



Using atmospheric radiocarbon (¹⁴CO₂) to estimate fossil and biogenic CO₂ fluxes in the LA megacity

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Project Goals

- Ultimately: invert ¹⁴C or CO₂ fos for FF emissions via CO (and other tracers.)
- Proximately: compare observed CO₂ fos to transported inventories.

Outline

- Background on megacities and ¹⁴CO₂
- Results and Analysis
- Conclusions and Future work

Why Megacities? Large emissions and large signals!

Population (Millions)	GHG Emissions (M tCO ₂ e)
1. China: 1,192	1. USA: 7,107
2. India: 916	2. China: 4,058
3. 50 Largest Cities: 500	3. 50 Largest Cities: 2,606
Duren and Miller, Nat. C.C., 2012 • Existing megacities (2012) • Projected new megacities (2025)	

Megacity > 10 million; 2010 = 22; 2025 = 38

¹⁴CO₂:¹²CO₂ (Δ^{14} C) is a robust tracer for fossil fuel fluxes: atmospheric Δ^{14} C looks just like fossil CO₂.

 $\Delta^{14}C_{ff} = -1000 \text{ per mil (i.e. zero } {}^{14}C)$ Scaling: -2.7 per mil $\Delta^{14}C = 1 \text{ ppm CO}_2$ -fossil





Includes only fossil fuel

Miller et al, 2012

CO₂ PBL enhancements (or depletions) can be partitioned into Ecosystem and Fossil fractions.

$$CO_{2}xs$$

$$C_{obs} = C_{bg} + C_{fos} + C_{bio}$$

$$(\Delta C)_{obs} = (\Delta C)_{bg} + (\Delta C)_{fos} + minor$$

Measurement precision = 1.7 per mil \rightarrow ~ 1 ppm C_{fos}

$$\Delta^{14}C = \left[\frac{({}^{14}C/C)_{sam}}{({}^{14}C/C)_{std}} - 1\right] \times 1000$$

 $\Delta_{\rm ff}$ = -1000 per mil; $\Delta_{\rm atm}$ ~ +20 per mil

Context: NOAA/INSTAAR ¹⁴CO₂ sites sensitive to significant fraction of Asian and North American emissions







CO₂xs, fos and bio time series



USC Granada Hills CS Fullerton

$\Delta^{14}CO_2$ -based source analysis



-750 per mil \rightarrow 25% biological

CO₂fos/bio source analysis

- <CO2bio/CO2xs> = 0.27 (0.09/+0.18)
- Also highly correlated (R = 0.8)
 → common source location and transport



Why is CO₂ bio as high as it is?

- Ethanol in gasoline (10% of 50% of fuel sources)
- Human Respiration (~1% of fuel sources)
- What is the rest? (~ 20%)
- Urban Biosphere?





Transforming in situ CO to CO₂fos

Granada Hills in situ data



- Just an example, for now...
- Yellow represents midday hours – i.e. only when our CO/CO₂fos values are valid.
- Evidence for diurnal variability in CO:CO₂

Wintertime biospheric CO_2 fraction averages ~40% of total CO_2 enhancement.



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Conclusions and Future work

- 1. $CO_2xs \neq CO_2ff$, even in L.A.
- Significant CO₂bio seems to be the rule not the exception.
- 3. Remote-sensing and in situ approaches need to account for biospheric CO₂.
- 4. $\delta^{13}CO_2$ measurements \rightarrow fuel type partitioning (oil v. gas)
- 5. Create continuos time series of CO_2 fos via CO_2 fos:CO ratios.

Mount Wilson Observations of 14C, CO2 and CO



MWO and LA Basin comparison



- Both show substantial Cbio, but MWO also shows substantial uptake.
- MWO signals are weaker.
- Differences between
 CO:CO2xs and CO:CO2ff are consistent with Cbio contributions, but...
- ... they are offset. Is this spatial or a time trend in combustion efficiency.

Future work

- Sampling:
 - continue at 3 or 4 days per week for ~ 2 years
 - Add a summer and winter diurnal cycle sampling campaign
 - Switch PFPs to other sites (Compton?, Irvine?, San Bernardino?)
- Measurement and QC:
 - Data flagging using in situ CO2, CO, ws and wd



CO₂ PBL enhancements (or depletions) can be partitioned into Ecosystem and Fossil fractions.



Fossil Fuel and $\Delta^{14}C$



•¹⁴C is produced from N in the stratosphere by cosmic rays.

- •It is oxidized to CO, then CO₂
- •¹⁴C has a half-life of ~6000 years.

•Thus, it is absent from fossil fuels, and thus an excellent tracer for these emissions.

- •In the USA, minor emissions from nuclear power plants
- •Also fluxes associated with gross C fluxes from oceans and terrestrial biosphere.

U.S. lower atmosphere data show expected depletions of ¹⁴C



INSTAAR+UCI Measurement Precision

- Measurement made by Accelerator Mass
 Spectrometry (AMS) at UC Irvine
- Precision = 1.6 per mil
- Accuracy = < 0.5 per mil (based on intercomparison of different labs)



 $en) = -31.46 \pm 1.63$