We are extremely grateful for the helpful comments of the referees, which we have used to improve the manuscript as described below.

Reply to Anonymous Referee

1) Are the footprints calculated on a grid, and the gridded footprints used for the inversion, or are they averaged for a whole basin, and the basin footprint used for the inversions? The authors comment on sub-basin spatial pattern differences between EDGAR and VULCAN later in the paper, suggesting the latter, but it isn't stated anywhere that I can find. If it is, please point me to the right place.

Footprints were calculated on a grid (the resolution of gridded footprints are discussed in Section 2.3). When coupled with gridded emissions the resulting concentration is summed over each air basin, and emissions are scaled by air basin in the inversion. We have included more specific language in Section 2.3.

2) The temporal patterns in the prior uncertainty (i.e. are there any, or do the experiments assume a constant prior uncertainty for the entire year), as well as the spatial patterns in the prior uncertainty (is it fixed for the whole basin, or is it gridded within the basin?). Prior uncertainty is specified relative to prior emissions, hence it differs in absolute magnitude for monthly differences in emissions. Over the state this variation is ~15% when comparing May/Oct-Nov to Jan-Feb (see Fig. 1d). Prior uncertainty is specified for the whole air basin. This is now clarified in Sections 2.2 and 2.5.3.

One assumption that is critical to the applicability of this paper to other studies with real data is an assessment of the interaction between the different types of uncertainties. In particular, spatial and temporal uncertainties in the prior are not independent from each other due to the effects of transport (though the signals here are diminished by the shorter run times). This is even more important for the experiments involving transport error, as the differences in footprints will interact with the differences in the temporal and spatial patterns. A limited set of experiments should be able to address whether these effects are significant, prior to having to do the full suite for multiple more cases.

The purpose of this paper was to quantify individual contributions that could potentially bias regional fossil fuel CO2 inversions. The reviewer makes an interesting suggestion to include experiments where the different sources of uncertainty are included to see how they interact. When we tried this the different sources of uncertainty interacted in various ways that were difficult to interpret and did not enable us to make general conclusions. The interactions can improve or worsen the results in different cases. Therefore, we prefer not to include these results in the paper. However, we have added to the discussion some comments on the individual nature of our experiments and how, in reality, multiple sources of uncertainty will interact.

“We note that while we have assessed individual contributions to uncertainty in the experiments formulated here, these contributions can also interact with each other. These interactions could
act to increase the resulting biases, or partly cancel them, depending on the combination used. The possibility for interacting effects implies that multiple prior emissions estimates and transport models should be used in inversions of real data.”

Specific Comments:
What is the period of simulation?
The period of simulation is three 1 month long campaigns in May 2014, October-November 2014 and January-February 2015, analogous to the observations presented in Graven et al. 2018. We have added some clarification in lines 117-121. The transport model simulations were run 7 days back in time from time of observation (see lines 189, 197, 220-221).

211-213: The scaling of 0.5 is definitely a free parameter that deserves some sensitivity analysis as it could strongly affect your results. You could look at the covariance between the three transport models as a first guess. How does this covariance compare to the factor of 0.5*mean signal that you assume?
We use the factor of 0.5 to recreate the previous inversion setup of, for example, Fischer et al., 2017, Graven et al., 2018, to examine the posterior bias under these assumed initial conditions. We have added text to the methods and two results sections on the effect of varying the free parameter (or as I call it the uncertainty parameter) to 0.3, 0.5 and 0.8. The differences are only a few percent and they do not change our conclusions.

229-231: Correlation in the transport uncertainties assumed in your experimental setup should at least be examined before assuming your uncertainty covariance is diagonal. Again, you could use the 3-model ensemble to test this assumption.
Here we follow the commonly used practice of assuming the uncertainty covariance is diagonal (e.g. Gerbig et al. 2003; Zhao et al. 2009; Göckede et al. 2010; Jeong et al. 2012a; 2012b; 2013; Fischer et al., 2017; Graven et al., 2018). Although interesting, to test for non-diagonal covariance was deemed to be out of the scope of this study. However, we note that an experiment on the use of correlation in the model-measurement mismatch uncertainty matrix was carried out in Jeong et al., 2016. The study compared statewide posterior emissions of CH₄ in California estimated using a full (i.e. diagonal and non-diagonal) uncertainty matrix to a diagonal uncertainty matrix, and found that mean statewide emissions differed by ~6%. However this difference was not significant relative to the uncertainty (see Text 3 from the Supplemental of Jeong et al., 2016).

I have included in lines 591-596 the following:
“Here, we have assumed the model-measurement mismatch uncertainty matrix is diagonal, following previous work (e.g. Gerbig et al. 2003; Fischer et al., 2017), however the consideration of correlated errors in the uncertainty matrix has also been found to affect posterior emissions for methane in California and reduce their uncertainty at the level of several percent (Jeong et al. 2016).”

Additional references:


Section 2.5.4: what is the prior flux estimate in this experiment?
Annually averaged Vulcan, see lines 334-335.

390 - 392: isn't this easy to test by looking at footprints?
Removed these lines.

393-401: I'm not sure I see the connection here. Why does being more concentrated in urban regions change the total? (this also applies to the conclusion in lines 530-533) It also appears that there might be a temporal offset happening, where the fluxes are biased low in summer and high in fall/winter. Is there a pattern in the prior uncertainty causing this? There isn't any way to tell given the lack of temporal information in the flux results images. Another cause is the seasonality in sensitivity of the observations to the fluxes, which can again be tested by looking at footprints.
It is due to the fact that EDGAR places more emissions in built up areas. Therefore simulating concentrations at observation sites in built up areas using EDGAR emissions will, on average, have higher simulated concentrations compared to Vulcan, while total emissions are the same over the whole air basin. This leads to a scaling down of the fluxes when observation sites are primarily in built-up areas when EDGAR is used as the prior. For example, the fluxes are consistently biased low in the South Coast (14.SC) in Fig 4b.

There is no temporal offset as emissions are temporally constant in all spatial error experiments. Likewise the prior uncertainty stays constant throughout spatial error experiments.

3.2.4: A nice conclusion of this section is that the removal of outliers improves the results from transport errors alone. That could be a strong recommendation to the community for working at these scales, which is done by many modelers, but not all. - this is mentioned in the discussion, but could be more strongly highlighted here.
Added highlight - thank you.
I would suggest that a more direct analysis of the impact of transport errors by season could be accomplished by looking at basin-wide sensitivity for each observation location by season, and how that varies by transport model. This would explain a lot of the inter-model differences you are seeing in many of the other experiments as well. That would support your PBLH analysis, which gets to the heart of why the footprints would be different, but doesn’t quantify the differences between the flux sensitivities directly.

We show in Fig 2 the air basin sensitivity for each observation location by season and how that varies by transport model.

537-540: This conclusion needs to be tested by altering the estimate of transport error assumed in the inversions themselves. My guess is that the answers might be sensitive to this parameter, but that needs to be tested.

See previous answer for testing the free parameter for transport error.

Fig 2: What is “signal”? Is it just the emissions run forward through the transport
Yes - see definition in brackets on lines 225-228.

Reply to Sourish Basu

1. Line 58, “INFLUX ref” is missing.
Corrected.

2. Lines 64-65, the phrasing “simulated and observed measurements” sounds awkward to me. I understand the authors phrased it this way because the Fischer et al (2017) study is an OSSE study. I suggest rephrasing this as “Recent studies with both real atmospheric measurements of $\Delta^{14}$CO$_2$ and $\Delta^{14}$CO$_2$ simulated in observing system simulation experiments (OSSEs) at a network of sites have shown that atmospheric $\Delta^{14}$CO$_2$ can be used to estimate monthly mean Californian ffCO$_2$ emissions with posterior uncertainties of 5-8%”, or something along these lines.
Revised this sentence.

3. Lines 72-73, I would omit the qualifier “broadly consistent”, since the range (-43% to +133%) is rather large. I would suggest just stating the result of Turnbull et al (2011) as “were found to be within +X/-Y% of Vulcan”.
Revised this sentence.

4. The $\beta$ of equation (1) is not discussed in the text other than to say that it includes the influence of other terms like the biospheric disequilibrium flux. Is it assumed that $\beta$ is perfectly
known? If so, that is fine, but that should be explicitly stated. Or, one could also given an estimate of β and say why it is unlikely to be a big factor for the estimates derived in the paper. Revised later sentence to state that uncertainty in β is included in total uncertainty.

5. Likewise, $\Delta_{bg}$ is not discussed after equation (1). I notice that the authors estimate a total emission outside of California in their inversion. Is this equivalent to estimating $\Delta_{bg}$? Clarified that terms in Eq 1 are not calculated explicitly and Revised later sentence to state that background uncertainty is included in total uncertainty.

