



GREENHOUSE GASES MEASUREMENT AND MODELING EFFORTS IN CALIFORNIA, USA

Research Division
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Executive Summary

The California Air Resources Board (CARB) in the United States has established an extensive greenhouse gas (GHG) monitoring network with seven stations strategically distributed across the State of California. These stations are equipped with a variety of state-of-the-art analyzers that measure a variety of GHGs including carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O). The sampling and measurement method developed at CARB's network follows a stringent approach as described in the Quality Assurance Project Plan (QAPP) to ensure the data meet the highest level of accuracy and precision. Data from the network provide the basis for comprehensive modeling and analysis approaches which allows us to generate a 'top down' estimate of greenhouse gas emissions.

CARB has performed three major modeling applications including atmospheric inverse modeling, tracer-tracer analysis, and source apportionment analysis to inform the emission inventory based on the observational data from the monitoring network. Atmospheric inverse modeling estimates GHG emissions with an atmospheric transport model, a prior inventory for GHG emissions, and a statistical technique to minimize differences between measured and predicted GHG concentrations. The tracer-tracer analysis provides estimation on GHG emissions based on their concentration correlations with other pollutants with known emission estimates in the well-mixed air mass with GHGs. With the availability of additional speciated concentration data of volatile organic compounds (VOCs), source apportionment analysis can be conducted to further identify major sources of GHGs in the absence of bottom-up GHG reporting programs.

This document describes the methods used for the measurement of greenhouse gases (GHGs) conducted by the California Air Resources Board (CARB) in California, United States, and the modeling efforts for inventory evaluation based on the observational data. An overview of CARB's GHG monitoring network and measurement methods are provided in Sections 1 and 2, atmospheric modeling and analysis approaches are described in Section 3.

1 Atmospheric Observation Sites

CARB has an extensive GHG monitoring and measurement program to study the regional and local emissions of important GHGs in California. CARB initiated the first subnational GHG monitoring network in 2010 (with pilot measurements in 2007) to study the regional GHG emissions trends throughout the state and evaluate the statewide inventories. The statewide inventory has also been regionally disaggregated to allow for regional evaluations of emissions. Data from this network form the basis of a comprehensive statewide inverse modeling and various trends assessment analyses allowing us to generate a 'top down' emissions estimate, and in certain cases disaggregate those emissions spatially and by industrial sector.

1.1 CARB's GHG Monitoring Network

The network currently has seven CARB-operated monitoring stations located at strategically selected regional sites throughout California (Figure 1 and Table 1). CARB has equipped these stations with state-of-the-art analyzers that measure a variety of important greenhouse gases such as carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O) with high levels of accuracy and precision. Additionally, carbon monoxide (CO) is also measured as a tracer for anthropogenic emissions. CARB is also deploying analyzers capable of measuring the isotopic signature of CO₂ and CH₄ to potentially further refine the source attribution of the inventory. The monitoring stations were selected to ensure measured CH₄ concentrations represent regional-scale monitoring and they are generally located in rural areas that are not directly impacted by local sources.

The seven GHG monitoring stations operated by CARB are located within the three key regions of California: Central Valley, and South Coast, where GHGs are largely emitted from anthropogenic sources. Sites TSB and STB are located in the north of the Central Valley, while MAD, TRA and ARV are located in the south of the Central Valley. These latter three sites measure methane from a complex mix of sources found in the Central Valley, including from oil and gas fields, landfills, and dairies. The SBC site is situated on a tall tower, which can cover a larger area and makes it possible to collect air samples at multiple heights. Such

measurements are useful to provide the vertical profile of measured GHGs. As a unique site, MWO will be discussed in detail in Section 1.2.



In addition, CARB also collaborates with research partners on several other monitoring locations throughout California, e.g., the Los Angeles Megacity Carbon Project (MCP, Figure 2 and Table 1). The MCP network was started in 2012 and now includes 13 tower/rooftop sites to conduct continuous measurements of CH₄ concentrations in the Southern California region (Verhulst et al., 2017). Two of these sites, WMO and SBC belong to the CARB's GHG Monitoring Network. Three of these sites (SCI, VIC and LJO) are considered background sites providing information on GHG levels without the influence of sources in Los Angeles. Every site is equipped with at least one in situ gas analyzer providing high-accuracy, continuous measurements of carbon dioxide (CO₂). Nearly every site also includes measurements of methane (CH₄) and about half of the sites currently measure carbon monoxide (CO).

