Characterization of Regional-Scale CO₂ Transport Uncertainties in an Ensemble with Flow-Dependent Transport Errors

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Abstract Inference of CO₂ surface fluxes using atmospheric CO₂ observations in atmospheric inversions depends critically on accurate representation of atmospheric transport. Here we characterize regional-scale CO₂ transport uncertainties due to uncertainties in meteorological fields using a mesoscale atmospheric model and an ensemble of simulations with flow-dependent transport errors. During a 1-month summer period over North America, transport uncertainties yield an ensemble spread in instantaneous CO₂ at 100 m above ground level comparable to the CO₂ uncertainties resulting from 48% relative uncertainty in 3-hourly natural CO₂ fluxes. Temporal averaging reduces transport uncertainties but increases the influence of CO₂ uncertainties from the lateral boundaries. The influence of CO₂ background uncertainties is especially large for column-averaged CO₂. These results suggest that transport errors and CO₂ background errors limit regional atmospheric inversions at two distinct timescales and that the error characteristics of transport and background errors should guide the design of regional inversion systems.

Plain Language Summary Accurate estimates of regional-scale CO₂ surface fluxes are essential to improve our understanding of the carbon cycle and to verify human CO₂ emission inventories. CO₂ surface fluxes can be inferred from atmospheric CO₂ measurements through inversion methods, which use atmospheric transport models to relate CO₂ concentration to fluxes. However, previous studies have shown that inversion results can be sensitive to errors in the simulated atmospheric transport. To better understand how to account for such transport errors, we characterize the uncertainties in simulated CO₂ concentration due to uncertainties in atmospheric transport by running an ensemble of perturbed transport simulations in a regional atmospheric model. Our results show that CO₂ uncertainties due to transport uncertainties are about half the magnitude as uncertainties due to erroneous CO₂ surface fluxes while displaying similar spatial and temporal patterns. Transport uncertainties are reduced when CO₂ is time averaged, but at the same time the influence of uncertainties in the CO₂ background concentration is increased at longer timescales. Thus, the flux signals in regional inversions are degraded by transport errors and CO₂ background errors at different timescales, and it is imperative to properly account for these errors to obtain reliable regional-scale CO₂ flux estimates.

1. Introduction

Atmospheric CO₂ inversions provide a method to infer CO₂ surface fluxes from observed atmospheric CO₂ mole fractions through the use of an atmospheric transport model (Ciais et al., 2010; Enting, 2002) and have been widely employed to gain insights into the global and regional carbon cycle (e.g., Tans et al., 1990). Moreover, inversions can potentially provide critical independent verification of anthropogenic CO₂ emission inventories to support future climate agreements (National Research Council, 2010). However, there is still a considerable divergence between results of inversions at the continental scale (Chevallier et al., 2014; Gurney et al., 2002), which inhibits our ability to draw reliable conclusions about regional CO₂ sources and

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sinks. Intercomparisons of inversion systems have revealed that the estimated CO₂ fluxes are often sensitive to the inversion setup (Gurney et al., 2002; Peylin et al., 2013). For example, the choice of atmospheric transport model can lead to contrasting conclusions about regional CO₂ fluxes (Stephens et al., 2007).

One of the largest sources of errors in atmospheric inversions is erroneous representation of atmospheric conditions by the atmospheric transport model (Gloor et al., 1999). Errors in atmospheric transport can lead to mismatches between modeled and observed CO₂ that are mistaken as flux signals. Despite the recognized importance of transport errors in atmospheric inversions, transport uncertainties are typically not fully accounted for in current inversion systems, to a large part because of a lack of complete understanding of the transport error characteristics. Transport uncertainties are typically prescribed and assumed to be uncorrelated in space and time. With denser observations, for example, from satellite measurements, it is expected that correlated transport errors will have a larger influence on inversion results (Chevallier et al., 2010).

