カナダ・チャーチルにおける炭素・水素同位体比の観測から推定された大気中 CH_4 濃度変動に対する北方湿地の寄与

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Contributions of regional boreal wetlands to atmospheric CH_4 variations at Churchill (Canada) estimated from carbon and hydrogen isotope measurements

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We conducted flask-based measurements of concentration, $\delta^{13}C$, δD of atmospheric CH_4 at Churchill, Canada (CHL; 58°44'N, 93°50'W) and Ny-Ålesund, Svalbard (NAL; 78°55'N, 11°56'E) during 2007–2014; CHL locates on the northern perimeter of the Hudson Bay Lowland (HBL); NAL is a background station remote from regional CH_4 sources. The CH_4 concentration at CHL is generally higher than that at NAL, while $\delta^{13}C$ and δD at CHL are lower than those at NAL, likely reflecting CH_4 emissions from regional to local boreal wetlands in nearby area of CHL. Clear seasonal cycles are observed in CH_4 and $\delta^{13}C$ with the respective seasonal maximum (minimum) values in January–February (June) and May (October). δD also shows a clear seasonal cycle, but it is not the case for CH_4 and $\delta^{13}C$, which exhibit large weekly-monthly variability. The summertime minimum of CH_4 concentration and maxima of $\delta^{13}C$ and δD at CHL are about 1 month earlier than those at NAL. A simple 1-box model indicates that contribution of biogenic CH_4 emissions peaks earlier at CHL than at NAL, causing the phase differences between the two sites. At CHL, short-term CH_4 variations are observed through the year but most pronounced in summer. By inspecting the relationships between CH_4 concentration and the isotope ratios, we estimated the source isotope signatures to be −63.4±2.8‰ for $\delta^{13}C$ and −316±24‰ for δD in summer (May–October), and −47.7±4.5‰ for $\delta^{13}C$ and −244±52‰ for δD in winter (November–April). These values indicate predominant contribution of wetlands emissions to CH_4 in summer and that of fossil fuel sources in winter. In addition, we use an atmospheric chemistry transport model at 1.12×1.12^{o} horizontal resolution to investigate the cause of seasonal and short-term CH_4 variations at the two sites. While the model reproduces the CH_4 concentration variations at NAL well, it overestimates summertime CH_4 level at CHL. Tagged tracer experiments imply that the highly elevated CH_4 concentrations come from emissions in boreal northern America, suggesting that our a-priori wetland flux in the region might be overestimated.