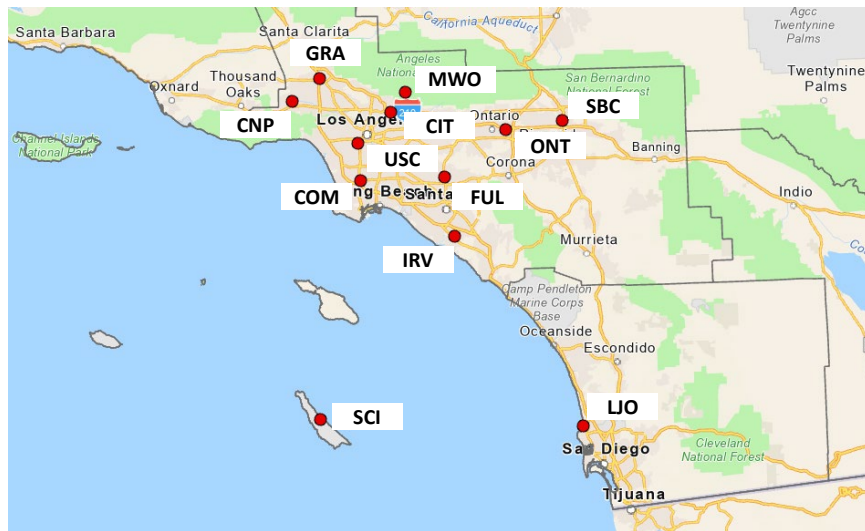


Table 1. Site information on CARB GHG monitoring network and Los Angeles Megacity Carbon Project

| Site Name | ID | Latitude | Longitude | Inlet Height (m) | Elevation (m) | Pollutants Measured |
|--|-----|----------|-----------|------------------|---------------|--|
| CARB GHG Monitoring Network | | | | | | |
| Mt. Wilson | MWO | 34.22 | -118.06 | 4 | 1732 | CH ₄ , iCH ₄ , CO ₂ , iCO ₂ , N ₂ O, CO, BC |
| San Bernardino | SBC | 34.09 | -117.31 | 58 | 300 | CH ₄ , CO ₂ , N ₂ O, CO |
| Arvin | ARV | 35.24 | -118.79 | 10 | 158 | CH ₄ , CO ₂ , N ₂ O, CO |
| Madera | MAD | 36.87 | -120.01 | 10 | 81 | CH ₄ , CO ₂ , N ₂ O, CO |
| Tranquility | TRA | 36.63 | -120.38 | 10 | 63 | CH ₄ , CO ₂ , N ₂ O, CO |
| Sutter Buttes | STB | 39.21 | -121.82 | 10 | 645 | CH ₄ , CO ₂ , N ₂ O, CO |
| Tuscan Buttes | TSB | 40.26 | -122.09 | 10 | 562 | CH ₄ , CO ₂ |
| Los Angeles Megacity Carbon Project | | | | | | |
| Compton | COM | 33.87 | -118.28 | 45 | 9 | CH ₄ , CO ₂ , CO |

| | | | | | | |
|----------------------------|-----|-------|---------|-----|------|--|
| Granada Hills | GRA | 34.28 | -118.47 | 51 | 391 | CH ₄ , CO ₂ , CO |
| Ontario | ONT | 34.06 | -117.58 | 41 | 9 | CH ₄ , CO ₂ |
| Victorville | VIC | 34.61 | -117.29 | 100 | 1370 | CH ₄ , CO ₂ |
| San Clemente Island | SCI | 32.92 | -118.49 | 27 | 489 | CH ₄ , CO ₂ , N ₂ O, CO |
| Downtown LA | USC | 34.02 | -118.29 | 50 | 55 | CH ₄ , CO ₂ , N ₂ O, CO |
| Fullerton | FUL | 33.88 | -117.88 | 50 | 75 | CH ₄ , CO ₂ , N ₂ O, CO |
| Irvine | IRV | 33.64 | -117.84 | 20 | 10 | CH ₄ , CO ₂ |
| Canoga Park | CNP | 34.19 | -118.60 | 15 | 245 | CH ₄ , CO ₂ |
| Pasadena | CIT | 34.14 | -118.13 | 10 | 230 | CH ₄ , CO ₂ , CO |
| La Jolla | LJO | 32.87 | -117.25 | 13 | 0 | CH ₄ , CO ₂ , N ₂ O, CO |

1.2 Mt. Wilson Observatory Station (MWO)

CARB also operates a unique site MWO, which is located on top of the San Gabriel mountain range (34°13'21" N, 118°3'42" W) that overlooks the Los Angeles Basin (LA Basin). Its unique location and meteorological conditions allow measurement of well-mixed urban air pollution that typically travels toward the monitoring site from regions between southeast (SE) and west-southwest (WSW). The upslope airflow, rapid atmospheric mixing from the growth of boundary layer height, and relatively consistent meteorological pattern between 10 AM and 6 PM in the LA Basin make this monitoring site ideal for long-term measurements of urban emissions (Figure 3).