Transport errors can be grossly divided into two components: (1) transport errors due to errors in the transport model, including errors in model parameters, and (2) transport errors due to errors in meteorological initial and boundary conditions. Previous studies have mostly focused on the first class of transport errors and quantified the uncertainties using different transport models (e.g., Engelen et al., 2002; Gurney et al., 2002) or perturbed model physics (e.g., Diaz-Isaac et al., 2018). However, even with a perfect transport model, there can exist significant transport errors due to errors in meteorological initial and boundary conditions combined with intrinsic atmospheric error growth. A few studies have quantified CO₂ transport uncertainties due to uncertainties in meteorological conditions at the global scale (Liu et al., 2011; Miller et al., 2015; Polavarapu et al., 2016) and the mesoscale (Lauvaux et al., 2009) and investigated ways to include transport uncertainties in atmospheric inversions (Kang et al., 2012; Lin & Gerbig, 2005), but no rigorous characterization of CO₂ uncertainties due to flow-dependent transport errors has been performed at the regional to continental scales.

In this paper we characterize transport uncertainties due to uncertainties in meteorological initial and boundary conditions on subseasonal timescales. We focus on a summer case during the month of July 2016 over North America, coincident with the first intensive Atmospheric Carbon and Transport (ACT)-America field campaign. ACT-America’s objectives include quantifying and reducing uncertainties in atmospheric inverse estimates of CO₂ fluxes via targeted comparisons of simulations and aircraft observations of atmospheric CO₂ mole fractions. This paper represents a preliminary exploration of the uncertainties in atmospheric CO₂ mole fractions associated with transport uncertainty. Through the use of an ensemble data assimilation system, the flow-dependent forecast error structures are represented by an ensemble of high-resolution model simulations and constrained by periodically assimilating simulated rawinsonde observations. To put the transport uncertainties in perspective, we performed two additional sensitivity experiments, one in which we perturbed only CO₂ surface fluxes and the other in which we perturbed only CO₂ lateral boundary conditions. Together, these three experiments shed light on how flux signals in atmospheric CO₂ mole fractions are degraded in regional inversions by uncertainties in atmospheric transport and CO₂ lateral boundary conditions.

2. Methods

2.1. Model and Data

We use the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem; Grell et al., 2005) version 3.6.1, which is a regional chemical transport model with online meteorology from WRF. WRF is a fully compressible, nonhydrostatic mesoscale model integrated on the staggered Arakawa C-grid and with mass-based terrain-following vertical model levels (Skamarock et al., 2008). Here WRF-Chem was run at a horizontal resolution of 27 km in a domain that covers most of North America (see Figure 1) and with 60 vertical levels extending up to 50 hPa. The vertical spacing is smallest in the boundary layer and increases gradually with height.

CO₂ is treated as an inert passive tracer; that is, the trace gas has no feedback on meteorological variables and is not part of any chemical reactions. Thus, the total mass of CO₂ in the model domain depends only on the surface fluxes of CO₂ inside the domain and the CO₂ lateral boundary conditions. We use a positive definite sixth-order diffusion scheme to more accurately simulate diffusive processes, the Mellor-Yamada CHEN ET AL.
Figure 1. (a) Map of the model domain and locations of simulated rawinsonde observations (yellow circles) that were used to constrain transport errors. The shading shows the ocean regions that were used to perturb oceanic CO2 fluxes. (b) Ecoregions and lakes used in the terrestrial CO2 flux perturbations (shading), based on the ecoregions defined by Olson et al. (1985; doi: 10.3334/CDIAC/lue.ndp017).

Nakanishi and Niino Level 2.5 PBL scheme (Nakanishi & Niino, 2006), to represent the planetary boundary layer physics, and the Kain-Fritsch convective scheme (Kain, 2004). See supporting information section S1 (Dudhia, 1988; Hong et al., 2004; Mlawer et al., 1997; Tewari et al., 2004) for more information about the WRF-Chem setup.

