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Long-term observations of ^{14}C-based atmospheric fossil fuel CO_2 ($FFCO_2$) and the CO/$FFCO_2$ ratio in the Heidelberg urban environment

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Urban environments are large emitters of fossil fuel CO_2 and of combustion-related pollutants, such as carbon monoxide (CO). While huge efforts are currently undertaken to agree on commitments to reduce fossil fuel CO_2 emissions, these have not really been successful yet, and the global atmospheric CO_2 abundance is still increasing. However, the implementation of emission controls on pollutants, e.g. from traffic, seem to have been more efficient, based on recent emission inventory data (e.g. *http://www.statistik.baden-wuerttemberg.de*). In order to verify this bottom-up information we present here ten years of quasi-continuous atmospheric observations of CO_2, fossil fuel CO_2 ($FFCO_2$) and CO in Heidelberg. The fossil fuel CO_2 component is determined from integrated radiocarbon ($^{14}CO_2$) measurements, exploiting the fact that fossil fuel CO_2 lacks ^{14}C (e.g. Levin et al., *Geophys. Res. Lett. 30*, 2003).

Our measurements show that the fossil fuel CO_2 level in the Heidelberg suburbs has not significantly changed (i.e. decreased) in the last decade. Observed inter-annual variations are rather due to inter-annual changes of atmospheric transport, as was already reported by Levin and Rödenbeck (*Naturwissenschaften 95*, 2008). However, we find a ca. 25% change in the CO/$FFCO_2$ ratio of the regional concentration offsets compared to background levels from about 14.5 ppb/ppm in 2002 to about 11 ppb/ppm in 2009. This observation is in very good agreement with the emission statistic for the closer catchment area of our measurement site. The Statistische Landesamt, Baden-Württemberg (*http://www.statistik.baden-wuerttemberg.de*) reported a change in the CO to $FFCO_2$ emission ratio for the city area of Heidelberg from 13.8 ppb/ppm in 2000 to 10.5 ppb/ppm in 2007. A more than 20% decrease of the CO/$FFCO_2$ emission ratio is also reported for the whole state of Baden-Württemberg (south-west Germany), but note that these ratios are generally smaller, between 11.5 and 9 ppb/ppm, due to a smaller contribution of traffic to the emissions. Our results clearly show that statistical emissions inventories, or at least the ratios of emissions and their changes, can be reliably verified on the local to regional scale by careful long term atmospheric observations.

A temporal change of the CO/$FFCO_2$ emission ratio has, however, also implications for the use of CO as surrogate tracer for high-resolution fossil fuel CO_2 estimates (e.g. Levin & Karstens, *Tellus 59B,* 2007): Without ongoing ^{14}C calibration of the CO/$FFCO_2$ ratio, decadal trends in CO-based fossil fuel CO_2 estimates can be largely biased by up to 20-30%.

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