6. Lines 143-144, the four inventories mentioned cover different time periods. Are they normalized to the same California total before calculating the spread? If yes, then what determines the prior uncertainty of the California total $ffCO_2$? If no, then isn’t the spread artificially large because the inventories span different years? They are from different time periods but the differences between inventories are much larger than the differences between years (end of that paragraph). As there are differing trends in emissions between different emissions estimates for those available for individual years, it is not clear that normalizing to one year would reduce the spread.

7. Lines 146-147, was the standard deviation calculated across the four inventories, or was the spread (max to min) across four inventories assumed to be the 1- σ uncertainty? The standard deviation was calculated across the four inventories, see updated lines 161-162 and Fischer et al., 2017.

8. Lines 184 and 205, the footprints are aggregated over six days, beyond day 1. Does this mean that the flux adjustments, beyond the first 24 hours, are all coherent across six days? Is that realistic? I’m curious why this was done, since I would assume the transport model would be able to distinguish between signals coming from flux 2 days ago vs 6 days ago (say). We simply mean that the impact of including diurnal emissions more than 24 hours before observation on the resulting concentration is not significant compared to invariant fluxes - these are findings by co-author Emily White from the University of Bristol. Added a statement to clarify “This approach captures the influence of temporally varying emissions that can be significant in the first 24 hours but we assume to be negligible for the period longer than 24 hours back in time.”

9. Lines 272-273, saying “where there are regional errors in the magnitude of prior emissions” is not quite exact, I think. I suggest rephrasing this as “… in the simplest case where the only errors in prior regional flux estimates are biases in their magnitudes”. Revised.

10. Lines 287-288, it’s common practice in OSSE studies to use the more realistic scenario as the truth (nature run) and the simpler scenario as the prior. However, here the authors use
annually averaged Vulcan (less realistic) as the truth and temporally varying Vulcan (more realistic) as the prior. Why?
Revised to clarify in lines 318-323:

“It may seem counter intuitive to choose the simpler scenario (i.e. time invariant) as true emissions, however this was unfortunately due to the simulations available; we did not have simulated ffCO₂ concentrations from each air basin for temporally invariant emissions coupled with W-S-LBL footprints, only the total ffCO₂ concentrations. We do not expect that switching the prior and true emissions would significantly affect our conclusions.”

11. Lines 289-290, I would have thought that annually averaged Vulcan would have the same total as temporally varying Vulcan, since averaging conserves the total. So why was scaling necessary? Was it because the inversions only covered a few months and not an entire year? The inversion only covers periods in May 2014, October-November 2014 and January-February 2015 (see lines 122-124). May and October-November are troughs in the annual emission cycle, whilst January-February are peaks (see figure 1b).

12. Lines 298-299, similar question as before. The authors used annually averaged Vulcan (simpler scenario) as truth and prior instead of the more realistic temporally varying Vulcan. Why?
Please refer to the answer for point 10.

13. Lines 458-459. While I certainly understand the value of more observations, and am all for increasing the observations coverage of the Δ₁⁴CO₂ network, I do not think that having more observations will necessarily reduce the impact of transport model uncertainty. As the authors have themselves noted, the impact of transport model uncertainty is higher for smaller regions, while for larger regions (entire California) there is some cancellation. This is because the difference between transport models is typically more prominent at smaller scales (e.g., in the CO₂ inversion world, the global total flux is the easiest thing to estimate). So having more observations from a denser network could also sample these model differences even more and increase the impact of transport uncertainty on posterior flux estimates.
We think the word dense caused some confusion here and so we have revised this sentence to delete “dense” and to add the phrase “with multiple sites in a variety of settings” for clarification.

14. Lines around 490, and figures 3-6. The posterior bias is typically lower than the posterior uncertainty, barring a few exceptions. This could either be because the posterior biases are low (good outcome), or because the posterior uncertainties are large (less desirable outcome). Let’s say that in an ideal world, we commit to making more Δ₁⁴CO₂ measurements of higher precision, which will reduce posterior uncertainty. Will that also decrease the biases in figures 3-6? Or will it increase some of the biases (see earlier point about transport uncertainty), and may
decrease others? Basically, what I’m trying to get at here is whether the good outcome for most of the flux estimates (bias < 2σ) is a happy accident of the specific 2018 network and measurement precision, or whether there is a more fundamental reason we can expect biases to be lower than posterior uncertainties under different (possibly increased) coverage scenarios. If you reduce uncertainty in observations, this will allow the inversion to scale emissions more (presuming prior uncertainty remains constant), hence transport bias could increase whilst posterior uncertainty decreases. Hypothetically this could drive the inversion to have a posterior bias that is larger than the associated 2σ posterior uncertainty. However we did conduct experiments (see response to Anonymous Referee) whereby we varied the transport error uncertainty parameter to 0.3 and 0.8 (hence changing the balance of prior - observation/transport uncertainty), with no significant differences in the result and all results were within 2σ of the truth.

15. Line 515, the Bagley et al (2017) reference is missing from the bibliography. Fixed.

16. Line 534, suggest changing “much larger uncertainties” to “much larger percent- age uncertainties”. Corrected.
Characterizing Uncertainties in Atmospheric Inversions of Fossil Fuel CO$_2$ Emissions in California

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Key Words: Fossil Fuel, Carbon Dioxide, Simulation Experiment, Inversion, Transport Error

Abstract
Atmospheric inverse modelling has become an increasingly useful tool for evaluating emissions of greenhouse gases including methane, nitrous oxide and synthetic gases such as hydrofluorocarbons (HFCs). Atmospheric inversions for emissions of CO$_2$ from fossil fuel combustion (ffCO$_2$) are currently being developed. The aim of this paper is to investigate potential errors and uncertainties related to the spatial and temporal prior representation of emissions and modelled atmospheric transport for the inversion of ffCO$_2$ emissions in the U.S. state of California. We perform simulation experiments based on a network of ground-based observations of CO$_2$ concentration and radiocarbon in CO$_2$ (a tracer of ffCO$_2$), combining prior (bottom-up) emission models and transport models currently used in many atmospheric studies. The potential effect of errors in the spatial and temporal distribution of prior emission estimates is investigated in experiments by using perturbed versions of the emissions estimates used to
create the pseudo data. The potential effect of transport error was investigated by using three different atmospheric transport models for the prior and pseudo data simulations. We find that the magnitude of biases in posterior state-total emissions arising from errors in the spatial and temporal distribution in prior emissions in these experiments are 1-15% of posterior state-total emissions, and generally smaller than the 2-σ uncertainty in posterior emissions. Transport error in these experiments introduces biases of -10% to +6% in posterior state-total emissions. Our results indicate that uncertainties in posterior state-total fCO₂ estimates arising from the choice of prior emissions or atmospheric transport model are on the order of 15% or less for the ground-based network in California we consider. We highlight the need for temporal variations to be included in prior emissions, and for continuing efforts to evaluate and improve the representation of atmospheric transport for regional fCO₂ inversions.

1. Introduction

The U.S. state of California currently emits roughly 100 Tg C of fossil fuel CO₂ (fCO₂) each year (CARB, 2017), or approximately 1% of global emissions (Boden et al., 2016). The passing of California’s “Global Warming Solutions Act” (AB-32) in 2006 requires that overall greenhouse gas emissions in California be reduced to their 1990 levels by 2020 (a 15% reduction compared to business as usual emissions) with further reductions of 40% below 1990 levels planned for 2030, and 80% below by 2050. The California Air Resources Board (CARB) is responsible for developing and maintaining a “bottom-up” inventory of greenhouse gas emissions to verify these reduction targets. However, previous studies have shown such inventories may have errors or incomplete knowledge of sources (e.g. Marland et al, 1999; Andres et al., 2012). Uncertainties in inventories of annual fCO₂ emissions from most developed countries (i.e. UNFCCC Annex I and Annex II) have been estimated to be between 5-
10% (Andres et al., 2012), and uncertainties can become much larger at subnational levels (Hogue et al., 2016). In a recent study Fischer et al., (2017) found discrepancies between bottom-up gridded inventories of ffCO$_2$ emissions were 11% of California’s state total emissions.

Previous research has shown that inferring ffCO$_2$ emissions from atmospheric measurements, including measurements of fCO$_2$ tracers, could provide independent emissions estimates on urban to continental scales (e.g. Basu et al., 2016; Lauvaux et al., 2016; Fischer et al., 2017; Graven et al. 2018). Such estimates are derived from observations through the use of an atmospheric chemical transport model and a suitable inverse method in a process often referred to as “inverse modelling” or an “inversion”. Distinguishing enhancements of CO$_2$ due to anthropogenic or biogenic sources can be done using measurements of radiocarbon in CO$_2$ ($\Delta^{14}$CO$_2$), since CO$_2$ emitted from fossil fuel combustion is devoid of $^{14}$CO$_2$ due to radioactive decay (Levin et al., 2003).