The station is equipped with the most comprehensive set of instrument/analysis capabilities. The instruments perform real-time measurements of CH₄, iCH₄, CO₂, iCO₂, N₂O, CO, BC, as well as isotopic CH₄, CO₂. Whole-air canister samples are also collected at MWO periodically to measure speciated volatile organic compounds (VOCs) that were derived from the LA Basin. Data collected at MWO have been used in various modeling practices including inverse modeling, tracer-tracer analysis, and source apportionment analysis, to understand GHG emissions in the LA Basin.

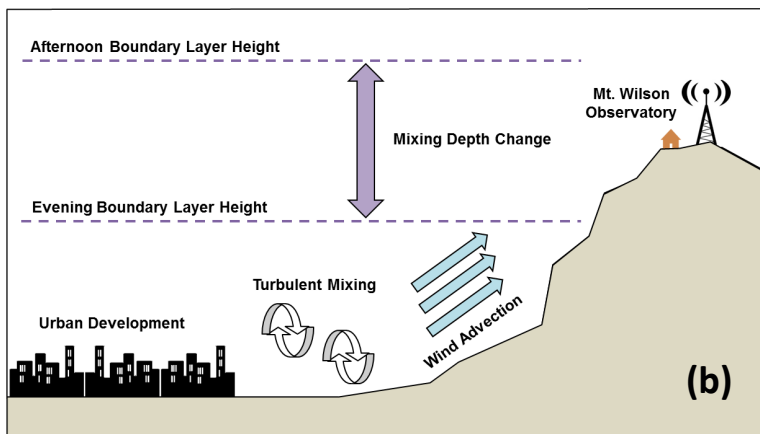


Figure 3. (a) View of the Los Angeles Basin from the MWO monitoring site; (b) Illustrated depiction of air mass transport to MWO (Kuwayama et al., 2019); (c) Instrumentation set-up at MWO

2 Measurement Methods

This section briefly describes the sampling and measurement method deployed at CARB's GHG monitoring network. More details are available in the Quality Assurance Project Plan (QAPP)(California Air Resources Board, 2019) . The QAPP document details a comprehensive project plan to develop, instrument, and implement the GHG monitoring network in California. The document also details CARB's data processing and review processes to ensure that the network data meets the World Meteorological Organization (WMO) standards. This approach allows CARB's network data to meet the required level of accuracy and precision to be consistent with the collaboratint research stations throughout California and to provide to provide the quality of data needed for a robust ambient based evaluation of the statewide emissions inventory.

2.1 Instrumentation

The CH₄ and CO₂ gas analyzers used at CARB's network stations are cavity ringdown analyzers that are capable of both high accuracy and fast measurements of CH₄ (range = 0 – 20 ppm), CO₂ (range = 0 – 1,000 ppm), and water vapor (range = 0 – 70,000 ppm). CARB's network uses three different models of cavity ring-down spectrometer (CRDS) instruments (Picarro Models G1301, G2201-i and G2301, Picarro Inc.). The Picarro 1301 is an older model that has been operated successfully at five sites. The 2301 and 2201-I models are newer. The 2301 model replaced the 1301 model as Picarro's offering for a CO₂, CH₄ and H₂O analyzer. The 2201-I model, used at MWO only, provides additional capabilities to measure both ¹²C and ¹³C isotopes in CH₄ and CO₂, allowing for source apportionment between biogenic and fossil fuel sources of CH₄ and CO₂.

While the instruments deployed at CARB's network stations have different model numbers, all instruments have very similar performances in terms of measurement precision and drift as listed in Table 2. All instruments can deliver highly accurate data when calibrated and maintained appropriately. The calibration of the instruments is described in Section 2.3.

Table 2. Information for three models of Picarro Cavity Ring down Spectrometer (CRDS)

| CRDS Instrument | Precision | Precision (1s - 600 secs) | Drift (Day) | Drift (Month) |
|-----------------|------------------------|---------------------------|-----------------|---------------|
| Picarro 1301 | <1.5ppb (1s - 30 secs) | n/a | 1.5 ppb (30hrs) | n/a |

| | | | | |
|-----------------------|---|----------|--------------------------------------|-------|
| Picarro 2201-I | 6 ppb @ 2000 ppb reading (1s - 30 secs) | n/a | < 2.3 ppb (24hrs) @ 2000 ppb reading | n/a |
| Picarro 2301 | < 0.5 ppb (1s - 5 secs) | 0.22 ppb | < 1 ppb (24 hrs) | 3 ppb |

The N₂O and CO instruments deployed at the network stations are Off-axis Integrated Cavity Output Spectroscopy N₂O/CO Analyzers (LGR Model 907-0015; Los Gatos Research Inc.). These monitors are capable of both high accuracy and fast measurements of N₂O (range = 0 – 10 ppm), CO (range = 0 – 10 ppm), and water vapor (range = 0 – 70,000 ppm).

