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Novel in-situ observations of δ^{13}C in an urban environment

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Cities are reportedly responsible for approximately 75% of anthropogenic Greenhouse Gases (GHG) emissions while comprising only two percent of the land mass [1]. A thorough investigation of urban emissions is thus indispensible. Previous studies found that the emission characteristics of rural and urban areas significantly differ as different emission sectors are involved [2]. Big cities or urban agglomerations, in our case the Greater Toronto Area (GTA), Canada, are known to utilize more natural gas for residential heating [3]. This study investigates if in-situ $^{13}CO_2$ measurements can be a tool to independently confirm this altered energy consumption behavior. We derive bottom-up estimates of the δ^{13}C signature of fossil fuel CO_2 emissions for the province of Ontario and a data set for the GTA using available CO_2 emission data [3, 4]. As one can assume different isotopic compositions of the fossil fuel CO_2 emissions from liquid, solid and gaseous fuels, we are left with a range of source signatures of -29‰ to -31‰ for Ontario, -30.5‰ to -33.5‰ for the GTA and -33‰ to -37‰ for the GTA during night-time, when accounting for the daily variations of residential heating and transportation. We further constrain the source signature for liquid and solid fuels using previous measurements of δ^{13}C in elemental carbon (EC) which shows that this fraction has a mean isotopic signature of -26.36$\pm$0.12‰ in winter, in our domain [5]. The main uncertainty for the bottom-up estimates is due to the δ^{13}C of the natural gas, which is assumed to range from -40‰ to -48‰. This translates to an uncertainty of $\sim$ 2‰ in the bottom-up estimate. From our hourly atmospheric $^{13}CO_2$ observations we derive an annual mean for δ^{13}C of -9.6‰ and a standard deviation (1-σ) of 1.1‰ for 2011-2012. The amplitude of the seasonal cycle is about 1‰ and the daily amplitude in the δ^{13}C depletion frequently surpasses 2‰. To exclude influences from biospheric CO_2 sources and sinks we restrict our analysis to winter data and only interpret the night-time increase (0-6 a.m. local time), which is dominated by local fluxes. Our observed night-time $\delta^{13}C_{source}$ data for January 2011 of -35.5$\pm$2.2‰ lies below the "bottom-up estimates" for the province of Ontario, but is in-line with the best estimate for the GTA of -36.8$\pm$2‰. Our result seems to suggest that in-situ δ^{13}C observations might hold the potential to identify air masses from regions where energy consumption behaviors are different, but this preliminary study also shows clear limitations of this approach: (I) The observations are not yet able to constrain the source signature better than by about 2‰. (II) The bottom-up estimates of $\delta^{13}C_{source}$, even after using an additional observational data set (δ^{13}C in EC), still rely on assumptions for the δ^{13}C of natural gas (i.e., the uncertainties of end-member). Additionally, this study emphasizes that the diurnal cycle of the different emission sectors has to be accounted for, when interpreting urban observations of δ^{13}C and CO_2.

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