Recent studies with both real atmospheric measurements of $\Delta^{14}$CO$_2$ and with observing system simulation experiments (OSSEs) at a network of sites have shown that atmospheric $\Delta^{14}$CO$_2$ can be used to estimate monthly mean Californian ffCO$_2$ emissions with posterior uncertainties of 5-8%, levels that are useful for the evaluation of bottom-up ffCO$_2$ emissions estimates. Furthermore, Graven et al., 2018 found their posterior emissions estimates were not significantly different from the California Air Resources Board’s reported ffCO$_2$ emissions, providing tentative validation of California’s reported ffCO$_2$ emissions in 2014-15. In another study using aircraft-based $\Delta^{14}$CO$_2$ measurements, Turnbull et al. (2011) found ffCO$_2$ emissions from Sacramento County in February 2009 had a mean difference of -17%, range: -43% to +133% with the Vulcan emissions estimate (Gurney et al., 2009).
Although atmospheric inversions may provide a method for estimating emissions that is useful for evaluating emissions reduction policies, such as AB-32, systematic errors can arise from the atmospheric transport and prior emission models (e.g., Nassar et al., 2014; Liu et al., 2014; Hungershoefer et al., 2010; Chevallier et al., 2009, Gerbig et al., 2003). Comparisons of CO$_2$ simulated by different transport models have been conducted globally (e.g. Gurney et al., 2003, Peylin et al. 2013), and on the European continental scale (Peylin et al., 2011). The latter found that transport model error resulted in differences in modelled fCO$_2$ concentrations that were 2-3 times larger than using the same transport model but different prior emissions, depending on the location and time of year. However, comparisons of fCO$_2$ simulated by different high resolution models (25 km or less) at regional scales are still lacking.

The objective of this paper is to examine the sensitivity of a regional inversion for Californian fCO$_2$ emissions to errors in the prior emissions estimate and transport model. We build on previous work by Fischer et al. (2017) that developed an Observation System Simulation Experiment to estimate the uncertainties in both California statewide fCO$_2$ emissions and biospheric fluxes that might be obtained using an atmospheric inversion. Their inversion was driven by a combination of in situ tower measurements, satellite column measurements from OCO-2, prior flux estimates, a regional atmospheric transport modelling system, and estimated uncertainties in prior CO$_2$ flux models, fCO$_2$ measurements using radiocarbon, OCO-2 measurements, and in atmospheric transport. In contrast to Fischer et al., 2017 we focus only on fCO$_2$ emissions and use a network of flask samples without incorporating satellite measurements.

Our approach is to use simulation experiments to quantify representation and transport error using the inversion setup and the observation network from Graven et al. (2018) as a test case.
Specifically we test whether the inversion can estimate the “true” emissions that were used to produce the pseudo data, within the uncertainties, when the prior emissions estimate includes spatial and temporal representation errors within the scope of current emissions estimates (Vulcan v2.2 and EDGAR v4.2 FT2010). We further test whether the inversion can estimate “true” emissions, within the uncertainties, when the transport model used for the prior simulation is different from the transport model used to produce the pseudo data, emulating transport error.

2. Data and Methods

The analysis approach applies a Bayesian inversion developed from previous work that combines atmospheric observations, atmospheric transport modelling, prior flux models, and an uncertainty specification (Jeong et al., 2013; Fischer et al., 2017). Here, the inversion scales prior emission estimates in 15 regions (Figure 1a, Table 1) termed “air basins”, classified by the California Air Resources Board for air quality control (https://www.arb.ca.gov/desig/adm/basincnty.htm).

2.1 Observation Network

As a test case to explore uncertainties in fCO₂ inversions, we use the observation network of 9 tower sites in California that was used to collect flask samples for measurements of CO₂ and radiocarbon in CO₂ in 2014-15 and simulate the same campaign periods (Figure 1a) (Graven et al. 2018). Three, month-long, campaigns were conducted: 1st – 29th May 2014; 15th October – 14th November 2014; and 26th January – 15th February 2015, with flasks sampled approximately every 2-3 days at 22:30 GMT (14:30 local standard time). We replicate the sample availability in Graven et al. (2018), including the reduction in observation sites used in Jan-Feb 2015. The time of observation was chosen as the planetary boundary layer is usually deepest in the afternoon so
that errors in the modelled boundary layer concentration are considered smaller (Jeong et al., 2013), and afternoon concentrations are more representative of large regions.

The observed fFCO₂ concentration at a given site can be calculated by (Levin et al., 2003; Turnbull et al. 2009):

\[
ffCO₂ = C_{\text{obs}} \left( \frac{\Delta_{bg} - \Delta_{obs}}{\Delta_{bg} - \Delta_{ff}} \right) + \beta \]  

(1)

Where \( C_{\text{obs}} \) is the total observed CO₂ concentration at a given site. \( \Delta \) refers to \( \Delta^{14}C \), the ratio of \(^{14}C/^{12}C\) reported in part per thousand deviation from a standard ratio, including corrections for mass-dependent isotopic fractionation and sample age (Stuiver and Polach, 1977). \( \Delta_{bg}, \Delta_{obs} \) and \( \Delta_{ff} \) are the \( \Delta^{14}CO₂ \) of background, observed and fossil fuel CO₂, respectively, where \( \Delta_{ff} \) is -1000‰ since fFCO₂ is devoid of \(^{14}CO₂\). The term \( \beta \) is a correction for the effect of other influences on \( \Delta^{14}CO₂ \), principally heterotrophic respiration (Turnbull et al. 2009). In the experiments we present here, we do not explicitly calculate \( \Delta^{14}CO₂ \) or the other terms in Equation 1, rather we simulate fFCO₂ and specify its uncertainty to be the same as the uncertainty in radiocarbon-based estimates of fFCO₂. Following Fischer et al. (2017), total observational uncertainty for fFCO₂ was assumed to be 1.5 ppm (1-σ), encapsulating measurement uncertainty, background uncertainty and uncertainty in \( \beta \). This is consistent with Graven et al. (2018), who estimated total uncertainty in fFCO₂ for individual samples of 1.0 to 1.9 ppm.

2.2 Prior Emissions Estimates and Prior Uncertainty

The two prior emissions estimates used here are gridded products produced by EDGAR (version FT2010) (EDGAR, 2011) for the year 2008 and Vulcan (version 2.2) for 2002 (Gurney et al., 2009). EDGAR is produced at an annual resolution whilst Vulcan has an hourly resolution.
two models use different emissions data and different methods to spatially allocate emissions with annually averaged statewide emissions differing by 17.8 TgC (~19% of mean emissions), and up to 11.6 TgC for individual air basins of California (Table 1). Although our campaigns took place in 2014-2015, we use emissions estimates from Vulcan for the year 2002 and EDGAR for 2008 as emissions estimates are not available from Vulcan and EDGAR for 2014-15. The difference in state total emissions between 2002, 2008 and 2014-15 is 3-6 TgC (CARB, 2017), much less than the EDGAR-Vulcan difference of 17.8 TgC.

We estimate prior uncertainty in the same way as in Fischer et al. (2017), using a comparison of four gridded emissions estimates in California (Vulcan v2.2, EDGAR FT2010, ODIAC v2013 and FFDAS v2) as well as a comparison across an ensemble of emissions estimates for one model (FFDAS v2, Asefi-Najafabady et al.,2014). Prior uncertainty is specified for the whole air basin. The relative 1-σ standard deviation across the four inventories is between 8% and 100% for individual air basins (Table 1), and this is what we use to specify the 1-σ uncertainty in the prior emissions from each air basin. This estimate of prior uncertainty is referred to as “SD prior uncertainty”. We also conduct tests with an alternative prior uncertainty of 70% for each air basin (referred to as “70% prior uncertainty”). This was done to test the sensitivity of our results to the choice of prior uncertainty. Emissions occurring outside California were assumed to have an uncertainty of 100% for both cases.

2.3 Atmospheric Transport Models

We simulate ffCO₂ using three different atmospheric transport models outlined in Table 2. These models are commonly used in regional atmospheric transport modelling and greenhouse gas inversion studies but to date have not been compared in California. Two of the transport models
use different versions and parameterizations of the Weather Research and Forecast (WRF) model combined with the Stochastic Time-Inverted Lagrangian Transport (STILT) model. The third transport model uses meteorology from the UK Met Office’s Unified Model (UM) combined with the Numerical Atmospheric dispersion Modelling Environment (NAME).