2.2 Sample Collection

At each site, the gas analyzers are housed at the base of the tower in a building or portable laboratory built in a trailer or modified sea container. Air is drawn down the tower through sampling lines. Each line has a high-surface-area PTFE 0.2 um filter capsule on the inlet.

Auxiliary pumps are used upstream of the analyzers so that air is pushed rather than pulled through the analyzers. Some advantages of this design are that (1) the condenser works more effectively at higher pressure, (2) the ambient air is delivered to the analyzers at a pressure similar to the calibration gases, and (3) there is reduced likelihood that leaks will affect the measurements. Disadvantages are that water is more likely to condense in the sampling lines and that the sample air is exposed to pumps and associated components, which are not included in the calibration path.

To reduce the measurement artifacts and system damage caused by the presence of water vapor, the sampling system minimizes humidity differences between the sample airstream and standards by passing sample and standard gas through Nafion membrane dryers. The sample air is dried, while standard gases are humidified.

2.3 Calibration

All the gaseous analyzers deployed at CARB's network are periodically calibrated to ensure the instruments' accuracy and precision. At each site, the "Periodic Two-point Calibration and Target Check" procedure is applied. Every four hours, each analyzer is challenged with two gas standards, with known gas concentrations, spanning typical ambient concentrations to calibrate the instrument. A third standard with known concentrations is then used to calculate the instruments' accuracy and precision. The three gases are 'secondary transfer standards, meaning they are provided by commercial gas suppliers, but their concentrations

are verified using the primary gas standards provided by the Earth System Research Laboratory at National Oceanic and Atmospheric Administration (ESRL at NOAA).

2.4 Data Review

CARB conducts Quality Assurance/Quality Control (QA/QC) procedures for data review and validation (California Air Resources Board, 2019). The first level review is conducted by CARB site operators. In this review, all the preliminary data of each month are reviewed and flagged for abnormal data points (e.g., instrument malfunction, filter changes, target accuracies). Upon the completion of the first level review, the field check sheets, monthly site activity log, and the review data are further reviewed by an assigned CARB second-level review staff to confirm data quality. CARB site operators fill out field check sheets to document field activities. Following standard air monitoring practices, CARB performs instrument calibrations every six months and after major instrument maintenance to verify the instrument accuracies.

2.5 Additional Measurements for VOC Speciation

In previous projects at MWO, CARB collected whole-air canister samples and quantified the concentration of VOCs using three gas chromatography systems (HP 6890, Agilent Technologies) that integrated electron-capture detectors, flame ionization detectors, and a quadrupole mass spectrometer. The data collected has been utilized in source apportionment analysis to evaluate major sources of GHGs in the Los Angeles basin (Hsu et al., 2010; Kuwayama et al., 2019). CARB is planning to deploy an auto-sampling GC-MS system at MWO to measure speciated concentration of VOCs and fluorinated gas (F-gases) at 30 minutes intervals, using Thermo Scientific ISQ LT Single Quadrupole GC-MS System coupled with a thermal desorption system (CIA Advantage and Unity 2) from Markes International. The system will be able to quantify compounds with target concentration less than or equal to 100 ppt with uncertainty of $\pm 20\%$, in both ambient and canister samples.

