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Interactive comment on “Continental-scale enrichment of atmospheric $^{14}\text{CO}_2$ from the nuclear power industry: potential impact on the estimation of fossil fuel-derived CO_2 ” by H. D. Graven and N. Gruber

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The Graven and Gruber paper presents a new and very much improved map of nuclear industry $^{14}\text{CO}_2$ emissions. This is a useful addition to our understanding of atmospheric $^{14}\text{CO}_2$, providing a reasonably realistic representation of the magnitude and spatial distribution of the nuclear $^{14}\text{CO}_2$ point sources. It represents a marked improvement over previous nuclear $^{14}\text{CO}_2$ emissions maps, which distributed nuclear $^{14}\text{CO}_2$ emissions evenly across the Northern Hemisphere land (e.g. Turnbull et al., 2009) or excluded them altogether (Randerson et al., 2002; Hsueh et al., 2007; Turnbull

et al., 2009b). Further refinements can and should be made to Graven and Gruber's nuclear $^{14}\text{CO}_2$ flux map (e.g. better estimates of emissions from individual reactors, temporal variability, other sources such as combustion of ^{14}C labeled waste, etc), but one suspects that this version will be in use for years to come until funding and data are available to make improvements.

However, the paper goes on to use an atmospheric transport model to examine how the nuclear $^{14}\text{CO}_2$ flux might bias the use of $^{14}\text{CO}_2$ measurements as a quantitative tracer for fossil fuel CO_2 emissions. Numerous previous studies (e.g. Suess, 1955; Meijer et al., 1995; Levin et al., 2003; Turnbull et al., 2006; Hsueh et al., 2007; Graven et al., 2009; Djuricin et al., 2010) have all demonstrated how $^{14}\text{CO}_2$ measurements can be used to constrain fossil fuel CO_2 emissions, taking advantage of the isotopic difference between ^{14}C -free fossil fuel CO_2 emissions and "natural" CO_2 sources with ^{14}C content close to that of the atmosphere. A number of studies using both observations and models have examined potential biases in the method, and shown that over the continents, there is a small bias due to biospheric CO_2 exchange (with ^{14}C content slightly higher than the atmosphere), and that there can be a small bias due to nuclear emissions in the vicinity of nuclear facilities, which can be corrected for (e.g. Levin et al., 2003; Turnbull et al., 2009). Graven and Gruber's study appears to contradict previous research in suggesting that nuclear industry $^{14}\text{CO}_2$ emissions can overwhelm the fossil fuel CO_2 signal in many regions. However, the presentation of the results can be somewhat misleading, since it is not apparent to the casual reader how much of the model analysis and interpretation is predicated on the choice of background.

In equation 1, the choice of $\Delta^{14}\text{CO}_2$ background is critical in the calculation of Cff – the calculated Cff value is the mole fraction of fossil fuel CO_2 above the background value. The equation implicitly assumes a quasi-Lagrangian system, where the background site is upwind of the region of interest, and the observation site is downwind. Since $^{14}\text{CO}_2$ measurements are difficult and expensive, alternatives to measuring the "ideal" upwind site have been identified. Several studies have assumed and/or demonstrated

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that high altitude sites are a reasonable proxy for the upwind site in many cases (e.g. Levin et al., 2003; Turnbull et al., 2009). Graven and Gruber show that they may not be a reasonable proxy for the upwind site when substantial nuclear ^{14}C emissions occur near the region of interest, since the nuclear emissions would then be treated as co-located with the fossil fuel CO_2 source. However, simply by selecting a more appropriate background site, the nuclear bias could be eliminated in many cases. For example, figure 2 shows a strong nuclear bias in Western Europe, but by selecting a background surface site in Northern France (instead of Jungfraujoch at altitude in Switzerland), one could potentially eliminate most of the nuclear bias in the Western European grid cells.

Equally importantly, the model results show averages over a long period of time, inherently averaging over all meteorological conditions. Judicious sampling in appropriate wind conditions can also reduce or eliminate the nuclear bias.

It should also be noted that the effect of the biosphere flux (its isotopic disequilibrium) cannot readily be disentangled from the fossil fuel flux in most cases, since the two fluxes are co-located, and the biosphere flux is dispersed across the continental regions. Hence ^{14}C -based Cff values have typically been corrected for an estimated biosphere bias. In contrast, the nuclear industry flux is comprised of point sources, which should allow the nuclear source flux to be separated from the fossil fuel flux simply by judicious choice of background site, and by making use of higher resolution atmospheric transport models that can better separate the influence of nuclear point sources from large area sources.

My motivation in writing this comment is that, as it stands, this paper may be misleadingly quoted years into the future as demonstrating the fallibility of the ^{14}C method for quantifying fossil fuel CO_2 emissions. Yet the true value of this paper is in showing that experimental design is vitally important, and that careful experimental design (such as choosing background sites and meteorological conditions carefully) and rigorous examination of the resultant data (to ensure that no significant nuclear bias exists) is

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needed to ensure that the ^{14}C method accurately quantifies fossil fuel CO₂.

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