WRF-Chem was initialized using meteorological initial conditions from the global European Centre for Medium-Range Weather Forecasts Interim Reanalysis (ERA-Interim; Dee et al., 2011) and driven by boundary conditions from ERA-Interim available at 6-hr intervals. For CO2 mole fractions and surface fluxes, we used the CarbonTracker Near-Real Time (CT-NRT) v2017 reanalysis (https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT-NRT/), which is provided by the National Oceanic and Atmospheric Administration. CT-NRT is an extension of the CarbonTracker system (Peters et al., 2007) and uses observations of atmospheric CO2 to optimize CO2 surface fluxes. Here we used the global CT-NRT product with a spatial resolution of $3 \times 2^\circ$ longitude-latitude and 25 vertical levels for atmospheric CO2 mole fractions, a spatial resolution of $1 \times 1^\circ$ longitude-latitude for surface CO2 fluxes, and 3-hourly time resolution for both CO2 mole fractions and fluxes. To simplify the interpretation of the results, we chose to focus on land biosphere and ocean fluxes and excluded emissions from fossil fuel combustion and wildfire. The latter two sources are often assumed to be relatively well-known in inversions (e.g., CarbonTracker and CT-NRT; ; Peters et al., 2007) and therefore not considered in the flux optimization.

2.2. Experiments
As noted above we performed three experiments for the month of July 2016, independently perturbing meteorological fields in the first (labeled “Transport” below), perturbing surface CO2 fluxes in the second (“Flux”), and perturbing CO2 lateral boundary conditions in the third (“Background”). These experiments are explained next.

For the Transport sensitivity experiment, we performed an observing system simulation experiment to quantify the effect of meteorological uncertainties on atmospheric CO2 mole fractions. In the observing system simulation experiment, we assume that the transport model is perfect and that the CO2 initial conditions, lateral boundary conditions, and surface fluxes are known; thus, errors in CO2 mole fractions arise only from uncertainties in meteorological initial and boundary conditions. These errors are characterized using an ensemble of 40 WRF-Chem forecasts with flow-dependent transport errors, which was constructed following the methodology of Meng and Zhang (2007). First, we perturbed the meteorological initial conditions at
0 UTC on 1 July 2016 using climatological background error statistics (supporting information section S2) to generate 41 ensemble members. The 41st member was taken as the “truth” and run forward in free forecast mode for 1 month to create a reference run. Atmospheric errors tend to grow rapidly with time (Lorenz, 1963), so to constrain the magnitude of transport errors in the other 40 ensemble members, we extracted pseudo-rawinsonde observations from the reference run with realistic added measurement noise (supporting information Figure S1) and assimilated them into the other members every 12 hr using the ensemble Kalman filter (Evensen, 1994; Houtekamer & Zhang, 2016). This study employed the Penn State University WRF-ensemble Kalman filter system, which is originally developed in Meng and Zhang (2008a, 2008b), but whose configuration and algorithms have been continuously updated and improved for a wide range of applications (e.g., Weng & Zhang, 2012, 2016; Zhang et al., 2016, 2019, 2011, 2009); see also supporting information section S2 (Gaspari & Cohn, 1999; Zhang et al., 2004). The locations of the rawinsonde observations are based on the operational network from the Meteorological Assimilation Data Ingest System and are shown in Figure 1a. All ensemble members in this experiment were initialized with identical CO2 initial conditions and forced with the same CO2 lateral boundary conditions and surface fluxes. This ensemble-based data assimilation approach allows us to explicitly model transport error structures and estimate transport error magnitudes based on the current meteorological observation network.

To quantify the sensitivity of atmospheric CO2 mole fractions to CO2 surface fluxes, we performed another sensitivity experiment (Flux) in which only the CO2 fluxes were perturbed using an approach typically found in CO2 inversions. Specifically, we followed the methodology of the CarbonTracker system (Peters et al., 2007): for each predefined subregion, a random scaling factor is drawn from a Gaussian distribution with mean 1 and standard deviation 0.4 (40% relative uncertainties) for ocean subregions (Figure 1a) and 0.8 (80% relative uncertainties) for land subregions (Figure 1b) and applied to the 3-hourly fluxes from CT-NRT in the subregion. Flux errors are assumed to be uncorrelated between different subregions. Unlike CarbonTracker, we do not optimize the scaling factors but instead keep them constant for the whole simulation period. Forty flux realizations were generated using this method and propagated to atmospheric CO2 mole fractions through the true transport from the reference run.