The first WRF-STILT model is run at Lawrence Berkeley National Laboratory (WS-LBL, Fischer et al. 2017; Jeong et al. 2016; Bagley et al. 2017) and uses WRF version 3.5.1 (Lin, 2003; Nehrkorn et al., 2010). Estimates for Planetary Boundary Layer Height (PBLH) are based on the Mellor–Yamada–Nakanishi–Niino version 2 (MYNN2) parameterization (Nakanishi and Niino 2004, 2006). As in Jeong et al. (2016), Fischer et al. (2017) and Bagley et al. (2017), two land surface models (LSMs) are used depending on the location of the observation site. A 5-layer thermal diffusion land surface model is used in the Central Valley for the May campaign whilst the Noah LSM (Chen et al., 2001) is used in the remaining campaigns and regions of California. We implement multiple nested domains, with the outermost domain spanning 16-59°N and 154-137°W with a 36km resolution, a second domain of 12km resolution over western North America, and a third domain of 4km resolution over California. Two urban domains of 1.3 km resolution are used in the San Francisco Bay area and the metropolitan area of Los Angeles. Footprints describing the sensitivity of an observation to surface emissions are calculated by simulating 500 model particles and tracking them backward for 7 days. The footprint of a given site and observation time is produced hourly for particles below 0.5 times the PBLH.

The second WRF-STILT model is from CarbonTracker-Lagrange (WS-CTL), an effort led at NOAA to produce standard footprints for greenhouse gas observation sites in North America (https://www.esrl.noaa.gov/gmd/ccgg/carbontracker-lagrange). WS-CTL uses WRF version 2.1.2 and the Yonsei University (YSU) (Hong et al., 2006) PBLH scheme coupled with the Noah
land surface model and the MM5 (fifth generation Pennsylvania State University-National Center for Atmospheric Research Mesoscale Model, Grell et al., 1994) similarity theory-based surface layer scheme. As with WS-LBL, simulations are run for 7 days and particles below 0.5 times the PBLH are used in the calculation of the footprint. Footprints have a spatial resolution of 0.1° for the first 24 hours and 1° for the remaining 6 days. Footprints are hourly dis-aggregated for the first 24 hours and then aggregated for the remaining 6 days. This approach captures the influence of temporally varying emissions that can be significant in the first 24 hours but we assume to be negligible for the period longer than 24 hours back in time. The 0.1° spatial resolution domain is 31° longitude by 21° latitude with the domain centered on the release location. The 1° resolution has a domain of 170°E to 50°E longitude and 10° N to 80° N latitude. The WRF domain covers most of continental North America (Fig. 1 in Nehrkorn et al., 2010) with 30 km resolution and has a finer nest with 10 km spatial resolution over the continental United States. WS-CTL simulates footprints for 500 particles released over a 2-hour period between 21:00 and 23:00 GMT (13:00 and 15:00 PST). An exception is Sutro Tower (STR), where footprints are only available for an instantaneous release of 500 particles at 22:10 GMT. Walnut Grove (WGC) footprints are available only for a release height of 30m a.g.l, which is lower than the sampling height of 91m a.g.l. used in the observation campaign (Graven et al. 2018) and used in the other two transport models. Footprints were available for 2014 but not for 2015, so the WS-CTL model is used for simulations of the May and Oct-Nov 2014 campaigns but not for the Jan-Feb 2015 campaign.

The third model, UM-NAME, is the UK Met Office’s NAME model, Version 3.6.5 (Jones et al., 2007), driven by meteorology from the Met Office’s global numerical weather prediction model, the Unified Model (UM) (Cullen et al., 1993). The UM model has a horizontal resolution of ~25
km up to July 2014, covering the period of the May 2014 campaign. The horizontal resolution was then increased to ~17 km covering both the October-November 2014 and January-February 2015 campaigns. The temporal resolution of the UM meteorology is every 3 hours for all campaigns. Following a similar approach as for the WRF-STILT models, 500 particles were released instantaneously at 22:30 GMT and simulated for hourly dis-aggregated footprints for the first 24 hours and aggregated for the remaining 6 days. The footprints are calculated for the same horizontal resolution as the UM meteorology (25 or 17km), where the particles present in the layer between 0 and 100 m above ground level are used to calculate the footprint. The computational domain covers 175.0°W to 75°W longitude and 6.0°N to 74°N latitude.

Simulated \( \text{fCO}_2 \) signals (the enhancement of CO\(_2\) concentration due to \( \text{fCO}_2 \) emissions within the model domain) are calculated by taking the product of the footprint and an emissions estimate, both with the spatial resolution of the footprint at the native footprint resolution. The resulting concentration is summed for individual air basins. Following previous work, we assume a transport model uncertainty of 0.5 times the mean monthly signal in the pseudo-observations at each site (referred to as the ‘uncertainty parameter’) (Jeong et al., 2013; Fischer et al 2017). We also test the effect of changing the uncertainty parameter to 0.3 and 0.8.

Ten ensembles were run for UM-NAME to test the effect of random errors on the calculation of the footprint. The RMSE was within 10% of the mean monthly signal for most observation sites. This is similar to the findings of Jeong et al. (2012), which the transport model uncertainty is based on. Two observation sites (THD and VTR) had slightly higher RMSE, but both were within 20% of the mean monthly signal.

### 2.4 Inversion Method
Our inversion method is a Bayesian synthesis inversion to scale emissions in separate regions of California. We follow the same approach as Fischer et al. (2017) to solve for a vector of scaling factors, \( \lambda \), for 15 air basins and a 16th region representing the area outside of California. Unlike Fischer et al. (2017), we do not split the San Joaquin Valley into two regions. The inversion uses the set of observations, \( c \), and the matrix of predicted \( \delta \text{CO}_2 \) signals from each air basin, \( K \), to optimize the cost function \( J \):

\[
J = (c - K\lambda)^T R^{-1} (c - K\lambda) + (\lambda - \lambda_{\text{prior}})^T Q_{\lambda}^{-1} (\lambda - \lambda_{\text{prior}}) 
\]

(2)

\( \lambda_{\text{prior}} \) is the prior estimate of the scaling factors (a vector of ones with length equal to the number of regions) and \( R \) and \( Q_{\lambda} \) are the error covariance matrices relating to observational and model transport errors, and prior emissions estimate errors respectively. The non-diagonal elements of \( R \) and \( Q_{\lambda} \) are zero, assuming uncorrelated errors in the prior emissions in each air basin and in the model and observations. This assumption for \( R \) is robust as we only generate one pseudo observation every 2-3 days. Included in \( R \) are observational errors and transport model errors, added in quadrature. Therefore if the average signal at an observation site is very small, then observational uncertainty (1.5 ppm) will dominate \( R \). Minimizing \( J \) using the standard least squares formulation under the assumption of Gaussian distributed uncertainties gives the posterior estimate for \( \lambda \) following:

\[
\lambda_{\text{post}} = \left(K^T R^{-1} K + Q_{\lambda}^{-1}\right)^{-1} \left(K^T R^{-1} c + Q_{\lambda}^{-1} \lambda_{\text{prior}}\right) 
\]

(3)

With the posterior error covariance given as:

\[
V_{\text{post}} = \left(K^T R^{-1} K + Q_{\lambda}^{-1}\right)^{-1} 
\]

(4)

\( \lambda_{\text{post}} \) and \( V_{\text{post}} \) are computed separately for each of the three campaigns outlined in section 2.1. Posterior emissions estimates are the product of \( \lambda_{\text{post}} \) and the prior emissions estimate from each air basin. State total emissions are then calculated by summing the emissions in each air basin.
Uncertainty in the state-wide Californian posterior flux, including error correlations, is calculated as:

$$\sigma^2 = E_{prior}V_{prior}E_{prior}'$$  \hspace{1cm} (5)

Where $E_{prior}$ is a vector of $\text{ffCO}_2$ emissions from each air basin.

2.5 Simulation Experiments

We conduct a series of experiments to test the performance of the inversion in estimating the true emissions when the emissions estimates or transport models used to produce pseudo-observations are different to those used to produce the prior simulations. The tests explore the effect differences in the magnitude, spatial distribution, and temporal variation of prior emissions have on posterior emissions. We also examine the effect of using different transport models to simulate pseudo observations and to simulate prior concentrations.

As part of these experiments, we evaluate the impact of outlier removal on the simulation experiments. Outlier removal is generally used in atmospheric inversions when there is an issue with the ability of the model to simulate a particular observation. We use the outlier removal method outlined in Graven et al. (2018) and compare with inversion results where no outliers are removed. In this outlier removal method, an observation (here, a pseudo-observation) is designated as an outlier if (1) the absolute difference between the $\text{ffCO}_2$ signals in the observation and the prior simulation is greater than the average of the observed and simulated $\text{ffCO}_2$, and (2) either the observed or simulated $\text{ffCO}_2$ is greater than 5 ppm.

