Diurnal and Seasonal Attribution of Anthropogenic CO₂ Emissions Over Two Years in the Los Angeles Megacity
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I. Introduction

In order to understand the role of anthropogenic greenhouse gases (GHGs), of which CO₂ is the most abundant, in climate change we must understand their sources. Since cities produce >70% of the anthropogenic GHGs, large signals there can be used to study the details of emission patterns. Treaties are being signed to regulate emissions, and there must be verification of compliance. The easiest way to get a broad picture of the distribution of CO₂ and its changes through time is to collect measurements of total column concentrations from space. Satellite borne instruments measure during midday, but is the distribution of emissions among the sources the same at all times of day for all times of year? Understanding the diurnal variation of the sources is important for comparison with bottom-up inventories, modeling, and results from new instruments such as C-3 (Eldering et al., 2013) and the Geostationary Carbon Power Investigation (X et al., 2014) which will see more of a diurnal cycle.

We have shown that CO₂, δ¹³C, and δ¹⁸O can be used to distinguish among gasoline combustion, natural gas combustion, and biosphere contributions to CO₂ in the atmosphere (Newman et al., 2015) in a top-down approach. Here we present in situ data for different times of day from the megacity of Los Angeles, CA, specifically from the Caltech campus in Pasadena (Figure 1), in order to understand diurnal variation using this method with CO₂ as a proxy for δ¹³C, for morning, midday, and evening for June 2013 through May 2015.

II. Data and Methods

- The data sets involved in this study include continuous measurements of CO₂ and δ¹³C (Hiscox Isotopic CO₂ Analyser) and CO₂ (LIR NDIR CO₂ EP Monitor) in Pasadena and background measurements on Mt. Wilson (CO₂, CO levels). δ¹³C measurements are determined for flask samples collected on alternate afternoons at 14:00 PST in Pasadena.

- We use multiple mass balance calculations on monthly averages to distinguish among the sources: fossil fuel combustion (CO₂), including gasoline (CO₂ gasoline), and background (CO₂, biosphere) emissions.

- The δ¹³C values of the flask samples give the fraction of CO₂ from CO₂ in the total local enhancement over background CO₂ (Eswaran et al., 2003).

- We use the S-15 method from the flask δ¹⁸O measurements and CO₂/δ¹³C from the continuous measure results for 13:00-15:00 to determine the emission ratios, RCO₂/CO₂, for each month (Figure 4; e.g., Tansbult et al., 2008, 2011; Vogel et al., 2010; Miller et al., 2012). This assumes that RCO₂/CO₂ does not vary diurnally, but only seasonally.

- The stable isotopic composition of the carbon in the CO₂ (δ¹³C, % relative to the standard VPD8) is then used to distinguish gasoline and natural gas combustion within CO₂.

- The monthly averages of δ¹³C of CO₂ for morning, midday, and evening on a single day (right column). Keeling plot intercepts and correlation coefficients are shown for each correlation line. Vertical lines on the diurnal plots indicate the times chosen between morning and midday and evening.

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- We have shown that the pattern of higher fossil fuel emissions observed in Pasadena during summer is due to the annual wind direction pattern (Newman et al., 2015). However, the different patterns for different times of day during the summer cannot be due to the winds, since the wind directions are the same at all time of day.

III. Results

- The monthly averages of the Keeling plot intercepts (Figure 5) show significantly different patterns at different times of day, especially between early mornings and later.

- The lowest values of δ¹³C are centered on warmer months during midday and evening, whereas they are centered in the fall in the morning. This suggests that gasoline combustion or the biosphere dominate the signal at different times of day.

- When these δ¹³C values are compared with values of the fraction of CO₂ from fossil fuel CO₂ (Figure 6), this dichotomy propagates through to different patterns for the proportions of natural gas and gasoline combustion of total CO₂ at different times of day (Figure 6 left). Although natural gas is the dominant source of local CO₂ emissions during the summer middays, when the satellite borne instruments observe, gasoline combustion is the dominant source during summer mornings and evenings.

- Winter early morning emissions are mostly from the biosphere, very different from midday, when the biosphere contributes at most ~20%. As expected, the biosphere is a sink for CO₂ during the spring and summer, although this sink is much less intense than elsewhere.

- The absolute contributions in CO₂ ppm (Figure 6 right) are very similar to the patterns for the fractions of CO₂ emissions for the morning, and are higher for mornings and evenings for midday, when the atmosphere is most well mixed and therefore mixing ratios are closest to the background values, resulting in lower CO₂ ppm. The seasonal pattern for the evening absolute contributions is similar for all three sources.

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IV. Conclusions

- The relative proportions of the different emission sources for CO₂ may not be the same at different times of day. At least during the summer, this is not obviously due to diurnal changes in wind direction bringing emissions from different regions.

- In the case of Los Angeles, the relative contribution from natural gas combustion is higher during the summers and lower during the winter at midday and during the evenings, when orbiting satellites take measurements, but is higher in winter-spring than in summer during mornings. This is probably because of shifting wind directions seasonally.

- The biosphere’s contribution is higher during the cooler months than during the warmer months for all times of day, as expected for this low mid-latitude semi-arid region. However, the early mornings always have the highest biospheric contributions of the day.

- Emissions from gasoline combustion do not have a clear seasonal cycle for these two years, during midday and evening.

- To improve this analysis, we must determine the emission ratio (RCO₂/CO₂) for different times of day. We must also improve modeling at times other than midday or evening, in order to quantitatively account for transport of emissions.

References