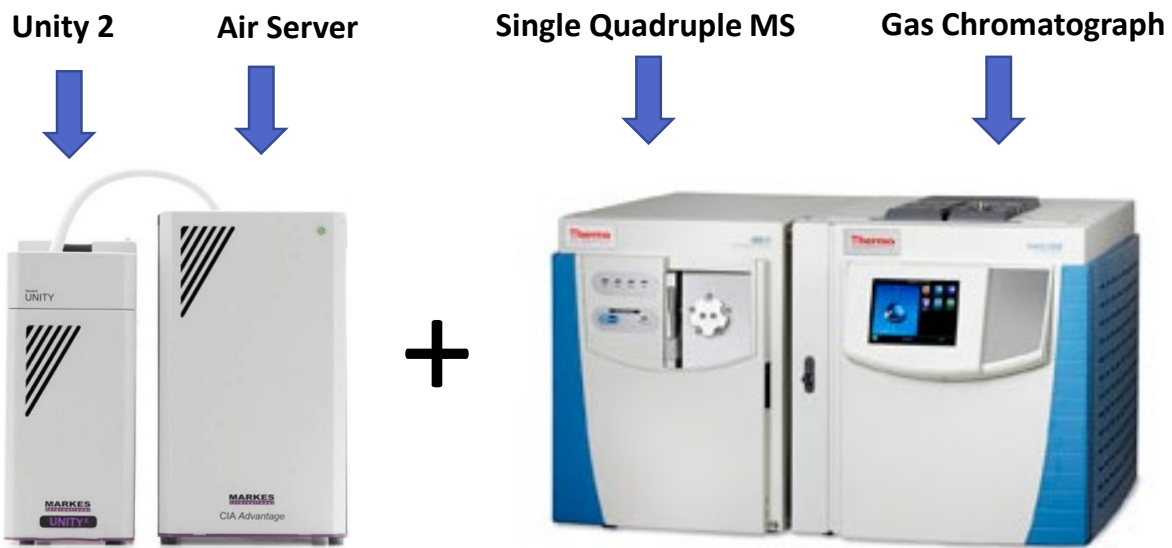


Figure 4. Schematic of the auto-sampling GC-MS system

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3 Atmospheric Modeling and Analysis

CARB has performed two major modeling applications based on the observational data obtained from the monitoring network (1) inverse modeling to evaluate the statewide emission inventory, (2) tracer-tracer analysis and source apportionment analysis to characterize regional emissions in the LA basin based on the data collected at MWO. Detailed information on CARB’s modeling and analysis approaches using observational data is provided in this section.

3.1 Inverse modeling

Atmospheric GHG measurements from networks of towers, when combined with inverse model estimation techniques provide a top-down estimate of current GHG emissions. It is a ‘second opinion’ on the inventory that can serve as an indicator of where more work may be needed to create a more robust inventory. Atmospheric inverse methods, which estimate the GHG emissions from in-situ (i.e., instrumentation located directly at the site and in contact with the air) and/or remotely sensed GHG concentration measurements and modeled meteorology, provides an independent test of GHG emission inventory and have been widely applied at both global and regional scales. In general, the components of atmospheric inverse modeling are GHG measurements, an atmospheric transport model, a prior inventory for GHG emissions, and a statistical technique to minimize differences between measured and predicted GHG concentrations (Figure 5).

