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COMMENTARY

Conflicting estimates of natural geologic methane emissions

B. F. Thornton[1,*], G. Etiope[2,3], S. Schwietzke[4], A. V. Milkov[5], R. W. Klusman[5], A. Judd[6], and D. Z. Oehler[7]

Global bottom-up and top-down estimates of natural, geologic methane (CH_4) emissions (average approximately 45 Tg yr^{-1}) have recently been questioned by near-zero (approximately 1.6 Tg yr^{-1}) estimates based on measurements of $^{14}CH_4$ trapped in ice cores, which imply that current fossil fuel industries' CH_4 emissions are underestimated by 25%–40%. As we show here, such a global near-zero geologic CH_4 emission estimate is incompatible with multiple independent, bottom-up emission estimates from individual natural geologic seepage areas, each of which is of the order of 0.1–3 Tg yr^{-1}. Further research is urgently needed to resolve the conundrum before rejecting either method or associated emission estimates in global CH_4 accounting.

Keywords: Methane, Geologic, Anthropogenic

1. Introduction

The role of natural versus anthropogenic sources of the fossil fraction of the atmospheric methane (CH_4) budget is the subject of an ongoing debate (Petrenko et al., 2017; Etiope and Schwietzke, 2019). Although not the dominant source of atmospheric CH_4, the fossil fraction is roughly a quarter of the global annual CH_4 emissions of approximately 576 Tg yr^{-1} (top-down accounting, Saunois et al., 2020). Fossil CH_4 is provided by CH_4 sources with carbon > approximately 50,000 years old, whose radiocarbon (^{14}C) content (as $^{14}CH_4$) is essentially zero. Importantly, fossil CH_4 emissions stem from both anthropogenic and natural sources. Anthropogenic fossil CH_4 emissions have been most recently estimated at 108–135 Tg yr^{-1} (range of bottom-up and top-down accounting, Saunois et al., 2020). They include principally emissions from the oil and gas industry and coal mine venting. The natural fraction of fossil CH_4 refers to geologic CH_4 emissions from oil and gas seeps (either onshore or offshore), mud volcanoes, diffuse degassing over petroleum basins (microseepage), geothermal-volcanic systems, and abiotic CH_4 from serpentinized ultramafic rock systems (Etiope et al., 2019). We refer to this natural fraction as "geo-CH_4."

Historically, there has been controversy in separating the anthropogenic fossil and geo-CH_4 fractions of fossil CH_4 sources due to isotopic composition overlaps and limited observations. However, both anthropogenic fossil and geo-CH_4 sources can be accounted for and scaled up via observations; such bottom-up accounting presently suggests that natural fossil CH_4 accounts for approximately 45 Tg yr^{-1}, while anthropogenic fossil CH_4 accounts for up to approximately 135 Tg yr^{-1} (**Figure 1**). Uncertainties have been determined from these source-level local measurements of geo-CH_4 combined statistically with global activity data to produce bottom-up extrapolations, yielding 27–63 Tg yr^{-1} (95% confidence interval [CI]). Top-down geo-CH_4 estimates, based on atmospheric CH_4, $^{13}CH_4$, and ethane measurements, yield 12–97 Tg yr^{-1} (95% CI), with mean values of 38–53 Tg yr^{-1} (Etiope and Schwietzke, 2019).

In a recent work, Hmiel et al. (2020)—hereinafter referred as Hmiel et al.—deepened this debate by providing a dramatically downsized estimate of global CH_4 emissions attributed to geo-CH_4. Based on a study of late preindustrial CH_4 retrieved from Greenlandic ice, and essentially a top-down model, they conclude that only 1.6 Tg yr^{-1} (95% CI, 0.1–5.4 Tg yr^{-1}) are attributable to geo-CH_4 sources. This would be a negligible part of global CH_4 emissions of approximately 576 Tg yr^{-1} (Saunois et al., 2020). As a consequence, the results of Hmiel et al. necessarily imply that present-day CH_4 emissions from the fossil fuel industries (the anthropogenic source of fossil CH_4 in the atmosphere) must be currently underestimated by 25%–40% (**Figure 1**).

[1] Department of Geological Sciences and Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden
[2] Istituto Nazionale di Geofisica e Vulcanologia, Rome, Italy
[3] Faculty of Environmental Science and Engineering, Babes-Bolyai University, Cluj-Napoca, Romania
[4] Environmental Defense Fund, Berlin, Germany
[5] Colorado School of Mines, Golden, CO, USA
[6] Alan Judd Partnership, Northumberland, United Kingdom
[7] Planetary Science Institute, Tucson, AZ, USA

