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Constraining N_2O emissions since 1940 by firn air isotope measurements in both hemispheres

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N_2O is currently the 3^{rd} most important anthropogenic greenhouse gas in terms of radiative forcing and its atmospheric mole fraction is rising steadily. To quantify the growth rate and its causes, we performed a multi-site reconstruction of the atmospheric N_2O mole fraction and isotopic composition using firn air data collected from Greenland and Antarctica in combination with a firn diffusion and densification model. The multi-site reconstruction showed that while the global mean N_2O mole fraction increased from (290±1) nmol mol^{-1} in 1940 to (322±1) nmol mol^{-1} in 2008 the isotopic δ values of atmospheric N_2O decreased by (- 2.2±0.2) ‰ for $\delta^{15}N^{av}$, (- 1.0±0.3) ‰ for $\delta^{18}O$, (- 1.3±0.6) ‰ for $\delta^{15}N^{\alpha}$, and (- 2.8±0.6) ‰ for $\delta^{15}N^{\beta}$over the same period. The detailed temporal evolution of the mole fraction and isotopic composition derived from the firn air model was then used in a two-box atmospheric model (comprising a stratospheric and a tropospheric box) to infer changes in the isotopic source signature over time. The precise value of the source strength depends on the choice of the N_2O lifetime, which we choose to be 123 a. Adopting this lifetime results in total average source isotopic signatures of (- 7.6±0.8) ‰ (vs. Air-N_2) for $\delta^{15}N^{av}$, (32.2±0.2) ‰ (vs. VSMOW) for $\delta^{18}O$, (- 3.0±1.9) ‰ (vs. Air-N_2) for $\delta^{15}N^{\alpha}$, and (- 11.7±2.3) ‰ (vs. Air-N_2) for $\delta^{15}N^{\beta}$ over the investigated period. $\delta^{15}N^{av}$and $\delta^{15}N^{\beta}$ show some temporal variability while the other source isotopic signatures remain unchanged. The ^{15}N site-preference (= $\delta^{15}N^{\alpha}$ – $\delta^{15}N^{\beta}$) can be used to reveal further information on the source emission origins. Based on the changes in the isotopes we conclude that the main contribution to N_2O changes in the atmosphere since 1940 is from soils, with agricultural soils being the principal anthropogenic component, which is in line with previous studies.