Regional CO2 inversions also need to consider background CO2 mole fractions that originate from outside the regional domain. In the third and final sensitivity experiment (Background), we assessed the uncertainties in atmospheric CO2 mole fractions stemming from uncertainties in the CO2 background using four ensemble members driven by lateral boundary conditions from four different global CO2 models: CarbonTracker CT2016 (3 × 2° lon-lat, 25 vertical levels; Peters et al., 2007), with updates documented (http://carbontracker.noaa.gov); CMS-Flux GEOS-Chem (5 × 4° lon-lat, 47 vertical levels; Liu et al., 2014); TM5 4DVAR (3 × 2° lon-lat, 25 vertical levels; Basu et al., 2016); and GEOS-Chem-CarbonTracker (5 × 4° lon-lat, 47 vertical levels; Schuh et al., 2015). We used 3-hourly resolution output from all global models and the fluxes in all members were set to 0. One caveat of this experiment is that we used data from 2010 instead of 2016 because that was the last year for which all products were available to us when this study was conducted. This point is not critical for our study because we are interested in the uncertainties (i.e., the resulting ensemble spread) rather than the actual CO2 mole fractions and focus on error statistics and orders of magnitudes. As in the flux sensitivity experiment, the background sensitivity simulations were run using the true transport from the reference run. To include uncertainties in CO2 initial conditions in the Background experiment, we started the simulations in this experiment from December the previous year to allow time for the lateral boundary CO2 to completely fill the domain.

These three experiments quantify the sensitivity of atmospheric CO2 mole fractions to uncertainties in transport, CO2 surface fluxes, and CO2 background mole fractions. Unless otherwise noted, all uncertainties are reported in terms of one ensemble standard deviation in CO2 mole fractions of all simulations in each experiment. Whenever uncertainties were integrated in space or time, we averaged the squared ensemble standard deviations (i.e., the variances) and then took the square root of the average.

3. Results and Discussion

Atmospheric transport errors were constrained in the data assimilation system to about 2.2 m/s for the instantaneous $u$ and $v$ components of the wind in terms of vertically and domain-integrated
root-mean-square errors between the mean of the ensemble simulations and the reference run (supporting information Figure S2). Although the ensemble simulations and the reference run use the same boundary layer parameterization scheme, there are significant errors in boundary layer height due to uncertainties in meteorological conditions, with domain-integrated root-mean-square errors of 100 m for the nocturnal boundary layer (6–12 UTC) to 250 m in the convective boundary layer (0 UTC). The wind errors overall are slightly smaller than but nevertheless highly consistent with the errors in Meng and Zhang (2008b; supporting information Figure S3). Meng and Zhang (2008b) found that their uncertainties in the \( u \) and \( v \) components of the wind are realistic when verifying against observations, which indicates that the transport errors in our experiment are reasonable.

Next, we focus on uncertainties in simulated atmospheric CO\(_2\) mole fractions. The temporal evolutions of vertically and domain-integrated transport, flux, and background uncertainties are shown in Figure 2a. Uncertainties in transport or CO\(_2\) fluxes quickly degrade the perfect atmospheric CO\(_2\) initial conditions. The uncertainties saturate after only 12 hr and maintain a consistent mean daily magnitude over time, except for an apparent increase on 9 July and a decrease on 30 July, which are due to changes in the weekly optimized CO\(_2\) fluxes in CT-NRT. The vertically integrated transport and flux uncertainties reflect mainly the large uncertainties close to the surface (Figure 2c). There is a distinct diurnal cycle in both transport and flux uncertainties, with large peaks in the early mornings local time related to the stratification of the nocturnal boundary layer. The diurnal variation in transport uncertainties is not seen in column-averaged CO\(_2\) (XCO\(_2\); see Figures 2g and 2h), suggesting that the large transport uncertainties in the nocturnal boundary layer are mostly due to uncertainties in vertical mixing. The near-surface flux uncertainties show a secondary and smaller peak in the afternoon local time (Figure 2d) due to the strong photosynthetic uptake of CO\(_2\) in summer, which is also reflected in XCO\(_2\) (Figure 2h).

