# REGIONAL PATTERNS OF FOSSIL FUEL $CO_2$ IN THE PLANETARY BOUNDARY LAYER ACROSS NORTH AMERICA

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## ABSTRACT

The rate at which fossil fuels are lofted off continents and mixed between the northern and southern hemispheres in atmospheric models critically determines the distribution and magnitude of surface sinks in top-down inversions that take advantage of atmospheric $CO_2$ observations. Tracers that can serve as potentially useful constraints on these mixing processes include water vapor, radon, and $\Delta^{14}C$. Past work using $\Delta^{14}C$ and radon suggests that fossil fuel emissions in Europe are more seasonal than that reported in monthly inventories [*Levin et al*., 2003]. $\Delta^{14}C$ is a particularly sensitive tracer of fossil fuel dispersion because fossil fuels contain no $^{14}C$. Here we used annual plants to map out patterns of atmospheric $\Delta^{14}C$ across North America. This information was then used to estimate fossil fuel-derived $CO_2$ concentrations in the planetary boundary layer across the continent, relative to a clean air region that we defined in the Rocky Mountains.

## METHODS

We collected corn (*Zea mays*) from 67 sites across North America during the summer of 2004. Our sampling protocol was designed to capture regional and continental scale patterns of fossil fuel $CO_2$ levels; we avoided areas directly influenced by point sources such as major roads or cities. Corn was chosen because it grows ubiquitously across North America and because of its C4 physiology. Leaf samples were converted to graphite using the process described by *Santos et al.* [2004] and analyzed for $\Delta^{14}C$ using the W.M. Keck Carbon Cycle Accelerator Mass Spectrometer at the University of California, Irvine. Because our objective was to measure variations in fossil fuels levels across the continent, we also estimated and removed the effects of biosphere-atmosphere, ocean, and stratospheric mixing on surface $\Delta^{14}C$ patterns using an atmospheric tracer transport model and prescribed surface and stratospheric fluxes [*Krakauer et al*. in prep.]. These effects were small, but non-negligible.

## RESULTS

In places where the plants were exposed to sustained and elevated levels of $CO_2$ from fossil fuel emissions, $\Delta^{14}C$ levels were reduced (Figure 1). We found that there was a drop of 7.5‰ ± 4.3‰ between the Mountain West (Idaho, Colorado, New Mexico and Alberta, Canada) and the Eastern U.S. (Massachusetts, New Hampshire, New York, Indiana, Ohio, Pennsylvania, Maryland, West Virginia, Kentucky, Tennessee, Virginia, Mississippi, Alabama, Georgia, Florida, and Halifax, Canada). This corresponds to a 2.9 ± 1.5 ppm increase in fossil fuel $CO_2$ levels in the east, relative to the west (Table 1). An even larger $\Delta^{14}C$ decrease was found between the Mountain West and the Ohio/Maryland region (Ohio, Pennsylvania, West Virginia, and Maryland), corresponding to 4.3 ± 1.0 ppm of added fossil $CO_2$.

Using emission factors from *Potosnak et al.* [1999] and the $\Delta^{14}C$-derived estimates of fossil fuel $CO_2$ described above, we estimate that CO levels were elevated by 73.1 ppb in the Eastern U.S. and by 108.3 ppb in the Ohio/Maryland region, as compared with background CO levels in the Mountain West.

## DISCUSSION AND CONCLUSIONS

The high $\Delta^{14}C$ values in the Rocky Mountains suggest that fossil fuel emissions from California and other western states are either substantially diluted by the time they reach this region or that these emissions leave the continent via another route (e.g., entrainment into a southerly flow around the Pacific high that forms during summer months). If the latter is correct, atmospheric sampling strategies designed to measure

North American continental outflow should sample southern as well as eastern borders. The variations in $\Delta^{14}C$ at a regional scale are substantial and indicate that ecosystem studies using $\Delta^{14}C$ as a tracer cannot assume that local atmospheric levels are the same as that recorded at remote marine boundary layer stations.

