



# Top-down methane emissions estimates for the San Francisco Bay Area from 1990 to 2012



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## HIGHLIGHTS

- Top-down methane emissions estimate used to evaluate a bottom-up emissions inventory.
- Estimates combine two decades of CH<sub>4</sub>:CO enhancement ratios at 14 sites with CO emission inventory.
- Resulting CH<sub>4</sub> emissions 1.5–2 times greater than bottom-up inventory.

## ARTICLE INFO

### Article history:

Received 10 November 2014

Received in revised form

28 January 2015

Accepted 29 January 2015

Available online 30 January 2015

### Keywords:

Methane emissions

Emissions inventory

Greenhouse gas

Inventory verification

Top-down estimates

## ABSTRACT

Methane is a potent greenhouse gas (GHG) that is now included in both California State and San Francisco Bay Area (SFBA) bottom-up emission inventories as part of California's effort to reduce anthropogenic GHG emissions. Here we provide a top-down estimate of methane (CH<sub>4</sub>) emissions from the SFBA by combining atmospheric measurements with the comparatively better estimated emission inventory for carbon monoxide (CO). Local enhancements of CH<sub>4</sub> and CO are estimated using measurements from 14 air quality sites in the SFBA combined together with global background measurements. Mean annual CH<sub>4</sub> emissions are estimated from the product of Bay Area Air Quality Management District (BAAQMD) emission inventory CO and the slope of ambient local CH<sub>4</sub> to CO. The resulting top-down estimates of CH<sub>4</sub> emissions are found to decrease slightly from 1990 to 2012, with a mean value of  $240 \pm 60$  GgCH<sub>4</sub> yr<sup>-1</sup> (at 95% confidence) in the most recent (2009–2012) period, and correspond to reasonably a constant factor of 1.5–2.0 (at 95% confidence) times larger than the BAAQMD CH<sub>4</sub> emission inventory. However, we note that uncertainty in these emission estimates is dominated by the variation in CH<sub>4</sub>:CO enhancement ratios across the observing sites and we expect the estimates could represent a lower-limit on CH<sub>4</sub> emissions because BAAQMD monitoring sites focus on urban air quality and may be biased toward CO rather than CH<sub>4</sub> sources.

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## 1. Introduction

From global to local scales, greenhouse gas (GHG) emission inventories are an essential element of efforts to address climate change. In California, USA, the State legislated mandatory reductions in GHG emissions to meet 1990 emissions levels by 2020 (AB-32, 2006). The San Francisco Bay Area of California has GHG emissions that are reported as greater than those of Austria, Peru or Portugal (WRI, 2014; BAAQMD, 2014). According to the current inventory of the Bay Area Air Quality Management District

(BAAQMD), the Bay Area emitted 125,500 metric tons of methane in 2011, or 3.5 million tons CO<sub>2</sub>-equivalent, assuming a 100 yr global warming potential (GWP = 28) from the IPCC 5th assessment (IPCC, 2013, Table 8.A.1).

Currently, regional, state, and federal agencies, including the BAAQMD, estimate GHG emissions using bottom-up inventory methods that rely on a combination of activity data, emission factors, biogeochemical models and other information. Recent emission evaluations based on ambient measurements show that methane emissions for the US as a whole (e.g., Miller et al., 2013) and in California (e.g., Wennberg et al. 2012, Hsu et al. 2010; Singh et al. 2010; Jeong et al. 2013; Peischl et al. 2013) are underestimated by ~50% or more depending upon the area. Specific to oil and gas sector methane emissions, two meta-analyses, one for the US

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(Brandt et al. 2014) and another for California (Jeong et al. 2014) reached similar conclusions. Relevant to urban areas, the US-EPA recently released a report identifying uncertainty in methane emissions from the natural gas distribution system as an area in need of further research (US-EPA, 2014). For the Bay Area, which has a number of refineries and extensive natural gas use, a 50% underestimation would imply additional annual emissions of about 3.5 million tons CO<sub>2</sub>-equivalent.

From national to local scales, GHG emissions inventories are relatively new compared with the inventories for criteria pollutants. In particular, emissions of a primary criteria pollutant like carbon monoxide (CO) are estimated with greater accuracy. Here we estimate emissions of methane (CH<sub>4</sub>) for the Bay Area using the top-down approach of Hsu et al. (2010), scaling the better estimated emissions of CO to CH<sub>4</sub> using the ratio of local enhancements, CH<sub>4,local</sub>:CO<sub>local</sub>, where “local” indicates local atmospheric mixing ratio increments above the background inflow of CH<sub>4</sub> and CO to the region of interest.

