Nitrate Isotopic Signature of Rainwater in Singapore: Implications for nitrogen sources and transformation in tropical regions.

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Abstract:

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Globally increased emissions of nitrogen oxides (NOx) in the atmosphere pose threats to global ecosystems and human health. The main sink of nitrogen oxides from the atmosphere to terrestrial systems is the wet deposition. Therefore, tropical regions with abundant precipitation are likely to experience greater impact from nitrogen pollution. To identify the nitrate sources in tropical rainwater, we collected daily rainwater samples in Singapore between May 2015 and July 2017 and measured coupled nitrogen and oxygen isotopes of nitrate in these samples. The results showed that the NO$_3^-$-N concentrations range from 0.05 to 3.28 mg/L, with an average of 0.64±0.54 mg/L (±1SD), showing a higher level in the inter-monsoon (IM, April to May and October to November) period relative to the Northeast (NE) monsoon (December to March) and Southwest (SW) monsoon (June to September). The δ$_{15}$N-NO$_3^-$ values range from -6.5‰ to +8.2‰, with an average of -0.6±2.6‰, and the δ$_{18}$O-NO$_3^-$ values range from +38.9‰ to +71.7‰, with an average of +58.0±6.4‰. Similar to the seasonal pattern of nitrate concentration, higher isotopic values of nitrate are also observed during IM period, while the NE monsoon had more negative δ$_{15}$N-NO$_3^-$ values. In combination with back trajectory analyses, the wide range of δ$_{15}$N-NO$_3^-$ values suggests multiple nitrate sources, including anthropogenic activities (power plants, industry, transportation and biomass burning) and natural sources (lightning and biogenic soil NOx). The seasonal pattern of δ$_{15}$N-NO$_3^-$ values implies proportional contributions from different nitrate sources. We believe that seasonal variability of δ$_{18}$O-NO$_3^-$ values is driven by the formation pathways, with lower δ$_{18}$O-NO$_3^-$ values resulting from the hydroxyl radicals and peroxy radicals, which compete with ozone.