Using atmospheric $^{14}CO_2$ measurements to quantify fossil fuel emissions and evaluate atmospheric transport

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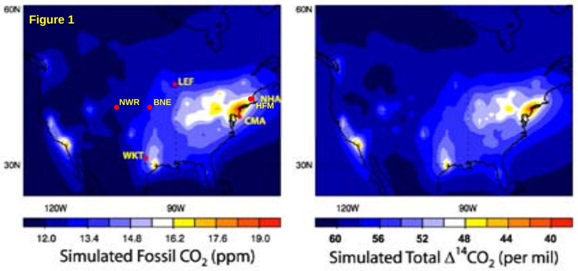
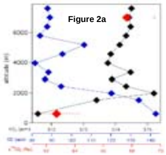
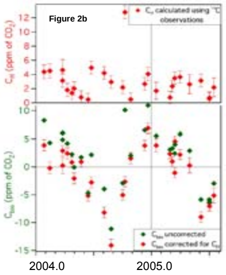
$^{14}CO_2$ as a tracer of fossil fuel emissions

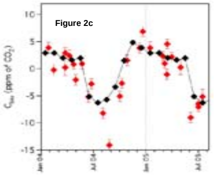


- TM5 model simulations show that total $\Delta^{14}CO_2$ is an excellent tracer for fossil fuel CO_2. The small differences result from the contributions of non-fossil fuel terms to the $^{14}CO_2$ budget.
- Four active vertical profile measurement sites (three heights only): Portsmouth, NH (NHA), Cape May, NJ (CMA), Park Falls, WI (LEF), Beaver Crossing, NE (BNE); one active surface site Niwot Ridge, CO (NWR); and one planned tower site, Moody, TX (WKT) are shown.

Separating Fossil and Biological Signals in Boundary Layer CO_2







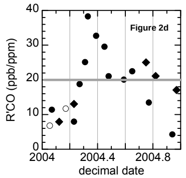
- Fig. 2a is an example from HFM showing both biological uptake and fossil fuel influence in the boundary layer. Here, fossil fuel CO_2 is masking the true extent of the biological uptake.
- In Fig. 2b we quantitatively separate the fossil fuel (C_{ff}) and biological (C_{bio}) influences on boundary layer CO_2.
- Fig. 2c shows that the calculated C_{bio} signal (red) agrees well with independent eddy-covariance measurements from Harvard Forest (black line).
- Fig. 2d shows that using C_{ff} to determine a CO:CO_2 emissions ratio results in a seasonally variable ratio (black symbols) that is significantly different from a bottom-up estimate from an EPA inventory (gray line). Note that this implies that if one were to use CO observations (Fig 2a) and the inventory, the derived values of C_{bio} would be incorrect.
- Equations 2a and 2b describe the atmospheric mass balances for boundary layer CO_2 and $\Delta^{14}C$ and how we determine C_{ff} and thus C_{bio}.

Eq. 2a

$$C_{obs} = C_{bg} + C_{ff} + C_{bio}$$
$$\Delta_{obs} C_{obs} = \Delta_{bg} C_{bg} + \Delta_{ff} C_{ff} + \Delta_{bio} C_{bio}$$

Eq. 2b

$$C_{ff} = \frac{C_{obs}(\Delta_{obs} - \Delta_{bg})}{(\Delta_{ff} - \Delta_{bg})} - \underbrace{\frac{C_{HR}(\Delta_{HR} - \Delta_{bg})}{(\Delta_{ff} - \Delta_{bg})}}_{\text{Biosphere disequilibrium Correction} \sim <0.2\text{ ppm}}$$



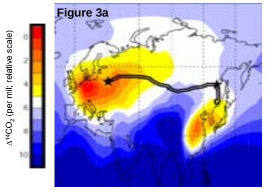
Introduction

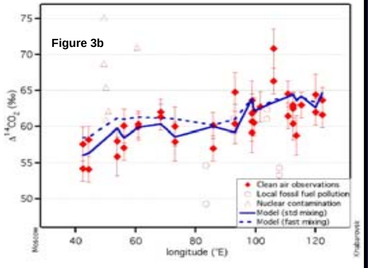
→ *Developing a reliable observational method for tracking fossil fuel emissions is a critical part of any carbon monitoring strategy,* because inventories need to be verified and, unlike observations, they are inevitably out of date.