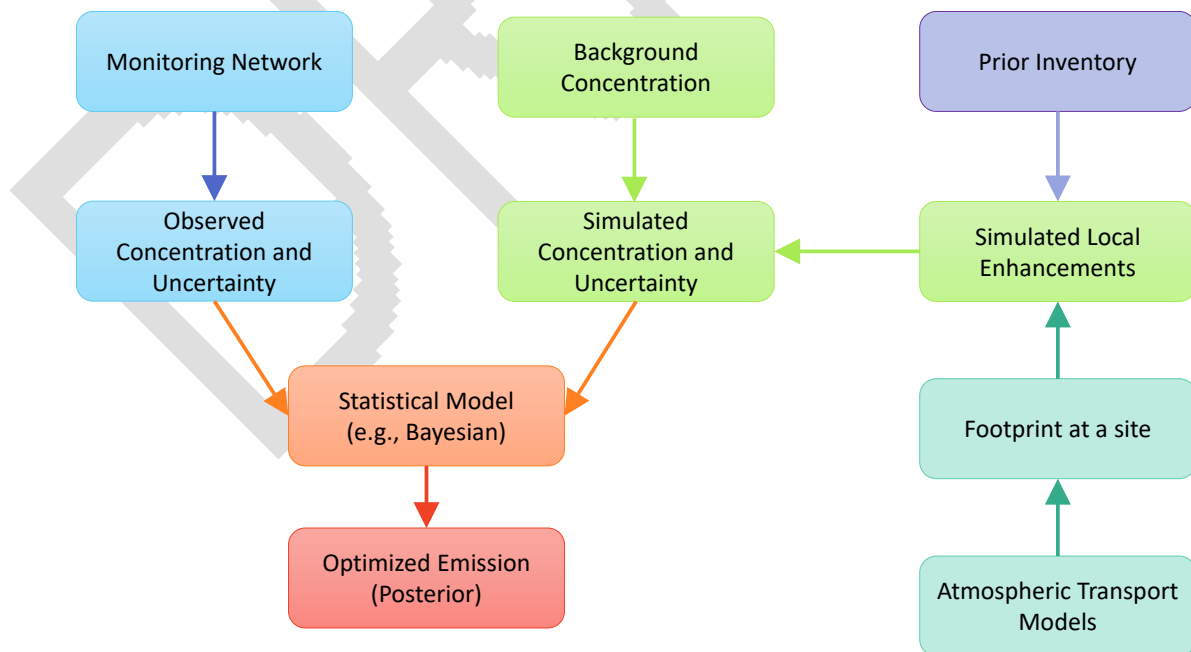


Figure 5. Schematic of the Inverse Modeling Framework

3.1.1 Atmospheric Transport Model

Atmospheric transport models calculate the atmospheric footprint of a site from particle trajectories simulated using a Lagrangian Particle Dispersion Model (LPDM) driven by a meteorological model. 'Footprint' represents the sensitivity of signals measured at the receptor location (i.e., measurement site) to surface emissions across the landscape (i.e., concentration change at the measurement site due to unit emission flux from each grid cell). The footprint provides information on how much the concentration at the receptor location changes when the surface emission changes by one unit.

Figure 6 shows an example of particles moving from upwind locations to the measurement site (i.e., receptor) using the transport model. In the time-reverse sense, the model transports ensembles of particles (air parcels) backwards in time from a receptor point. The model defines paths traveled by parcels of air, or trajectories, which are a basis for footprint estimates. By releasing particles backwards, the model identified the origin of emission sources that contribute to the receptor. The identified contribution from each location (e.g., certain grid cell) is combined to generate a map of the averaged (e.g., hourly) measurement sensitivity to emissions, or footprint (Figure 7). This footprint is then multiplied by the prior emissions to produce GHG concentrations that account for upwind emission sources.

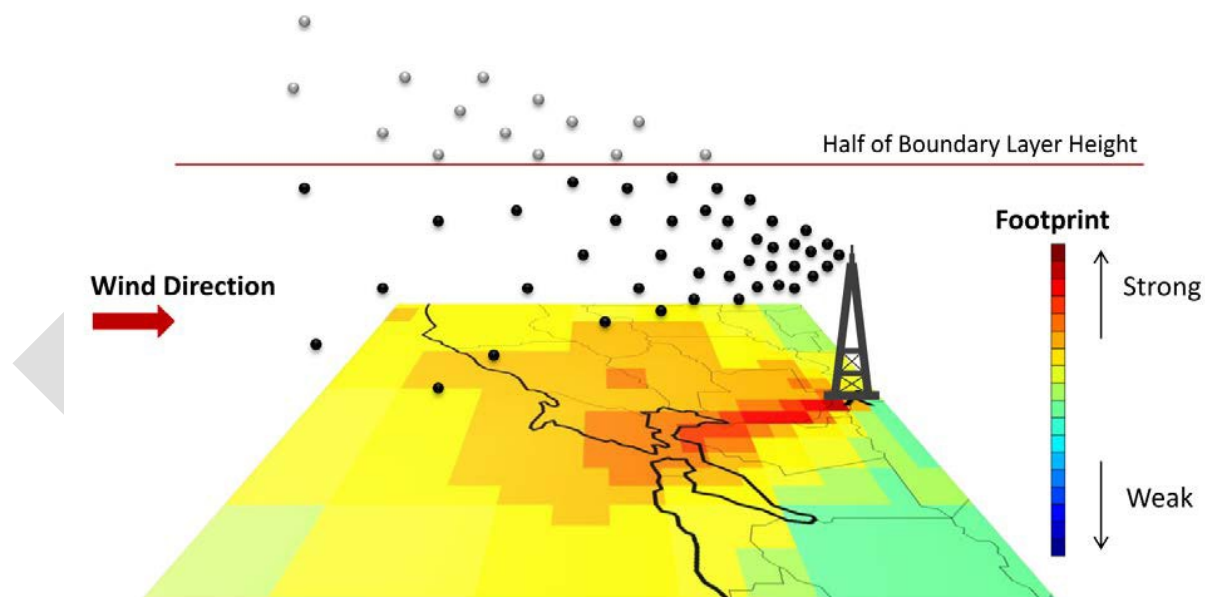


Figure 6. Schematic diagram showing simulations of backward (opposite to the wind direction) particle trajectories from a tower that are used for footprint calculations. Footprints at the grid cells near the receptor are strong because more particles pass over those grid cells before they are advected and dispersed into different places. The particles above $\frac{1}{2}$ PBLH (colored in gray) do not contribute to the footprint strength (Fischer et al., 2016).

