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Verification of methane emissions from atmospheric observations of CH_4, $^{13}CH_4$, and related tracers at the Lutjewad tower

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Abstract

In this study we evaluate surface emissions estimates of CH_4 based on observations from the Lutjewad atmospheric measurement station in the Netherlands (6.3529º E, 53.4037º, 1 m asl) for the years 2006 to 2014. An established approach to estimating fluxes from continuous observations is the "Radon-Tracer-Method" where ^{222}Radon soil emission flux estimates are used to scale concurrent fluxes of greenhouse gases within the catchment area of the observation site. However, the Radon-Tracer-Method suffers from considerable uncertainties from estimating the appropriate catchment area and the ^{222}Radion soil flux in the catchment area. On a regional to continental scale inverse modelling studies using observational input are a more sophisticated tool to evaluate emission budgets of CH_4 and other greenhouse gases. To better understand and quantify the CH_4 emissions in the station catchment area we evaluated the agreement between observed and model-based hourly mean time series of CH_4 and ^{222}Radon as well as $\Delta CH_4/\Delta^{222}$Radon ratios. Model results are based on simulations from the TM3-STILT model system (**S**tochastic **T**ime-**I**nverted **L**agrangian **T**ransport model coupled with the TM3 chemistry-Transport model to determine the boundary conditions outside Europe) with "a-priori" CH_4 emissions from the EDGARv4.2 emission database and a new Radon flux map developed within InGOS. Overall, we find that the ^{222}Radon is simulated quite well by the TM3-STILT model system within a range of typically ±10% whereas the CH_4 can be underestimated up to a factor of two. These findings indicate that current methane emission estimates (as a whole) based on EDGARv4.2FT emission inventory in the catchment area of Lutjewad might be significantly underestimated. In addition, we use the isotopic compositions of ^{13}C and ^{2}H (Deuterium) in CH_4 from concurrent flask measurements taken at Lutjewad to partition CH_4 sources and find that the fraction of fossil related EDGARv4.2 emissions is slightly overestimated, but within the uncertainty. Furthermore, we study the tracer correlations of CH_4 with CO_2 and CO to point to the potential sources under various conditions.