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TITLE: Gridded maps of geological methane emissions and their isotopic signature

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ABSTRACT:

Abstract. Methane (CH<sub>4</sub>) is a powerful greenhouse gas, whose natural and anthropogenic emissions contribute ~20 % to global radiative forcing. Its atmospheric budget (sources and sinks), however, has large uncertainties. Inverse modelling, using atmospheric CH<sub>4</sub> trends, spatial gradients and isotopic source signatures, has recently improved the major source estimates and their spatial-temporal variation. Nevertheless, isotopic data lack CH<sub>4</sub> source representativeness for many sources, and their isotopic signatures are affected by incomplete knowledge of the spatial distribution of some sources, especially those related to fossil (radiocarbon-free) and microbial gas. This gap is particularly wide for geological CH<sub>4</sub> (geo-CH<sub>4</sub>) seepage, i.e. the natural degassing of hydrocarbons from the Earth's crust. While geological seepage is widely considered a major source of atmospheric CH<sub>4</sub>, it has been largely neglected in 3-D inverse CH<sub>4</sub> budget studies given the lack of detailed a priori gridded emission maps. Here, we report for the first time global gridded maps of geological CH<sub>4</sub> sources, including emission and isotopic data. The 1°×1° maps include the four main categories of natural geo-CH<sub>4</sub> emission: (a) onshore hydrocarbon macro-seeps, including mud volcanoes, (b) submarine (offshore) seeps, (c) diffuse microseepage and (d) geothermal manifestations. An inventory of point sources and area sources was developed for each category, defining areal distribution (activity), CH<sub>4</sub> fluxes (emission factors) and its stable C isotope composition (δ<sup>13</sup>C-CH<sub>4</sub>). These parameters were determined considering geological factors that control methane origin and seepage (e.g. petroleum fields, sedimentary basins, high heat flow regions, faults, seismicity). The global geo-source map reveals that the regions with the highest CH<sub>4</sub> emissions are all located in the Northern Hemisphere, in North America, in the Caspian region, in Europe and in the East Siberian Arctic Shelf. The globally gridded CH<sub>4</sub> emission estimate (37 Tg yr<sup>-1</sup> exclusively based on data and modelling specifically targeted for gridding, and 43–50 Tg yr<sup>-1</sup> when extrapolated to also account for onshore and submarine seeps with no location specific measurements available) is compatible with published ranges derived using top-down and bottom-up procedures. Improved activity and emission factor data allowed previously published mud volcanoes and microseepage emission estimates to be refined. The emission-weighted global mean δ<sup>13</sup>C-CH<sub>4</sub> source signature of all geo-CH<sub>4</sub> source categories is about -49 ‰. This value is significantly lower than those attributed so far in inverse studies to fossil fuel sources (-44 ‰) and geological seepage (-38 ‰). It is expected that using this updated, more <sup>13</sup>C-depleted, isotopic signature in atmospheric modelling will increase the top-down estimate of the geological CH<sub>4</sub> source. The geo-CH<sub>4</sub> emission grid maps can now be used to improve atmospheric CH<sub>4</sub> modelling, thereby improving the accuracy of the fossil fuel and microbial components. Grid csv (comma-separated values) files are available at <https://doi.org/10.25925/4j3f-he27>.

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