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Source Attribution of Atmospheric CO₂ Using $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ as Tracers in Chinese Megacities

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Identifying the sources of atmospheric Carbon dioxide (CO₂) is an important prerequisite for developing effective mitigation strategies. Here we conducted regular observations of the atmospheric CO₂ mixing ratio and its carbon isotope compositions (i.e., $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$) in Xi'an and Beijing during winter, to estimate source contributions of CO₂ emissions in Chinese megacities. The results showed that CO₂ emissions in both Xi'an and Beijing originated mainly from fossil-fuel sources, which contributed $65 \pm 3\%$ and $82 \pm 2\%$ of the total CO₂ enhancement, respectively, during the sampling period; the results also revealed a substantial biogenic CO₂ contribution during winter. We further separated the fossil-fuel sources into contributions from coal, oil and natural gas combustions. We found that coal combustion was the dominant anthropogenic source in Xi'an, accounting for $54 \pm 4\%$ of the total fossil-fuel emissions, and oil and natural gas contribute almost equally to the emissions. In contrast, emission from natural-gas combustion was the main fossil-fuel source in Beijing, accounting for more than half of the total fossil-fuel emissions, whereas, coal combustion contributed only $17 \pm 10\%$. These top-down results are generally consistent with emission inventory when seasonal variations of emissions are considered; some differences between the two methods indicated that the inventory for Xi'an might be underestimating the emissions from oil consumption. This study confirms the potential of direct verification between top-down and bottom-up methods from the perspective of source attribution. We further combined inventory data sets and $\Delta^{14}\text{C}$ measurements to quantitatively evaluate the contribution of human respiratory emission in Beijing, and further isolate the emissions from fossil fuels and biogenic CO₂ sources. We found that the human respiratory emissions could increase atmospheric CO₂ concentration by about 2 ppm, accounting for $14\% \pm 6\%$ of average CO₂bio concentration in winter. This study highlights the importance of human respiration in carbon emissions in megacities and has implications for a better understanding of the regional carbon budget.

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