Flux uncertainties in the free troposphere above the boundary layer take about a week to saturate (Figure 2e). There is not much CO\(_2\) error growth due to erroneous CO\(_2\) surface fluxes beyond 1 week because air that has
been affected by the regional fluxes is continuously advected out of the limited-area domain. As a result of this loss of flux signals through the lateral boundaries, transport and background uncertainties have a larger influence in the free troposphere than flux uncertainties in our experiments. Moreover, our results suggest that CO$_2$ errors due to transport uncertainties or background uncertainties are significantly correlated vertically and therefore do not cancel out when averaged vertically; thus, these error sources can considerably influence XCO$_2$ estimates (Figure 2g).

Figure 3 shows the vertical distribution of time- and domain-integrated CO$_2$ uncertainties. For this and all subsequent time integrations we excluded the first week to account for the spin-up time of uncertainties.

Figure 4. Horizontal distribution of time-integrated uncertainties in CO$_2$ mole fractions (ppm) in terms of one ensemble standard deviation for transport uncertainties (left column), flux uncertainties (middle column), and background uncertainties (right column) for (a–c) CO$_2$ at 100 m above ground level (AGL), (d–f) vertically integrated CO$_2$ uncertainties in the free troposphere, and (g–i) column-averaged CO$_2$. 

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Flux and transport uncertainties are largest close to the surface, as expected, while background uncertainties vary only weakly across vertical model levels. In the afternoon, when atmospheric CO₂ mole fractions in the boundary layer are typically representative of larger areas, the time- and domain-integrated flux uncertainties near the surface are almost twice as large as the transport uncertainties (Figure 3c), which suggests that near-surface CO₂ observations are primarily influenced by flux uncertainties (roughly four times larger variance than transport uncertainties) during this time.

Figure 4 shows the horizontal distributions of time-integrated CO₂ uncertainties. Because we used scaling factors to perturb the CO₂ surface fluxes, the flux uncertainties at 100 m above ground level are large in areas with strong fluxes (Figure 4b). The same areas tend to have large near-surface transport uncertainties (Figure 4a) due to the stronger horizontal and vertical CO₂ gradients created by the fluxes. Background uncertainties are comparably small near the surface (Figure 4c) but considerable in the free troposphere (Figure 4f) and for XCO₂ (Figure 4i). Much of the background uncertainties originate from the northern lateral boundaries (supporting information Figure S4). In the free troposphere, transport and flux uncertainties are larger in the northern part of the domain where there is more extratropical cyclone activity (Figures 4d and 4e). Uncertainties in XCO₂ (Figures 4g–4i) reflect the joint uncertainties at both the surface and in the free troposphere.

Finally, we quantify how transport, flux, and background uncertainties vary from subdaily to monthly timescales. To this end, we calculated moving averages of 3-hourly instantaneous atmospheric CO₂ mole fractions over different-length time windows and then calculated the mean domain-integrated uncertainties for the different time-averaged CO₂ fields. Figure 5 shows the CO₂ uncertainties for the different error components with varying averaging window size. At 100 m above ground level, transport and flux uncertainties exhibit a similar exponential decay with increasing averaging window size, while background uncertainties decrease by only a small amount when time averaged (Figure 5a). The magnitude of near-surface flux uncertainties initially decreases fastest with increasing window length when averaged over the daily cycle. As the analysis is extended to longer averaging windows, transport uncertainties decrease until they are the smallest of the error components (Figure 5b). These results suggest that background uncertainties have the
largest spatial and temporal error correlation structures, while transport uncertainties have smaller error correlation length scales than flux uncertainties. For instantaneous CO₂ at 100 m above ground level, the transport uncertainties in our experiment yield a similar domain-integrated ensemble spread in CO₂ mole fractions as 48% uncertainties in 3-hourly surface fluxes (see gray lines in Figure 5a). When averaged over 1 week the transport uncertainties correspond to about 36% relative uncertainties in CO₂ fluxes, and over the whole month the correspondence asymptotes to 24%.

