Atmos. Chem. Phys. Discuss., 11, C6010–C6013, 2011
www.atmos-chem-phys-discuss.net/11/C6010/2011/


Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Continental-scale enrichment of atmospheric $^{14}CO_2$ from the nuclear power industry: potential impact on the estimation of fossil fuel-derived $CO_2$" *by* H. D. Graven and N. Gruber

**H. D. Graven and N. Gruber**

heather.graven@env.ethz.ch

Received and published: 5 July 2011

Dear Jocelyn,

We appreciate your interest in our paper and your input. We would like to respond to the issue you raised on the influence of the chosen background site on $\Delta^{14}$C-based estimates of fossil-fuel derived $CO_2$.

In calculations of fossil-fuel derived $CO_2$ from $\Delta^{14}$C measurements assuming a quasi-Lagrangian framework, the choice of reference site is critical. Air measured at the



reference site should be representative of the composition of air before interaction with the catchment area of interest. Selecting the reference site also determines the spatial scale of the catchment area. If emissions over a catchment area of a single urban center are of interest, then selecting a reference site 50 km upwind, for example, would result in measured $\Delta^{14}$C gradients that are highly sensitive to emissions in the local urban area but less sensitive to emissions several hundred kilometers away. This approach is likely to be effective in reducing or eliminating the influence of the nuclear industry on measured $\Delta^{14}$C gradients at urban scales, particularly if the region of interest is far from nuclear sources. We noted that this approach would reduce the potential nuclear bias at urban scales in the Abstract (last sentence), and in the Discussion (end of 2nd paragraph).

While you suggest that local background sites and meteorological selection criteria should always be implemented in order to eliminate the potential nuclear bias, this requirement unnecessarily restricts the $\Delta^{14}$C-method to be applied to small-scale, quasi-Lagrangian studies as described above. Rather than trying to eliminate any influence of nuclear $^{14}$C emissions in observations of $\Delta^{14}$C gradients, our work aims toward improving our understanding of nuclear influences. Indeed, by dismissing regions that might be influenced by nuclear $^{14}$C emissions or by examining $\Delta^{14}$C gradients at local scales only, we would not be able to employ $\Delta^{14}$C to improve upon fossil fuel emissions estimates that are most relevant to society, i.e. nationally-aggregated emissions from industrialized countries.

By addressing this issue, we lessen the restriction on spatial scales and locations at which the $\Delta^{14}$C-method of estimating fossil fuel-derived $CO_2$ can be employed and reduce the likelihood that nuclear influences on $\Delta^{14}$C will be attributed to fossil fuel influences. Our goals in this paper were twofold: 1. to provide a spatially-resolved estimate of $^{14}$C emissions from the nuclear industry, including an assessment of uncertainty, and 2. to point out and attempt to quantify $\Delta^{14}$C gradients at continental scales caused by nuclear emissions.

The purview of our study is fundamentally different from the urban-scale, quasi-Lagrangian study described above and from the studies that evaluate or correct for $\Delta^{14}$C enhancement in local areas surrounding nuclear sites (e.g. Levin et al., *Health Phys.*, 1988; Levin et al., *GRL*, 2003; Dias et al., *J. Environ. Rad.*, 2008). Our study examines continental-scale, steady-state $\Delta^{14}$C gradients caused by nuclear and fossil fuel influences on $\Delta^{14}$C using an Eulerian framework. Previous studies using observations and models have shown that continental-scale, steady-state $\Delta^{14}$C gradients result from fossil fuel emissions (Randerson et al., *GBC*, 2002; Hsueh et al., *GRL*, 2007; Turnbull et al., *JGR*, 2009), also shown in our paper. Implementing observation sites along such continental-scale $\Delta^{14}$C gradients may allow continental-scale fossil fuel emissions to be estimated with regional inversion schemes such as CarbonTracker (Pacala et al., NRC report, 2010). By design, these inversion schemes exploit continental-scale gradients to estimate continental-scale fluxes. In order to make inversion-based estimates of fossil fuel fluxes, all other influences on $\Delta^{14}$C gradients must be known and corrected for.

Therefore, we are interested in $\Delta^{14}$C gradients that are produced when $^{14}$C emissions from nuclear sites are mixed into the larger atmosphere - could a high density of nuclear sources or even one large nuclear source create large-scale regions of high $\Delta^{14}$C, relative to areas on the same continent without nuclear sites or to the free troposphere? An analogy could be made to $SO_2$ emissions from coal-fired power plants causing acid rain deposition over a large-scale region that extends several hundred kilometers downwind of the power plants.

We maintain that comparisons to continental reference sites or to the free troposphere are appropriate in our assessment of continental-scale gradients. These comparisons have direct bearing on observational studies that also make comparisons to continental reference sites or to the free troposphere (Levin et al., *GRL*, 2003; Turnbull et al., *GRL*, 2006; Levin and Roedenbeck, *Naturwissenshaften*, 2008; Vogel et al., *Tellus*, 2010; Van der Laan et al., *Tellus*, 2010; Molnár et al., *J. Rad. Nucl. Chem.*, 2010), and on



observational and modeling studies that examine continental-scale gradients in $\Delta^{14}$C (Hsueh et al., *GRL*, 2007; Turnbull et al., *JGR*, 2009; Turnbull et al., *ACP*, 2009). In equation 1, the excess fossil fuel-derived $CO_2$ ($\delta C_{ff}$) compared to the reference site is the main determinant of the fossil fuel dilution in $\Delta^{14}$C ($\delta\Delta_{ff}$). The global mean values we used to specify $\Delta^{14}$C and $CO_2$ mixing ratio at the reference sites ($\Delta_R$ and $C_R$) may include errors of $\pm$5 per mil and $\pm$1.6 ppm but these potential errors in $\Delta^{14}$C and $CO_2$ mixing ratio at the reference sites would cause an error of less than $\pm$0.8 $\%$ of $\delta\Delta_{ff}$ (less than $\pm$0.05 per mil, on average; p. 14590, lines 14-17). While the value of $\delta\Delta_{ff}$ at a particular site may change if another reference site was chosen, continental-scale gradients in $\delta\Delta_{ff}$ would not change.

We hope that others will improve upon our work in the future by producing better estimates of nuclear $^{14}$C emissions, potentially through the implementation of $^{14}$C emissions monitoring at more nuclear sites, and by producing better estimates of the $\Delta^{14}$C gradients that result from nuclear $^{14}$C emissions, potentially through the use of higher resolution or more sophisticated transport models.

Thank you and best regards, Heather Graven and Nicolas Gruber

---

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14583, 2011.