2.5.1 Difference in magnitude of emissions

First we test how well the inversion estimates the true emissions if the prior emissions have a systematic error in magnitude, but no error in the spatial or temporal distribution of emissions.
and no error in atmospheric transport. In this experiment, the prior emissions estimate is given by EDGAR and the true ffCO$_2$ signals were generated by scaling the EDGAR emissions in each air basin to match the annually averaged Vulcan emissions in that air basin. These differences range from 0.1 TgC in San Diego to 11.6 TgC in the San Joaquin Valley (Table 1). The EDGAR state total emissions are 12% higher than Vulcan, so the bias in the prior estimate in the state total ffCO$_2$ emissions is +12%. The experiment is run for all the transport models with no temporal variation in emissions. This experiment assesses the performance of the inversion and the strength of the data constraint provided by the observation network in the simplest case where the only errors in prior regional flux estimates are biases in their magnitudes. Prior uncertainty is fixed per air basin for all experiments.

2.5.2 Difference in spatial distribution of emissions

To investigate the bias in the posterior emissions estimate that could result from errors in the spatial distribution of prior emissions within each air basin, we now use annually averaged Vulcan emissions as the true emissions and EDGAR emissions scaled in each air basin to match the annually averaged Vulcan emissions in that region as the prior estimate of emissions. In this experiment, the prior estimate of the total emissions in each air basin is unbiased, and we assess how differences in the spatial distribution of emissions between Vulcan and EDGAR in each air basin may lead to a bias in the posterior emissions estimate. As shown in Figure 1c, the most significant discrepancies in spatial distribution are in the major urban areas of Los Angeles and the San Francisco Bay. This experiment is also run for all the transport models using the same transport model for both the true and prior simulation and including no temporal variation in emissions.
2.5.3 Difference in temporal variation of emissions

To assess the impact of temporally-varying emissions on the inversion result, we generated true \( \text{fCO}_2 \) signals with temporally-invariant annually-averaged Vulcan emissions and prior \( \text{fCO}_2 \) signals with temporally-varying Vulcan emissions. It may seem counter intuitive to choose the simpler scenario (i.e. time invariant) as true emissions, however this was dictated by the simulations available; we did not have simulated \( \text{fCO}_2 \) concentrations from each air basin for temporally invariant emissions coupled with W-S-LBL footprints, only the total \( \text{fCO}_2 \) concentrations. We do not expect that switching the prior and true emissions would significantly affect our conclusions. We scaled the temporally-varying Vulcan emissions in each air basin so that the total \( \text{fCO}_2 \) emissions were the same magnitude as the total \( \text{fCO}_2 \) emissions in the annually averaged Vulcan emissions for each field campaign. As shown in Figure 1d, scaling was less than 10% of annual mean emissions with campaigns occurring during maxima and minima of the annual emissions cycle. Here the prior estimate is again unbiased, and we assess how differences in the diurnal variation of emissions (see Fig 1b) may lead to a bias in the posterior emissions estimate. This experiment is also run for all the transport models using the same transport model for both the true and prior simulation. Prior uncertainty is specified relative to prior emissions, hence it differs in absolute magnitude for monthly differences in emissions. Over the state this variation is ~15% when comparing May/Oct-Nov to Jan-Feb (see Fig. 1d).

2.5.4 Difference in Atmospheric Transport

To test the effect of differences in the simulated atmospheric transport of emissions, the same emissions estimate (annually-averaged Vulcan) is coupled with two different transport models to generate prior and true \( \text{fCO}_2 \) signals. This experiment investigates potential effects of transport...
errors, within the variations in transport across the three models we use. WS-LBL is considered
the “true” atmospheric transport while UM-NAME and WS-CTL are used for the prior
simulation in individual experiments. Here the prior estimate is again unbiased, and we assess
how differences in the modeled atmospheric transport may lead to a bias in the posterior
emissions estimate.

3 Results

3.1 Simulated ffCO₂ Observations

Before presenting the results of the inversion experiments, we first examine simulated ffCO₂
contributions different regions at each of the 9 observation sites. This allows us to quantify
which air basins have the largest influence on simulated concentrations at observation sites and
better interpret the results of the experiments. Figure 2 shows simulated concentrations at
observation sites resulting from emissions in the 6 highest-emitting air basins in California, and
from outside California. The highest signals (> 10 ppm) are simulated at urban sites (e.g. CIT
and SBC) for emissions from urban air basins (e.g., South Coast, 14.SC). The 9 air basins not
shown in Fig. 2 contributed signals below 0.1 ppm due to the small size or low emissions of the
air basin (e.g. Lake County and Lake Tahoe), or distance from the observation network (e.g.
Northeast Plateau, Great Basin Valleys and Salton Sea). In general, the northern sites (THD to
SLT in Fig 2) are sensitive to northern air basins (Sacramento and San Joaquin Valleys and SF
Bay), and the southern sites (VTR to SIO) are sensitive to emissions from southern air basins
(Mojave Desert, South Coast and San Diego). All transport models show the observation sites
are sensitive to more air basins in the Oct-Nov and Jan-Feb campaigns, compared to the May
campaign (Fig. 2). Signals simulated by WS-CTL come from fewer air basins than UM-NAME or WS-LBL, particularly in May.

In our simulation experiments, signals from outside California are generally small compared to the total signal for most sites (<10% on average), although they can average 20-50% for STB, STR, SLT and SIO for individual campaigns. For THD, located near the northern border of the state, a larger influence from outside California is found, 10-90%, due to a combination of relatively low local emissions and northerly winds transporting emissions from the northwestern United States and Canada.

3.2.1 Difference in magnitude of emissions

Figure 3 (a) shows the statewide inversion result for the experiment testing the effect of a bias in magnitude in regional emissions in the prior simulation. In this figure, and similar figures that follow for the other experiments, prior estimates are represented by black markers and posterior estimates are represented by colored markers, with the 2-σ uncertainty on the x-axis and the bias relative to the truth on the y-axis. The diagonal lines show 1:1 and 1:-1 lines, so that a marker lying to the right of these lines indicate the posterior bias is smaller than the posterior uncertainty, whereas a marker to the left of these lines indicate the posterior bias is larger than the posterior uncertainty. Filled markers show results using SD prior uncertainty and empty markers show results using 70% prior uncertainty. Prior and posterior uncertainties are expressed as 2-σ.

For all transport models and campaigns, the inversion is able to reduce prior bias and scale posterior emissions towards the truth. The +12% bias in the statewide emissions in the prior was reduced to a posterior bias of between 0 and +9% (mean bias = +5%) for SD prior uncertainty.
Using 70% prior uncertainty reduced prior bias to between -3 and +6 (mean = +1%). Statewide posterior uncertainty was 10-14% (mean 12%) and 14-32% (mean = 21%) for SD and 70% prior uncertainty respectively, where uncertainty is expressed as 2-σ, lower than the statewide prior uncertainties of 16% for SD and 69% for 70% prior uncertainty. There were no outliers identified in this experiment.

To determine what is driving the statewide results, we examine the individual air basin inversion results. Figure 3 (b) shows the inversion results for the six main emission regions of California, with San Joaquin Valley (8.SJV) and South Coast (14.SC) having the largest prior biases. For the San Joaquin Valley (8.SJV) and South Coast (14.SC) regions with the largest prior bias, the biases are reduced in most cases, however, only the posterior estimates from the 70% prior uncertainty experiment overlap the true emissions. The posterior estimates for SD prior uncertainty do not overlap with the truth, indicating that the 2-σ prior uncertainty of 24% in South Coast (14.SC), for example, restricts the inversion from eliminating biases of 30% in these regions (Table 1), given the observations available. The 9 air basins omitted from Fig. 3(b) are generally not being scaled by the inversion due to a lack of constraint from the observation network, low emissions, or small prior uncertainty (Figure S1).

The bias in the posterior estimate of statewide emissions is larger in May than in Oct-Nov and Jan-Feb (Fig 3a, triangles). This poorer performance of the inversion in May can be largely attributed to the San Joaquin Valley (8.SJV), where the posterior emissions are largely unchanged from the prior in May. There is no observation site in the San Joaquin Valley, and as shown in Fig. 2, emissions in the San Joaquin Valley do not reach observation sites in neighboring air basins in May, but they do reach these sites in Oct-Nov and Jan-Feb. In contrast, the South Coast (14.SC) influences the two observation sites, CIT and SBC, located in the region...
as well as several other sites (Fig. 2). Both CIT and SBC show prior signals are too high compared to true signals for all campaigns and models (Fig. 3c), reflecting the positive bias in prior emissions in the South Coast region, which is reduced in the posterior. Changing the uncertainty parameter from 0.5 to 0.3 or 0.8 had the result of decreasing the ability of the inversion to scale state-wide emissions towards true emissions by 1-4%, with an increase in posterior uncertainty by a similar percentage.