In terms of time- and domain-integrated uncertainties in XCO₂, the influence of background uncertainties exceeds the other uncertainty sources at all timescales in our month-long experiments (Figure 5c). Further, the relative importance of background uncertainties increases at longer timescales (Figure 5d). Although local XCO₂ estimates can be more sensitive to CO₂ fluxes in the close vicinity of strong sources and sinks (Figure 4h), these results indicate that background uncertainties can be a major error source for XCO₂ estimates, especially when considering longer timescales.

4. Concluding Remarks

In this study, we quantified and contrasted uncertainties in simulated atmospheric CO₂ mole fractions in a regional model from three error sources: atmospheric transport errors, errors in CO₂ surface fluxes, and errors in CO₂ background mole fractions from the lateral boundaries. Transport uncertainties were derived from an ensemble of simulations with perturbed meteorological fields. The uncertainties in meteorological variables reflect the current capabilities of estimating meteorological conditions based on the operational network of rawinsonde observations and state-of-the-art data assimilation methods. It is possible that meteorological uncertainties can be further reduced by assimilating, for example, satellite observations, but considering that we ignored other error sources in our idealized experimental setup (e.g., model error, representation error, and biased meteorological observations), we are likely underestimating transport errors, which is also indicated by our overall smaller wind errors compared with Meng and Zhang (2008b). Nevertheless, we found that transport uncertainties due to only imperfect meteorological initial and boundary conditions significantly affect forward simulations of CO₂ mole fractions in our experiment.

The fluxes in our flux sensitivity experiment were perturbed by applying ecoregion-specific scaling factors to the fluxes following the methodology used by, for example, the CarbonTracker system. Although these flux error covariances are highly simplified, they correspond to the flux uncertainties that are presently used in some inversion systems including CarbonTracker. In our study we kept the flux scaling factors constant over the whole simulation period, but despite this long-term persistence in flux errors, the fluxes leave a limited footprint on CO₂ mole fractions in our regional model because the flux signals are advected out of the limited-area domain. One caveat of our modeling experiments is that the convective parameterization scheme we used does not include tracer transport and we therefore most likely underestimate atmospheric flux uncertainties due to neglected convective transport, especially in the southern part of the domain. Nonetheless, we do not expect the inclusion of convective transport to change the conclusions of this study. Uncertainties in CO₂ lateral boundary conditions resulted in persistent differences in the CO₂ background that may arise from biases in the global systems, or from systematic errors in the interpolation of the global CO₂ mole fractions to the regional lateral boundary conditions.

The findings of this study have several implications for regional atmospheric inversions. Because erroneous CO₂ fluxes do not lead to long-term accumulation of errors in CO₂ mole fractions in a limited-area domain, regional inversions need to take into account the relatively large transport and background uncertainties when assimilating free tropospheric CO₂ observations, including aircraft measurements outside the boundary layer and column-averaged XCO₂ from satellites. At short timescales the flux signals in near-surface CO₂ mole fraction estimates are degraded by transport uncertainties, which have similar spatial and temporal patterns as CO₂ uncertainties due to erroneous fluxes. At longer timescales transport uncertainties are partially averaged out but compensated by uncertainties in CO₂ background mole fractions. In our experiments, background uncertainties exceed transport uncertainties when the averaging window size exceeds 5 days for CO₂ observations at 100 m above ground level. Finally, we found that CO₂ lateral boundary conditions from different global modeling systems produce persistent differences in CO₂ background mole fractions, and it may be advantageous for regional inversions to also continually optimize the CO₂ background as part of the inversion procedure.
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