- $^{14}CO_2$ is an ideal fossil fuel tracer because radioactive decay (half life = 5700 years) leaves all fossil fuels devoid of ^{14}C. In contrast, all other reservoirs exchanging carbon with the atmosphere are relatively rich in ^{14}C.
- Figure 1 demonstrates this by showing that simulated patterns of total $\Delta^{14}CO_2$ and the fossil component of CO_2 over North America are very similar.
- Knowing the fossil fuel contribution to atmospheric CO_2 is important both for verifying stated fossil fuel emissions and also for isolating the biological contributions to observed CO_2, an example of which is given in Figure 2.
- Fig. 3 gives an example of how $^{14}CO_2$ measurements can also be used together with inventories of fossil fuel emissions to test our knowledge of atmospheric transport.
- Fig. 4 describes our North American measurements.
- As we make more and more measurements, we will be able to test not just model transport accuracy but our atmospheric top-down source estimation techniques themselves (data assimilations and inversions) by attempting to directly estimate fossil fuel emissions from atmospheric $^{14}CO_2$ data.

Using $^{14}CO_2$ to evaluate Atmospheric Transport

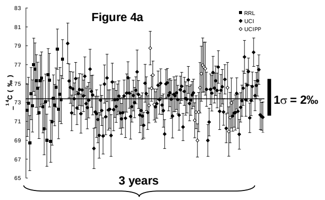
- Fossil fuel emission is the best quantified flux in the carbon cycle. Knowing the source and the atmospheric distribution of a tracer ($\Delta^{14}C$) for that source allows us to test our knowledge of atmospheric transport.
- As we make more and more measurements of $\Delta^{14}C$ over the United States and other continents, these will become powerful constraints for atmospheric transport models.
- Fig. 3a shows the simulated surface distribution of $\Delta^{14}C$ resulting from the European fossil fuel emissions. We predict a large east-west gradient across Siberia that is confirmed by observations (Fig. 3b) sampled during the TROICA-8 rail-based air sampling mission in April 2004.
- Fig. 3b shows the observations and model simulations. Two variants of TM5 ('fast' and 'standard' mixing) were compared with the observations. The 'fast' mixing version appears to slightly underestimate the east-west gradient. Although signal-to-noise is low in this case because the variants of TM5 were very similar, this illustrates the potential of using $\Delta^{14}C$ as a tracer of atmospheric transport.





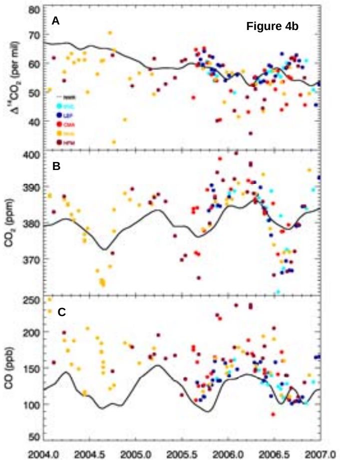
North American $^{14}CO_2$ measurements

Long-term Measurement Stability



- Over three years, the stability of the calibration of our measurements is very high: 2 per mil.
- Such stability is critical for constructing time series of atmospheric change.
- This 'long-term precision' is assessed by analyzing air from a single air tank every time we analyze our actual air samples.
- The stability calculation includes samples measured at two different labs, showing that our calibration is robust.

Boundary-layer time series of $\Delta^{14}C$, CO_2, and CO



- Sample are collected at five aircraft sites shown in Fig 1. (circles) and the NWR high altitude site (line), which serves as a reference of unpolluted air.
- All samples are collected in the planetary boundary layer (PBL) and warm colors represent more polluted east coast sites (NHA, HFM, CMA) and blues represent less polluted Midwestern sites (BNE, LEF)
- $\Delta^{14}C$ (panel A) tends to be much lower at the eastern sites, indicating a large influence of fossil fuel emissions, even during summer when CO_2 (panel B) shows net uptake by plants.
- CO (panel C) also contains information about fossil fuel emissions, but this is convolved with information from numerous other CO sources and sinks, like biomass burning, hydrocarbon oxidation and removal by OH (see Fig. 2d).
- $\Delta^{14}C$ data higher than the NWR reference line probably indicate air flow from the Gulf of Mexico where background $\Delta^{14}C$ is higher (see Fig. 1).
- These data will allow for important tests of both the transport and fossil fuel emissions currently used in the CarbonTracker system.

References and further reading

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