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Isotopologue signatures of N2O from groundwater, tile drainage water, riparian wetlands and waste water treatment wetlands

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Reactive nitrogen from anthropogenic activity released to groundwater, wetlands and open water bodies can be transformed and/or removed via nitrification and denitrification. The greenhouse gas N_2O is produced during these processes, but N_2O fluxes to the atmosphere are lowered by N_2O reduction to N_2, the last step of the denitrification pathway. The magnitude of N_2O fluxes and the removal of reactive N via denitrification as well as the related process dynamics are poorly known. Isotopologue signatures of N_2O such as $\delta^{18}O$, average $\delta^{15}N$ ($\delta^{15}N^{bulk}$) and ^{15}N site preference (SP = difference in $\delta^{15}N$ between the central and peripheral N positions of the asymmetric N_2O molecule) have be used to differentiate between nitrification and denitrification as source processes and they might be suitable to estimate N_2O reduction to N_2.

In denitrifying aquifers, it had been found that only a small fraction of denitrified N was released as N_2O (Weymann et al., 2008; 2009). Isotopic signatures of dissolved N_2O were extremely high with values of SP and $\delta^{18}O$ up to $>$ 80 ‰ and with close correlations between these signatures (Well et al., 2005). We hypothesized that this resulted from the almost complete reduction of N_2O to N_2 which is probably due to the slow diffusion within the water phase as compared to non-saturated systems like surface soils.

The aim of our study was to investigate isotopologue signatures of N_2O in a variety of water-saturated environments. In some of the sites, intense denitrification has been found in previous studies (denitrifying sandy aquifers in Northwest Germany, waste water treatment wetlands in Estonia). Here, we wanted to check if the isotope pattern is similar to the previous groundwater data and can thus be used as a general indicator for denitrification as the main source of N_2O. In further sites (riparian wetland in Estonia, tile-drain outfall from artificial drained arable soil in Northeast Germany), no independent information on denitrification was available and isotopic fingerprints were used as qualitative indicator for the dominance of nitrification or denitrification.

In the groundwater and wetland samples, isotopic signatures exhibited a relatively large variability of $\delta^{18}O$ and SP in most of the data-sets with typical width of ranges between 10 and 30 ‰ and a close correlation between these signatures. This shows that denitrification with N_2 as dominating product was evident in most cases. In the drained site, a single tile outfall was sampled monthly during one winter season. $\delta^{15}N^{bulk}$ and $\delta^{18}O$ were somewhat variable (-20 to -30 ‰ and 45 to 51 ‰ respectively) and were positively correlated. SP was relatively high (33 to 43 ‰) and exhibited a negative correlation with $\delta^{18}O$ and $\delta^{15}N^{bulk}$. The relatively small variations of all signatures suggest that process conditions were comparably constant. However the overall isotope pattern is neither typical for nitrification nor for denitrification and thus needs further clarification.

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