We estimate CH<sub>4</sub> emissions from 1990 to 2012 using a record of CH<sub>4</sub> and CO measurements from a dozen sites across the Bay Area and background CO estimates. The following sections describe the data used to estimate the ratio of local enhancements—the change in concentration from the addition of local emissions—of CH<sub>4</sub> and CO, the quality of the CO emission inventory, the resulting estimates of CH<sub>4</sub> emissions, and a discussion of the limitations and implications of the study.

## 2. Methods

We follow a scaling ratio approach of CH<sub>4</sub>: CO applied previously (e.g., Wennberg et al. 2012, Hsu et al. 2010; Singh et al. 2010), applying data from multiple sites for both local and background air. For oceanic background CH<sub>4</sub> and CO, we compare data from multiple sites, and then subtract the background CH<sub>4</sub> and CO from measurements at the Bay Area sites to estimate local enhancements. The resulting estimates of local CH<sub>4</sub> and CO are analyzed in 3 yr periods, and the resulting CH<sub>4</sub> emissions are estimated using the CH<sub>4</sub>:CO ratios combined with the BAAQMD inventory estimates of CO emissions.

### 2.1. Bay Area CH<sub>4</sub> and CO measurement sites

The BAAQMD operated monitors that measured CH<sub>4</sub> hourly starting with 9 sites in 1980, peaking at 12 sites from 1986 to 1994, then tapering off to 7 sites in 2010 and none in 2013 (Fig. 1). Two instruments were used to measure CH<sub>4</sub>, a Bendix instrument (Model 8202A) that measured CH<sub>4</sub> and total hydrocarbons, and a Thermo Electron (TE-55), that measured CH<sub>4</sub> and non-methane hydrocarbons. Both instruments were calibrated to CH<sub>4</sub> using standard gas additions. While the focus of the measurements was on the non-methane hydrocarbons, the CH<sub>4</sub> measurements are reasonably consistent over time and among sites, and exhibit concentrations consistent with background for some sites. CO was measured with Thermo Electron (TE-48) CO monitors, calibrated following a US-EPA method (40 CFR 53), except for San Jose, where a trace-level CO analyzer (TE-48TC) was used. In this analysis, we estimate local mixing ratio enhancements for CH<sub>4</sub> and CO from 24-h values for days with at least 18 h of valid data. (The numbers of valid observations by year, site and pollutant are shown in Supplementary materials S5.)

### 2.2. Drift in CO instruments

The TE-48 CO instruments were subject to drift on the order of 200 ppb, typically on the timescale of weeks. Instruments were re-zeroed after this amount of drift based upon measurements of CO-free air made every two days. These measurements were available in digital form from 2009 through 2012. Subtracting these zero measurements from the CO measurement appeared to correct much of the drift. Because of the limited availability of zero-air measurements, this adjustment could only be made in the most recent, 2009–2012, period. Applying the drift correction had little effect on the estimated ratios of local CH<sub>4</sub>:CO. (See Supplementary materials S4 for details.)

### 2.3. Estimating seasonal background CH<sub>4</sub> and CO

Ambient atmospheric CH<sub>4</sub> concentrations are typically enhanced by small fractions above oceanic background inflow to the Bay Area. In contrast, ambient CO is more strongly enhanced by local emissions. In both cases, background CH<sub>4</sub> and CO

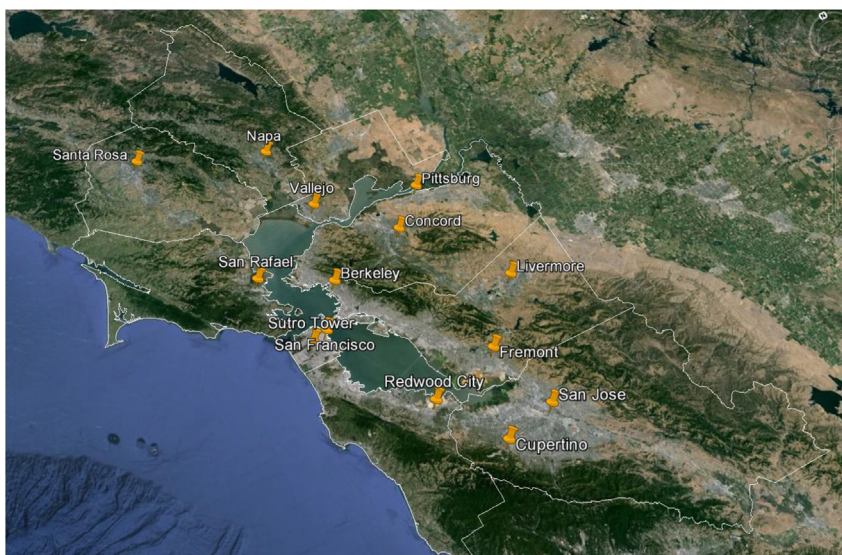


Fig. 1. Locations of CH<sub>4</sub> and CO sampling sites in the Bay Area used for this study.

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