### 3.2.2 Difference in spatial distribution of emissions

The statewide inversion results for the experiment including errors in the spatial distribution of emissions are shown in Figure 4 (a). In this case the magnitude of prior emissions in each air basin is equal to true emissions and we aim to quantify how errors in the spatial distribution of emissions (EDGAR as prior and Vulcan as true distribution) lead to bias in posterior emissions estimates. Posterior emissions are negatively biased, apart from WS-LBL in January-February. Posterior bias was between -10% and +1% (mean = -4%) for SD prior uncertainty and between -10% and +4% (mean = -4%) for 70% prior uncertainty across transport models and campaigns. As might be expected from the experimental setup with an unbiased prior, posterior emissions estimates generated using SD prior uncertainty have a smaller mean bias and smaller range of posterior estimates compared to those generated using 70% prior uncertainty. Statewide uncertainty was reduced from 16% to 10-14% (mean = 12%) for SD prior uncertainty and from 58% to 14-21% (mean = 18%) for 70% air basin prior uncertainty. Biases induced are smaller than the 2-σ posterior uncertainty across all transport models, campaigns and choice of prior uncertainty.
Posteriors results in the two largest emitting air basins (the San Francisco Bay and South Coast) are also negatively biased in most cases (Fig 4b). In several cases, posterior biases are larger than the associated posterior uncertainties, for example in the South Coast for WS-LBL in all cases. Considering Figure 4(c), prior fCO₂ signals are being overestimated more often than underestimated, particularly for the relatively more urban sites CIT and SLT.

Since the prior emissions from EDGAR have been scaled to have the same total as Vulcan (the true emissions) in each region, the pattern of more negative posterior emissions is only caused by the sub-regional spatial distribution of emissions. Comparing Vulcan and EDGAR native grid cell emissions in Figures 1c and S2, EDGAR tends to have greater emissions in high-emission grid cells. In other words, the emissions are more concentrated in EDGAR and more dispersed in Vulcan. This pattern explains the negative bias in posterior emissions for the urban South Coast air basin. The opposite effect does not appear to hold for rural observation sites and regions, perhaps because rural emissions are already rather dispersed and have less of an influence on the observations.

In these experiments, 0-3% of observations were identified as outliers, but excluding outliers did not change the statewide result significantly (<1% change in mean bias).

### 3.2.3 Difference in temporal variation of emissions

Figure 5(a) shows the statewide inversion result for the experiment where the emissions are Vulcan temporally-varying in the prior simulation (see Fig. 1b) but Vulcan temporally-invariant in the true simulation. Posterior bias was between -13 and +5% (mean = -3%) for SD uncertainty and between -15% and +6% (mean = -3%) for 70% prior uncertainty. Posterior uncertainty was 11-15% (mean = 12%) for SD prior uncertainty and 15-24% (mean = 18%) in posterior

**Deleted:** Sacramento Valley (3.SV) and the San Joaquin Valley (8.SJV) have higher posterior emissions in WS-LBL in most cases, possibly due to the inversion compensating for reduced posterior emissions in the San Francisco Bay (13.SFB) and South Coast (14.SC).
emissions for SD (70%) prior uncertainty. Outlier removal resulted in 0-1% (mean = 0%) of data points being removed, which did not change the statewide results.

The posterior estimate for WS-LBL in May with SD prior uncertainty has a significant negative bias of -13%, approximately the same magnitude as the associated 2-σ posterior uncertainty. As can be seen by the air basin results of Figure 5 (b), the statewide bias for WS-LBL in May is being driven by a large regional bias in the South Coast, but also in the San Francisco Bay and San Diego air basins. These regional biases are larger than their associated posterior uncertainties. Figure 5 (c) shows the prior ffCO₂ signals at CIT average ~7ppm too high in May for WS-LBL. In contrast, prior ffCO₂ signals at CIT and SBC are too low in Oct-Nov for WS-CTL, leading to a high bias in posterior emissions from the South Coast. San Diego also exhibited both high and low biases in the posterior emissions. Overall, temporal variations in emissions led to posterior biases generally within ±6%, but as large as 15%; however, a consistent pattern in the posterior bias due to the temporal representation in emissions was not found.

3.2.4 Difference in Atmospheric Transport

The statewide inversion results for the experiment where the atmospheric transport in the prior simulation uses WS-CTL or UM-NAME but the atmospheric transport in the true simulation uses WS-LBL are shown in Figure 6 (a). Outliers were identified in these experiments and we present results for inversions including all data and for inversions where outliers were removed. When all data are included, differences in atmospheric transport model introduce a bias in statewide posterior emissions of between -42% and -3% (mean = -12%) for SD prior uncertainty and between -32% and 0% (mean = -15%) for 70% prior uncertainty. For one case, using WS-
CTL to generate prior signals in October-November, the bias in the posterior emissions estimate was larger than the 2-σ uncertainty for both SD and 70% prior uncertainty. Changing the uncertainty parameter from 0.5 to 0.3 or 0.8 resulted in posterior emissions remaining closer to true emissions by 0-4% and increasing the posterior uncertainty by 1-5%.

Removing outliers significantly improved the inversion results (Figure 6 b): the mean bias was between -10% and 0% (mean = -3%) for SD prior uncertainty and between -9% and +6% (mean = -5%) for 70% prior uncertainty when outliers were removed. Posterior uncertainty was 9-15% (mean = 12%) and 15-24% (mean = 18%) for SD and 70% prior uncertainty respectively, with all posterior estimates within 2-σ of the true statewide emissions. The reduction in posterior bias when outliers are removed is mostly due to the removal of a few large positive outliers in prior simulated signals by WS-CTL (Figure 7). Figure 7 illustrates the time series of simulated fCO₂ in each model with outliers shown as an x. Outliers removed were between 6.9% and 20.6% of all observations (mean = 10.5%). This is similar to the fraction of outliers identified in Graven et al. 2018 using the same method with real data (~8%). It is also similar to that of Jeong et al., 2012a and b (0-27%) for monthly inversions of CH₄ in California using a different method of identifying outliers where model-data residuals are larger than 3-σ of model-data uncertainty.

This is an important result for the atmospheric inversion community working at such spatial scales, as it highlights the benefits of removing outliers.

While the statewide posterior emissions estimate is significantly biased in only one case (WS-CTL in Oct-Nov) when outliers are not removed, the posterior emissions estimates for the main emissions regions are significantly biased in several cases (Fig 6c). The largest bias is in the South Coast region where posterior estimates are biased by more than -75% (with 1% posterior uncertainty) in Oct-Nov when using WS-CTL to generate prior signals. This large posterior
emissions bias in the South Coast and the statewide total can be attributed to overestimates in prior $\text{fCO}_2$ signal of more than 6ppm on average at CIT and SBC and more than 2ppm at WGC and STR (Fig. 6e) due to some high outliers in the WS-CTL simulations (Fig. 7). Posterior estimates for San Francisco Bay, South Coast and San Diego were also significantly biased in some other cases, particularly for 70% prior uncertainty but also for SD prior uncertainty. This indicates that regional biases caused by differences in atmospheric transport appear to compensate over the statewide scale, and that results for individual regions are less robust than aggregate results for the statewide network. It also suggests that an observation network with multiple sites in a variety of settings is beneficial to reducing the impact of uncertainty in atmospheric transport.

To investigate the differences in simulated $\text{fCO}_2$ and assess whether these could be attributed to specific aspects of modelled meteorology, we compared PBLH and wind speed in WS-LBL and the UM for 5 of the 9 observation sites where PBLH output was available. PBLH was not available for WS-CTL. Estimates for PBLH in WS-LBL are based on the Mellor–Yamada–Nakanishi–Niino version 2 (MYNN2) parameterization scheme that estimates PBLH using localized turbulence kinetic energy closure parameterization (Nakanishi and Niino 2004, 2006). Estimates of PBLH are calculated internally within the UM. PBLH and wind speed were averaged over 6 hours from 12 to 6pm Pacific Standard Time to compare the afternoon means (Seibert et al., 2000). We found no consistent correlation between differences in PBLH or wind speed and differences in simulated $\text{fCO}_2$ between models across sites and campaigns (Figure S3). Absolute values of wind direction and $\text{fCO}_2$ did not show consistent correlations either. The lack of correlation suggests we cannot attribute differences in simulated $\text{fCO}_2$ to any single meteorological variable estimated at any individual station in the transport models.
We also examined if differences in simulated \( \text{fCO}_2 \) signals across transport models could be explained by the differences in spatial resolution of the models. WS-CTL footprints were re-gridded from a 0.1° native grid to the coarser UM-NAME grid of 17 or 25km and then used to simulate \( \text{fCO}_2 \). For this comparison, we simulated \( \text{fCO}_2 \) every day over the campaign period.

We found no consistent reduction in mean \( \text{fCO}_2 \) bias between sites over the 2 campaigns, however there is a reduction in spread of bias at 4 sites for both campaigns (WGC, SLT, SBC and SIO), suggesting model resolution could potentially have an impact for these sites. In general however, we cannot say that transport model resolution error in atmospheric transport is a key driver of \( \text{fCO}_2 \) signal bias across observation sites (Figure S4).

