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Source Attribution of Atmospheric CO2 Using Δ14C and δ13C 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., Δ14C and δ13C) 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 ± 3% and 82 ± 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 ± 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 ± 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 Δ14C 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% ± 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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