Throughout the decades, CARB has conducted independent studies utilizing two types of atmospheric transport models, with the Weather Research and Forecasting (WRF) model coupled with different types of LPDM, including the Stochastic Time-Inverted Lagrangian Transport model (STILT) (Bagley et al., 2017; Fischer et al., 2016; Jeong et al., 2013; Jeong et al., 2016) or FLEXible PARTicle dispersion model (FLEXPART) (Cui et al., 2019). Model setups of WRF differed by the extension and spatial resolution of nested domains, initial boundary meteorological conditions and configuration of physical schemes for radiative transfer, planetary boundary layer, land surface, microphysics, and cloud formation. The uncertainty in the transport model predictions is generally evaluated using a combination of meteorological measurements, such as temperature, relative humidity, wind direction and wind speed and planetary boundary layer heights (PBLH), as well as the measured concentration of tracers with well-established inventory (such as CO).

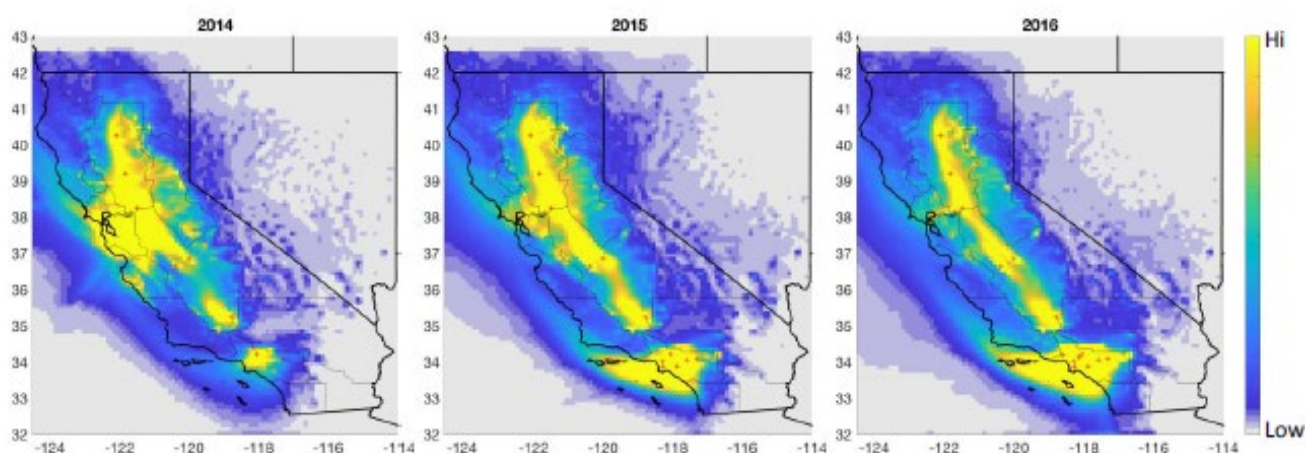


Figure 7. Examples of averaged footprint maps of the receptors to the surface fluxes (Cui et al., 2019)

3.1.2 Prior Inventories

Emission estimated from prior inventories is used to multiply by the simulated footprint to produce GHG concentrations that account for upwind emission sources. Two types of prior inventories have been used in CARB's inverse modeling framework (Figure 8), including California Greenhouse Gas Emission Measurements (CALGEM) and U.S. Environmental Protection Agency (USEPA) inventory of US greenhouse gas emissions and sinks (EPA-GHGI).

The CALGEM emission model provides emissions by sector at a high spatial resolution ($0.1^\circ \times 0.1^\circ$) for California. The CALGEM model has seasonal components for wetlands and crop agriculture only, and these seasonal emissions are combined with nonseasonal emissions to construct monthly emission maps for inversions. The CALGEM prior emissions distributions are scaled to match 2012 CARB state totals for anthropogenic emission sectors

(California Air Resources Board) with small adjustments for some regions and sectors. The EPA-GHGI inventory includes emission data for California region spatially extracted from a gridded national inventory of U.S. methane emissions which is designed to be consistent with the 2016 edition U.S. Environmental Protection Agency (EPA) inventory of US greenhouse gas emissions and sinks (GHGI) for the year 2012 (U.S. Environmental Protection Agency) Both of these inventories were gridded at 0.1×0.1 degree spatial resolution, and the annualized aggregated emissions were extracted for the emission analysis. The sectors of wetland and crop agriculture have seasonal variations in CALGEM, and the sectors of manure management, natural gas and petroleum production, stationary combustion, and forest fires have monthly/daily variations in EPA-GHGI.