4 Discussion

Our results show that atmospheric inversions can reduce a hypothetical bias in the magnitude of prior \( \text{fCO}_2 \) emissions estimates for the U.S. state of California using the ground-based observation network from Graven et al. (2018), under the idealized assumptions of perfect atmospheric transport and perfect spatio-temporal distribution of emissions in the prior estimate. By exploring differences in model transport and spatio-temporal distribution of prior emissions, we found that biases of magnitude 1-15% in monthly posterior estimates of statewide emissions can result from differences in the temporal variation, spatial distribution and modelled transport of the prior simulation. However, these biases were less than the 2-\( \sigma \) posterior uncertainty in state-total emissions, when outliers were removed. In some cases, the biases in posterior emissions for individual air basins were significant, compared to the posterior uncertainties, suggesting that estimates for individual regions are less reliable than the aggregate estimates of the state-wide total.
The largest bias in statewide posterior estimates was found to be caused by errors in the temporal variation in emissions. This highlights the necessity for temporally-varying emissions to be estimated and included in prior emissions estimates, particularly for urban regions. Similar results have been found in other regions including Indianapolis (Turnbull et al. 2015) and Europe (Peylin et al. 2011), and more generally, for high-emission regions around the globe (Zhang et al. 2016). Although the afternoon sampling is near to the diurnal maximum in emissions in California (Fig. 1c, Gurney et al. 2009), which might be expected to lead to higher simulated \( \text{ffCO}_2 \) in temporally-varying vs temporally-invariant emissions, we did not find consistently positive biases in \( \text{ffCO}_2 \) but rather both positive and negative biases. This suggests the overall impact of temporally-varying emissions depends on the model transport and the characteristics of the observation site. Furthermore, uncertainties in the temporal distribution of emissions at an hourly resolution have not yet been fully quantified (Nassar et al., 2013).

Errors in model transport, as represented in our experiments by using different transport models, were shown to bias posterior \( \text{ffCO}_2 \) emissions by 10% or less, when outliers were removed. These biases related to transport error are somewhat lower than estimated by similar simulation experiments for \( \text{ffCO}_2 \) emissions estimates for the U.S. by Basu et al. (2016) using different transport models (>10%), although their spatial scale was larger and the alternate model they used was intentionally biased. In contrast, the three models we use are all actively applied in regional greenhouse gas inversions. Our results are comparable to the estimate of ±15% uncertainty in atmospheric transport in WS-LBL using comparisons with atmospheric observations of CO in California (Bagley et al. 2017).

The fraction of pseudo-observations we identified as outliers in these transport error experiments (10.5%, range 6.9-20.6%), was similar to Graven et al., 2018, where 8% of all observations were
removed as outliers using the same method. The outliers in our experiments were primarily high
\( \text{ffCO}_2 \) signals simulated by WS-CTL in Oct-Nov. When included in the inversion, these did lead
to significant biases in the posterior estimates for the experiment on model transport. This
highlights the need for careful examination of simulated \( \text{ffCO}_2 \) and consideration of outliers in
atmospheric \( \text{ffCO}_2 \) inversions.

Attributing differences in simulated \( \text{ffCO}_2 \) between the different transport models to specific
meteorological variables proved inconclusive, and model resolution error did not appear to
explain the differences in simulated signals, although we were not able to investigate aggregation
error in comparison to the high-resolution WS-LBL model. Wang et al. (2017) found
aggregation error to be only a minor contributor to errors in simulated \( \text{ffCO}_2 \) in Europe, while
Feng et al., (2016) found that high-resolution gridded emissions estimates could be more
important than high resolution transport models for simulations of greenhouse gases in Greater
Los Angeles. We found that differences in the spatial representation of prior emissions in
EDGAR compared to Vulcan led to consistently lower, although not significantly different,
posterior state-wide estimates due to the emissions in EDGAR being more concentrated in urban
regions. The spatial allocation of emissions between urban and rural regions in gridded emissions
estimates have much larger percentage uncertainties than national totals (Hogue et al. 2016),
suggesting that several different gridded emissions estimates should be used in regional \( \text{ffCO}_2 \)
inversions to capture this source of uncertainty.

The results of these experiments suggest that the choice of prior emissions estimate and transport
model (among those considered here and currently used in the community) used in our \( \text{ffCO}_2 \)
inversion would result in differences of 15% or less in posterior state-wide \( \text{ffCO}_2 \) emissions in
California, using the observation network from Graven et al. (2018). These differences are
generally not significant, compared to the posterior 2-σ uncertainties of 10 to 15%. In comparison, Graven et al. (2018) found that posterior state-wide ffCO₂ emissions were not statistically different when using temporally-varying emissions from Vulcan, as compared to annual mean emissions from Vulcan or EDGAR, with posterior uncertainties of ±15 to ±17%. Our results may be specific to the California region, observation network and inversion setup we consider here, but we expect that similar differences of 1-15% are likely to be found elsewhere in similar inversions at comparable regional scales. We note that while we have assessed individual contributions to uncertainty in the experiments formulated here, these contributions can also interact with each other. These interactions could act to increase the resulting biases, or partly cancel them, depending on the combination used. The possibility for interacting effects implies that multiple prior emissions estimates and transport models should be used in inversions of real data.

In our results, emissions from many small or rural air basins did not have a significant contribution to the local enhancement of ffCO₂ at the observation sites and were not adjusted by the inversion in most cases (Figure 2, Figure S1). Combined with our experimental setup specifying the magnitude of prior emissions to be equal to true emissions, it might be expected that our results could underestimate the predicted biases in posterior emissions. However, these experiments were designed specifically to quantify representation and transport error using the inversion setup and the observation network from Graven et al. (2018) as a test case. Here, we have assumed the model-measurement mismatch uncertainty matrix is diagonal, following previous work (e.g. Gerbig et al. 2003; Fischer et al., 2017), however the consideration of correlated errors in the uncertainty matrix has also been found to affect posterior emissions for methane in California and reduce their uncertainty at the level of several percent (Jeong et al.,...
Fischer et al. (2017), showed in individual simulation experiments that using either EDGAR, or a spatially uniform flux of 1 μ mol m$^{-2}$ s$^{-1}$ as a biased prior, produced posterior emissions that are substantially closer to true emissions, but only if the prior uncertainties are set large enough to encompass biases in prior emissions. Therefore, further experiments using a different experimental setup such as choice of mismatch error or specification of inversion regions (e.g. to change the inversion region size based on proximity to the observation network, Manning et al., 2011), would help to characterize uncertainties in regional fCO$_2$ inversions and the robustness of posterior estimates to the choices made in the inversion setup.

**Conclusion**

We have shown that atmospheric inversions for the U.S. state of California can reduce a hypothetical bias in the magnitude of prior emissions estimates of fCO$_2$ in California using the ground-based observation network from Graven et al. (2018). Experiments to characterize the effect of differences in the spatial and temporal distribution in prior emissions resulted in biases in posterior state-total emissions with magnitudes of 1-15%, similar to monthly posterior estimates of Basu et al., 2016 for the western United States. Our results highlight the need for (1) temporal variation to be included in prior emissions, (2) different estimates of the spatial distribution of emissions between urban and rural regions to be considered, and 3) representation of atmospheric transport in regional fCO$_2$ inversions to be further evaluated.

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Table 1: The 15 air basins of California with respective emissions as estimated by Vulcan and EDGAR. Also shown are the SD prior uncertainty estimate (Fischer et al., 2017), and difference in magnitude between Vulcan and EDGAR for each air basin. Air basin numbers correspond to those marked in Figure 1.

