-ref      | 1,056         | 1,056                 | 1,760         | 1,760                 | 2,640         | 1,056                 |
| LAMpert-pert     | 264           | 102                   | 377           | 352                   | 528           | 352                   |
| LAMref-pert      | No cross-point | No cross-point        | 64            | 79                    | 112           | 117                   |
| LAMpert-ref      | 264           | 28                    | 352           | 293                   | 440           | 278                   |

Note. Numbers in parenthesis indicate corresponding pressure levels.
Table 3
Wavelength That Possess 90% or 95% of Total Variances of the LAMref-Ref Experiment, Accumulated From The Largest Wavelength

| Layer 1 | Layer 2 | Layer 3 |
|---------|---------|---------|
|         |         |         |
| July    | December| July    | December| July    | December|
| 90%     | 377     | 480     | 311     | 406     | 377     | 440     |
| 95%     | 203     | 264     | 165     | 220     | 203     | 264     |

Note. Unit is km.

July, the LAMref-clim experiment loses its predictability at smaller scales than the LAMclim-clim experiment, followed by the LAMclim-ref experiment. Thus near the surface, having correct meteorological ICs is more important than having correct meteorological LBCs in July. At layers 2 and 3, the LAMref-clim and LAMclim-ref experiments lose predictability at similar spatial scales. The resolvable scales of the LAMref-clim and LAMclim-ref are degraded as the impact of surface forcing decreases. Errors from lateral boundaries degrade the quality of the weather forecast effectively at high altitudes. In addition, larger biases and SDs for wind components at higher altitude relative to near the surface are provided by meteorological LBCs for the climate cycle experiments (Figures 4a and 4b). Horizontal wind speeds at higher altitudes are faster than those at lower altitudes, and the information of surface forcing exits the lateral boundaries of the model before reaching upper levels. This result suggests that using correct meteorological ICs is more important than correct meteorological LBCs at lower levels to resolve the motion of CO$_2$ at smaller spatial scales (e.g., from O(10 km) to O(100 km)). This result is more evident in July than in December, due to the larger magnitude of surface forcing in July. In December, having the correct LBCs is relatively more important.

Greater predictability is attained in perturbation cycle experiments (dashed curves in Figure 11). Smaller spatial scales of CO$_2$ can be resolved compared to the climate cycle experiments (Table 2), as expected, due to the smaller meteorological errors in ICs and LBCs for perturbation cycle experiments relative to the climate cycle experiments (Figures 2–5). With flow-dependent error structures both in the ICs and LBCs in the LAMpert-pert experiment, the resolvable scales in layer 1 (the surface—790 hPa) exceed 264 km (July) and 102 km (December). Interestingly, the LAMref-pert experiment has predictability for all spatial scales at layer 1 both in July and December (Figures 11a and 11d, blue dashed curves). Even at higher layers, it can resolve smaller scales down to 117 km (Table 2). The transport error in layer 1 is not sensitive to imperfect LBCs of meteorology. This is a promising result for systems that use atmospheric CO$_2$ measurements near the surface (e.g., in-situ tower and flask measurements) in LAM atmospheric inversions of CO$_2$. The LAMpert-ref experiment loses predictability at the same wavelength as the LAMpert-pert experiment at layer 1 in July since the influence of imperfect LBCs is weaker at lower levels. The importance of correct LBCs on CO$_2$ transport increases in altitude, as in the climate cycle experiments (Figures 11e and 11f). But, in contrast with the climate cycle experiments, its impact is still weaker than the correct IC. Thus, improving the quality of ICs is of primary concern for reducing CO$_2$ transport error on regional scales. Adding meteorological observations with high spatial density would be beneficial, as shown in Deng et al. (2017).

In addition, at higher altitudes, perfect ICs alone are not enough to optimally resolve finer spatial scales of CO$_2$. The remaining unresolved scales in the LAMref-pert experiment are attributed to the imperfect LBCs. Therefore, improved accuracy of LBCs is also important for resolving finer spatial scales particularly at higher layers.

The smallest spatial scales of CO$_2$ that can be resolved with our system in the context of realistic meteorological analysis errors are 102 km (December) and 264 km (July) (Table 2, LAMpert-pert). Given that our model has a grid spacing of 10 km and the finest resolvable scale by a spectral decomposition is 20 km (Nyquist wavelength), these results may seem discouraging in that meteorological errors limit the capability to resolve smaller spatial scales of CO$_2$ in our modeling framework. However, not all spatial scales contribute equally to spatial variations in CO$_2$. Variations of CO$_2$ in the LAMref-ref experiment have different magnitudes across spatial scales (Figure 11, black curves). Larger spatial scales have higher variances than smaller spatial scales. To get a sense of how much of the total variance of CO$_2$ in the LAM domain can be captured by each experiment, wavelengths that possess 90% or 95% of total variances of the LAMref-ref experiment accumulated from the largest wavelength are indicated by vertical dashed lines in Figure 11 and their numbers are summarized in Table 3. When it comes to resolving the spatial variations of CO$_2$ within the LAM domain, the perturbation cycle experiments perform better than climate cycle experiments. Over 95% of total variance in the reference field is resolved by the LAMref-pert experiment with perfect IC (Tables 2 and 3). Although not all spatial scales are resolved with given uncertainties, most of them can be resolved with accurate meteorology. While in the LAMpert-pert experiment, in the presence of realistic error, the portion of resolved variance is slightly lower than the LAMref-pert experiment due to the imperfect
meteorology. At layer 1, 90%–95% (July) or over 95% (December) of total variances of CO₂ are resolved. At layer 2 and 3, under 90% (July) and 90%–95% (December) of total variances of CO₂ are resolved. Hence, more improvement in the meteorology might be necessary in layers 2 and 3 in July.