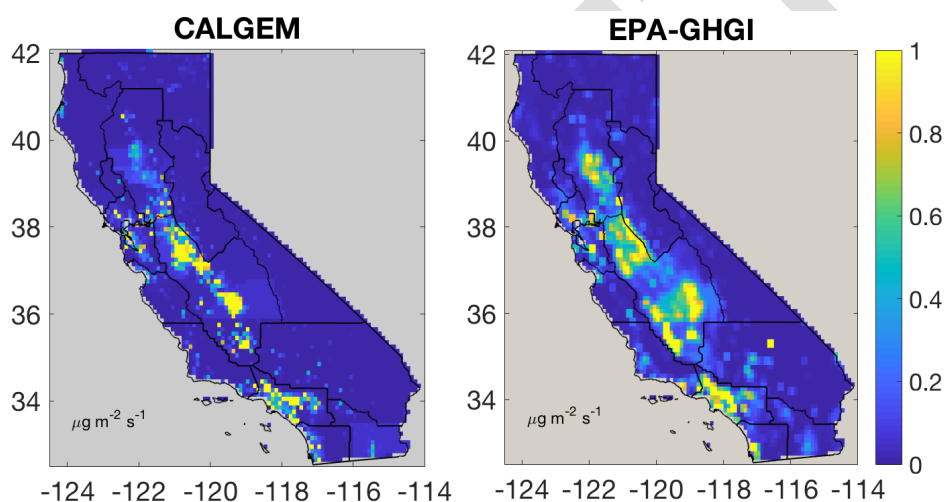


Figure 8. Two prior inventories (CALGEM and EPA-GHGI, unit: $\mu\text{g m}^{-2} \text{s}^{-1}$) used in the inversion estimates (Cui et al., 2019).

A multivariate analysis by combining the different sector groups from the two inventories in different combinations to construct a hybrid inventory that would provide the additional spatially resolved prior information. The hybrid adds another dimension to reducing the prior inventory uncertainty in the inverse system and can be used to better understand the emission sectors in the region.

3.1.3 Statistical Technique - Bayesian Inference

Predicted concentration from the footprint and prior inventory represents local enhancements of emissions. Upwind background concentrations entering the study domain are added, to compare predictions with ambient observations which include both local and background signals. A statistical technique is then deployed to optimize GHG fluxes in such a way that simulated atmospheric GHG concentrations agree better with observations.

Most atmospheric inverse modeling efforts are based on Bayesian inference and implemented in a variety of ways. The Bayesian inversion model optimizes a set of scaling factors, minimizing the difference between measurements and predictions given the uncertainties in the prior emissions and model predictions. Uncertainties of model predictions are propagated from the uncertainties associated with atmospheric measurements, background concentration and the atmospheric transport model. The result of the Bayesian inverse model is a set of optimized scaling factors for source emissions. The final product of the inverse modeling approach - optimized posterior emissions, is calculated by multiplying the optimized scaling factors by a priori emissions.

3.2 Tracer-Tracer Analysis

The tracer-tracer analysis is used to understand GHG emissions based on their concentration correlations with other pollutants with known emission estimates in the well-mixed air mass with GHGs. CARB has applied this approach to the observational data collected at MWO to characterize GHG emissions in the LA basin. Consistent ratio of CH₄ and CO collected at MWO indicates that these gases are well-mixed before reaching the site and the emission source contributions of both compounds are reasonably constant. Since CH₄ and CO are considered non-reactive on the time scale of dispersion within the Los Angeles urban area and their emission sources are likely to be similarly distributed (e.g., associated with human activities), they are subject to similar scales of atmospheric transport and dilution. CARB studies used linear regression models between long-term ambient CH₄ and CO measurements to derive the statistical relationship between ambient CH₄ and CO concentrations and such relationship was used as a metric to calculate CH₄ emissions in the LA basin using well-documented CO emissions (Hsu et al., 2010; Kuwayama et al., 2019).

3.3 Source Apportionment Analysis

Source apportionment analysis helps evaluate the major sources of GHGs and can be conducted if additional VOC speciation data are available (Kuwayama et al., 2019). Source apportionment models are generally receptor models that use mathematical procedures to identify and quantify the sources of air pollutants and their impacts at a certain site (the receptor), primarily based on the speciated concentration measurements at the receptor. One typical source apportionment model is the Positive Matrix Factorization (PMF) Model developed by USEPA (U.S. Environmental Protection Agency). It is a multivariate receptor model that quantitatively determines the contribution of air pollution concentrations in a data matrix through the identification of mathematically unique signatures, or chemical profiles. CARB performed source apportionment analysis based on concentration of 21 chemical compounds (including CO, CO₂, CH₄, alkanes, alkenes, benzene, ethylbenzene and xylene)

from whole-air canister samples collected at MWO between 2014 and 2016. The analysis identified major emission categories for CH₄, such as landfills, petroleum gas and industrial processes, fossil fuel combustion, petroleum refining, natural gas and biogenic sources (Kuwayama et al., 2019).

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