<table>
<thead>
<tr>
<th>Air Basin</th>
<th>Name</th>
<th>Code</th>
<th>Vulcan (TgC/yr)</th>
<th>EDGAR (TgC/yr)</th>
<th>SD Prior Unc. 1-σ (%)</th>
<th>Vulcan - EDGAR (TgC/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>North Coast</td>
<td>1.NC</td>
<td>1.0</td>
<td>1.6</td>
<td>59</td>
<td>-0.6</td>
</tr>
<tr>
<td>2</td>
<td>Northeast Plateau</td>
<td>2.NP</td>
<td>0.4</td>
<td>1.3</td>
<td>96</td>
<td>-1.0</td>
</tr>
<tr>
<td>3</td>
<td>Sacramento Valley</td>
<td>3.SV</td>
<td>6.8</td>
<td>7.4</td>
<td>8</td>
<td>-0.7</td>
</tr>
<tr>
<td>4</td>
<td>Mountain Counties</td>
<td>4.MC</td>
<td>2.2</td>
<td>2.0</td>
<td>51</td>
<td>0.1</td>
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<tr>
<td>5</td>
<td>Lake County</td>
<td>5.LC</td>
<td>0.1</td>
<td>0.2</td>
<td>65</td>
<td>-0.2</td>
</tr>
<tr>
<td>6</td>
<td>Lake Tahoe</td>
<td>6.LT</td>
<td>0.1</td>
<td>0.1</td>
<td>42</td>
<td>0</td>
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<tr>
<td>7</td>
<td>Great Basin Valleys</td>
<td>7.GBV</td>
<td>0.2</td>
<td>0.6</td>
<td>100</td>
<td>-0.4</td>
</tr>
<tr>
<td>8</td>
<td>San Joaquin Valley</td>
<td>8.SJV</td>
<td>8.6</td>
<td>20.2</td>
<td>35</td>
<td>-11.6</td>
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<tr>
<td>9</td>
<td>North Central Coast</td>
<td>9.NCC</td>
<td>6.0</td>
<td>2.2</td>
<td>71</td>
<td>3.8</td>
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<tr>
<td>10</td>
<td>Mojave Desert</td>
<td>10.MD</td>
<td>6.1</td>
<td>4.3</td>
<td>17</td>
<td>1.8</td>
</tr>
<tr>
<td>11</td>
<td>South Central Coast</td>
<td>11.SCC</td>
<td>4.4</td>
<td>3.4</td>
<td>21</td>
<td>1.0</td>
</tr>
<tr>
<td>12</td>
<td>Salton Sea</td>
<td>12.SS</td>
<td>1.4</td>
<td>1.7</td>
<td>55</td>
<td>-0.3</td>
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<tr>
<td>13</td>
<td>San Francisco Bay</td>
<td>13.SFB</td>
<td>16.4</td>
<td>17.5</td>
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<td>-1.2</td>
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<tr>
<td>14</td>
<td>South Coast</td>
<td>14.SC</td>
<td>26.9</td>
<td>35.5</td>
<td>12</td>
<td>-8.6</td>
</tr>
<tr>
<td>15</td>
<td>San Diego</td>
<td>15.SD</td>
<td>6.6</td>
<td>6.5</td>
<td>10</td>
<td>0.1</td>
</tr>
<tr>
<td><strong>Total California</strong></td>
<td></td>
<td></td>
<td><strong>89.6</strong></td>
<td><strong>104.7</strong></td>
<td><strong>8</strong></td>
<td><strong>-17.8</strong></td>
</tr>
</tbody>
</table>
Figure 1: a. The location of the 9 tower sites in the observation network (marked with black circles): Trinidad Head (THD), Sutter Buttes (STB), Walnut Grove (WGC), Sutro (STR), Sandia-Livermore (LVR), Victorville (VTR), San Bernardino (SBC), Caltech (CIT) and Scripps Institute of Oceanography (SIO). The 15 air basins are marked out with black lines with region 16 representing emission from outside California within the model domain. Underlayed is a map of annual mean ffCO$_2$ emissions from the Vulcan v2.2 emission map within the United States and EDGAR v4.2 (FT2010) for emission from outside the U.S. b. Vulcan diurnal emissions normalized to campaign averaged emissions for the 3 campaigns, c. Scaled EDGAR subtracted from Vulcan emissions map, where EDGAR has been scaled to have the same air basin total emissions. The inset shows an enlarged view of southwestern California. d. Average monthly emissions normalized to Vulcan annual emissions. Shown in both b and d is EDGAR annual invariant emissions (grey).
<table>
<thead>
<tr>
<th>Transport Model</th>
<th>Meteorology</th>
<th>Domain</th>
<th>Model Resolution</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>WS-LBL (v3.5.1)</td>
<td>WRF</td>
<td>North America</td>
<td>1km, 4km, 12 km, 36 km</td>
<td>50 / 16 km, 1 hour (Lin, 2003; Nehrkorn et al., 2010);</td>
</tr>
<tr>
<td>WS-CTL</td>
<td>WRF (v2.1.2)</td>
<td>North America</td>
<td>0.1°, 1°</td>
<td>29 / 25 km, 1 hour (Carbon Tracker, 2017)</td>
</tr>
<tr>
<td>UM-NAME</td>
<td>Unified Model</td>
<td>Global</td>
<td>17km, 25km</td>
<td>59 / 29 km, 3 hours (Ryall et al., 1998)</td>
</tr>
</tbody>
</table>

Table 2: Comparison of the three atmospheric transport models used in this study.
Figure 2: The average $f$FCO$_2$ signal (ppm) simulated by each atmospheric transport model as a result of emissions from the 6 largest emitting air basins and one outside California region at each observation site over the three measurement campaigns. Signals were simulated based on the EDGAR emission map.
Figure 3: (a) Statewide and (b) individual air basin inversion results for an error in the magnitude of prior emissions. Prior emissions are given by EDGAR and true emissions are given by EDGAR scaled to Vulcan total in each air basin. Air basin results are shown for Sacramento Valley (3.SV), San Francisco Bay (13.SFB), San Joaquin Valley (8.SJV), Mojave Desert (10.MD), South Coast (14.SC) and San Diego (15.SD). Prior results are presented by black markers and posterior results are represented by colored markers. Filled markers show results using SD prior uncertainty and empty markers show results using 70% prior uncertainty. The prior bias in each air basin is given by the dashed lines in (b) with SD prior uncertainty (dark grey) and 70% prior uncertainty (light grey). Prior and posterior uncertainties are expressed as 2-σ. The bottom plot (c) shows the mean signal error in simulated average fCO₂ concentration. Mean signal error is calculated by subtracting the average true signal from the average prior signal. Error lines are drawn between the maximum and minimum signal bias per campaign.
Figure 4: (a) Statewide and (b) individual air basin inversion results for an error in the spatial distribution of prior emissions. Prior emissions are given by EDGAR scaled to Vulcan emissions totals in each air basin and true emissions are given by Vulcan. The bottom plot (c) shows the mean signal error in simulated average $\text{ffCO}_2$ concentration.
Figure 5: (a) Statewide and (b) individual air basin inversion results for an error in the temporal distribution of prior emissions. Prior emissions are given by temporally varying Vulcan and true emissions are given by annually averaged Vulcan. Prior emissions were scaled to be the equal in magnitude to annually averaged Vulcan emissions. The bottom plot (c) shows the mean signal error in simulated average fCO$_2$ concentration.
Figure 6: Inversion results for the experiment where the atmospheric transport in the prior simulation uses WS-CTL or UM-NAME but the atmospheric transport in the true simulation uses WS-LBL. Posterior statewide emissions (a, b), individual air basin emissions (c, d), and percentage error in simulated average fCO₂ concentration (e, f) are shown with no outlier removal (first column) and outliers removed (second column). Prior and true emissions are given by annually averaged Vulcan.
Figure 7: All simulated ffCO$_2$ from May (first column), October-November (second column), and January-February (third column). Simulated ffCO$_2$ using W-S-LBL are shown in black markers (triangles for May, squares for Oct-Nov and diamonds for Jan-Feb) whilst prior W-S-CTL signals are shown in blue and UM-NAME signals in magenta. All simulated signals are generated using the Vulcan gridded emissions map. The fourth column shows true vs prior ffCO$_2$ signals, with colors corresponding to models and markers corresponding to campaigns. Outliers omitted from the standard inversion are shown by an x.
Supplementary
Figure 1: Posterior air basin emissions for an error in the magnitude of emission. Filled markers show posterior results using SD prior uncertainty and clear markers represent 70\% prior uncertainty. The prior bias in each air basin is given by the dashed lines with SD prior uncertainty (dark grey) and 70\% prior uncertainty (light grey). Prior and posterior uncertainties are expressed as 2-\sigma.
Figure 2: EDGAR grid cells compared to Vulcan (regridded to EDGAR native 0.1° resolution). Mean emissions are in units of gCO₂ m² yr⁻¹.
Figure 3: PBLH error versus $\text{ffCO}_2$ error (top) and wind speed versus $\text{ffCO}_2$ error (bottom). Error in PBLH/wind speed was calculated by subtracting UM from WRF estimates. $\text{ffCO}_2$ signal error was calculated by subtracting UM-NAME from WS-LBL signals.
Figure 4: ffCO₂ signal bias for aggregation error (black) and no aggregation error (red) in modelled atmospheric transport. For each box the central mark indicates the median, and the left and right edges indicate the 25th and 75th percentiles, respectively. Dashed lines extend to the most extreme data points not considered outliers. Error was calculated by subtracting WS-CTL signals (generated using native 0.1° resolution and UM-NAME resolution footprints respectively) from UM-NAME signals.
Figure 5: PBLH versus ffCO$_2$ (top) and wind speed versus ffCO$_2$ (bottom) using MYNN2 PBLH, Noah/LSU wind speed, and WS-LBL ffCO$_2$ signal estimates.