**FIGURES**

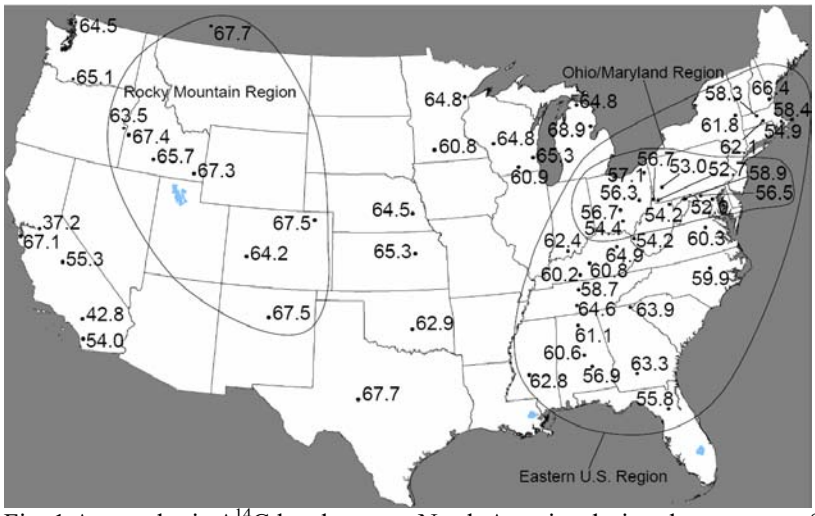


Fig. 1 Atmospheric $\Delta^{14}C$ levels across North America during the summer of 2004.

Table 1. Continental-scale $\Delta^{14}C$, fossil fuel $CO_2$ and fossil fuel CO during 2004.

| | Mountain West | Eastern U.S. | Ohio/Maryland |
|---|---|---|---|
| Mean $\Delta^{14}C$ | 66.3 ‰ ± 1.7 ‰ | 58.8 ‰ ± 3.9 ‰ | 55.2 ‰ ± 2.3‰ |
| Decrease in $\Delta^{14}C$ relative to Mountain West | - | -7.5 ‰ ± 4.3 ‰ | -11.1 ‰ ± 2.9 ‰ |
| Biosphere + ocean + stratosphere contribution | - | 0.7 ‰ ± 0.3 ‰ | 1.0 ‰ ± 0.5 ‰ |
| Estimated $\Delta^{14}C$ change due to added fossil fuel | - | -8.2 ‰ ± 4.3 ‰ | -12.1 ‰ ± 2.9 ‰ |
| Added fossil fuel $CO_2$ | - | 2.9 ppm ± 1.5 ppm | 4.3 ppm ± 1.0 ppm |
| Added fossil fuel CO | - | 73.1 ppb ± 38.9 ppb | 108.3 ppb ± 28.6 ppb |

**REFERENCES**


Levin, I., et al., (2003) A novel approach for independent budgeting of fossil fuel $CO_2$ over Europe by $^{14}CO_2$ observations *Geophysical Research Letters 30*(23), 2194, doi: 10.1029/2003GL018477.

Krakauer, N.Y., J.T. Randerson and F.W. Primeau. (2005) The air-sea gas exchange rate in light of ocean radiocarbon uptake. In prep. For submission to JGR Biogeosciences.

Santos, G.M., J.R. Southon, K.C. Druffel-Rodriguez, S. Griffin and M. Mazon (2004). Magnesium perchlorate as an alternative water trap in AMS graphite sample preparation: a report on sample preparation at the KCCAMS Facility at the University of California, Irvine, *Radiocarbon 46*(1), 165-173.

Potosnak, M.J. et al. (1999) Influence of biotic exchange in combustion sources on atmospheric CO2 concentrations in New England from observations at a forest flux tower. *J. of Geophysical Research 104*(D8), 9561-9569.