This work shows that there is a relationship between the magnitude of simulation error and the resolvable spatial scales of CO₂. Lower simulation errors indicate that finer spatial scales of CO₂ transport can be resolved. When meteorological errors are realistic in amplitude, the unresolved scales comprise only a small portion of the spatial variability of the CO₂ field. The simulation error results from the loss of predictability at smaller spatial scales.

5. Concluding Remarks

In this study, we investigate the predictability of CO₂ in the context of a LAM using the coupled weather and GHG transport model at ECCC. In contrast to our global model, where only uncertainty in meteorological IC was considered, the impact of LBCs is additionally investigated. As such, the limit of resolvable scales of modeled CO₂ concentrations due to imperfect meteorology of IC and LBC are explored through a series of experiments. Atmospheric conditions used in experiments differ but other model configurations such as surface CO₂ fluxes and the LBC of CO₂ are the same for all experiments so as to isolate CO₂ transport error due to meteorological inputs from other possible sources. We perform two sets of 1-month simulations for July and December 2015 and one set of 1-year simulations for 2015 to investigate the predictability of CO₂ in a LAM on weather and monthly timescales. For the shorter simulations, the simulation error saturates within few days due to the limit of atmospheric predictability. The magnitude of simulation error depends on the magnitude of error imposed in IC and LBC. For the longer simulations, resolvable spatial scales of CO₂ transport associated with the error of meteorological forecasts were identified. The predictability of CO₂ at smaller spatial scales (e.g., from O(10 km) to O(100 km)) is much more sensitive to errors in ICs and LBCs of meteorology, while predictability of the largest spatial scales (e.g., O (1,000 km)) is attributed mainly to surface CO₂ fluxes. The correct meteorological IC is more important than having correct meteorological LBCs in resolving CO₂ at smaller scales, which is partially due to the higher horizontal resolution of ICs (approximately 10 km) compared to LBCs (approximately 45 km). With perfect ICs and a reasonable quality of LBCs of meteorology, the predictability of CO₂ can be attained for all spatial scales at lower levels. At upper layers, the importance of the meteorological LBC is not negligible because the contribution of surface forcing is weaker and that of lateral boundaries is stronger. Examination of the predictability of CO₂ has shed light on the relative importance of errors in ICs and LBCs of meteorology on CO₂ transport error in a LAM, as well as the impact of common surface CO₂ forcing. A natural extension of this work is to use this experimental framework to study the errors in the IC for CO₂ and surface CO₂ forcing in the context of realistic meteorological IC and LBC.

With realistic error levels for meteorological fields, CO₂ simulation errors are remarkably low compared to the case where meteorological errors are large due to the absence of updated meteorological analyses. Errors remain low for the entire month in both July and December. This result is consistent with the findings of Chen et al. (2019) and Feng, Lauvaux, Keller, et al. (2019) that meteorological uncertainty is not the main source of uncertainty in CO₂ simulation error in a LAM, and that this error is most important on shorter time scales (around 1 day). These results are encouraging since they suggest that CO₂ flux estimation with a LAM can safely attribute mismatches between modeled and measured values to surface fluxes and CO₂ initial and boundary conditions.

The results shown in this study suggest some implications for inverse modeling systems which aim at resolving finer spatial scales of CO₂ at regional scales. When assimilating CO₂ observations near the surface, using correct meteorological ICs is important in resolving fine scales of CO₂ variability because CO₂ transport error at lower levels is more sensitive to meteorological IC and surface CO₂ fluxes rather than to meteorological LBC. If the grid configuration of the meteorological ICs is not identical with that of the LAM, a horizontal interpolation will be necessary to convert input data onto a model grid, acting as an additional source of error in a forward model run and providing additional errors in resolving small scales of CO₂ motion. Typically, to avoid this problem, forecasts at early hours are discarded. Using a data assimilation system to obtain a consistent and accurate meteorological IC for finer spatial scales could also be helpful (e.g.,
Deng et al., 2017. In contrast, when assimilating aircraft CO₂ measurements which contain information at altitudes above the PBL, using correct meteorological LBCs is also important. For example, estimated fluxes constrained by aircraft profiles are not sensitive to transport error associated with PBL height and vertical mixing in a global transport model (Verma et al., 2017). The assimilation of XCO₂ measurements would be more complex since they include information from the whole column (Basu et al., 2018). In reality, neither meteorological LBC provided by a global weather forecast nor CO₂ LBC provided by a global transport model is perfect. Biases of modeled CO₂ by a LAM are often attributed to biases in CO₂ LBCs (Feng, Lauvaux, Davis, et al., 2019; Kim et al., 2020). Correcting LBCs through data assimilation might be of importance in the forward and inverse model run. Indeed, it was shown that an ensemble of CO₂ LBCs obtained through a perturbation method and multiple global models also contribute the uncertainty of CO₂ in the regional model domain (Chen et al., 2019; Feng, Lauvaux, Keller, et al., 2019). Thus, investigating the impact of different CO₂ LBCs on the resolvable scales of CO₂ might be an interesting topic for future study. Recently, EC-CAS (ECCC Carbon Assimilation System) for carbon monoxide (CO) has been developed, utilizing an operational global EnKF system to simultaneously estimate the state of atmosphere and target tracer (Khade et al., 2021). In the future, it could be envisioned to utilize this system to quantify CO₂ transport error using ensemble simulations.

Data Availability Statement
CarbonTracker CT2016 results provided by NOAA ESRL, Boulder, Colorado, USA from the website at http://carbontracker.noaa.gov.

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