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Impact of atmospheric radiocarbon and stable isotope measurements on understanding the global CH4 budget over 1850–2015

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Measurements of stable isotope ratios of atmospheric CH_4 ($\delta^{13}C$-CH_4, δD-CH_4) have been utilized to evaluate contributions of individual CH_4 sources and sinks to global atmospheric CH_4 budget. However, given the uncertainty of both the source isotope signatures and kinetic isotope effects, recent estimates of the global atmospheric CH_4 budget using stable isotope observations are still inconclusive. Radiocarbon measurements ($\Delta^{14}C$-CH_4) could provide stronger additional constraint on the fossil-fuel CH_4 sources (i.e., ^{14}C-free), but the uncertainty of $^{14}CH_4$ emissions from nuclear power facilities and a lack of data have limited such utilization. Here we describe a new approach to estimate plausible global CH_4 emissions and sinks scenarios over 1850–2015 using observations and one-box model simulations of atmospheric CH_4, $\delta^{13}C$-CH_4, δD-CH_4, and $\Delta^{14}C$-CH_4. As inputs to the model, we prepare a priori bottom-up CH_4 emission inventories, total atmospheric CH_4 lifetime, source and sink isotope signatures, nuclear power facility database, and atmospheric $\delta^{13}C$-CO_2 and $\Delta^{14}C$-CO_2 observations and their uncertainties. We then run a Monte Carlo simulation of atmospheric CH_4, $\delta^{13}C$-CH_4, δD-CH_4, and $\Delta^{14}C$-CH_4 over the period using the inputs with the uncertainties. By using the observational CH_4 and three isotope constraints, we derive the best combinations of biogenic, anthropogenic fossil-fuel, natural geologic, biomass-burning, and nuclear power facility emissions and total CH_4 lifetime. We find that reconciling CH_4, $\delta^{13}C$-CH_4, δD-CH_4, and $\Delta^{14}C$-CH_4 observations indicates that (1) natural geologic emissions are likely smaller than the recent bottom-up estimate 43–50 Tg CH_4 yr^{-1} reported by Etiope et al. (2019), (2) biomass burning and anthropogenic fossil emissions are larger than current bottom-up estimates, and (3) biogenic emissions are somewhat smaller than current bottom-up estimates. Our finding suggests multiple isotope measurements, including $\Delta^{14}C$-CH_4, have a strong potential to evaluate the current and future bottom-up global CH_4 emission inventories.