* Corresponding author:
Email: brett.thornton@geo.su.se

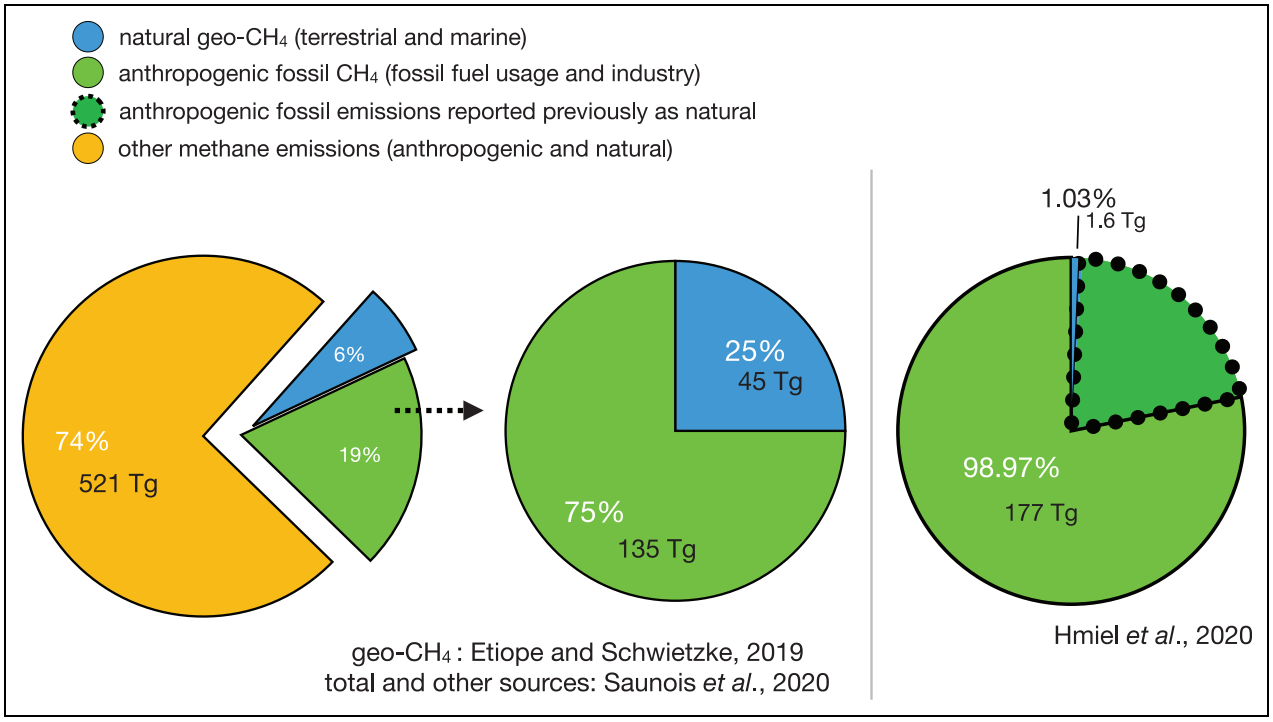

Figure 1. Global natural versus anthropogenic fossil CH_4 emission estimates. Saunois et al. (2020) reported annual bottom-up fossil fuel emissions of approximately 180 Tg, 25% of which were natural emissions (Etiope and Schwietzke, 2019), out of a global annual total of 747 Tg. In contrast, Hmiel et al. (2020) suggested median annual geo-CH_4 emissions of only 1.6 Tg. DOI: https://doi.org/10.1525/elementa.2021.00031.f1

The conclusions of Hmiel et al. are based on late preindustrial-era ice-core radiocarbon $^{14}CH_4$ measurements, and the assumption that natural geo-CH_4 emissions have not substantially changed since preindustrial times. The $\Delta^{14}CH_4$ measured in the ice-core CH_4 was close to that expected for the global biogenic methane source in the late preindustrial era, leaving almost no space for a significant global contribution of geo-CH_4 to the atmosphere at the time the gas was trapped in the ice. We note, however, that for the main conclusions and present-day natural global geo-CH_4 emission estimate, Hmiel et al. relied on only two ice-core samples from Summit in Greenland, dated approximately 1755 and approximately 1880 AD (although 1880 is within the Industrial Era), as other samples in the study are well within the industrial period of hydrocarbon extraction and production. Details of the complex and sophisticated methods of sample collection, gas extraction, chemical analysis, data treatment and correction, and modeling utilized by Hmiel et al. are not discussed in this article.

The main objective of this article is to contrast the results based on the ice-core $^{14}CH_4$ measurements with a wide body of data from many authors, which support the conclusion that natural geo-CH_4 emissions are far from negligible in the global CH_4 budget, contrary to the conclusion of Hmiel et al. We present the case that near-zero levels of natural geo-CH_4 emissions are incompatible not only with global both bottom-up and top-down geo-CH_4 estimates (**Figure 2a**) but also with direct measurements of local scale emissions from individual seepage zones (**Figure 2b**). We contend that numerous studies determining far higher natural geo-CH_4 emissions have not been adequately disproven by the limited measurements of Hmiel et al.

This article cannot resolve this scientific conundrum *per se*, but we argue that the Hmiel et al. results do not overturn the existing overwhelming contrasting evidence. This evidence is not highlighted in Hmiel et al. and has created a bias in the current scientific discussion. We provide arguments that (i) cast doubt on the validity of any near-zero geo-CH_4 estimate; (ii) show evidence that the existing literature on geo-CH_4 emissions are not erroneous by an order of magnitude—or more—as suggested in Hmiel et al.; and (iii) suggest that the level of confidence expressed by Hmiel et al. for their results is premature or misplaced. Thus, we illustrate the need for the reconciliation of these various lines of evidence to confidently quantify present-day CH_4 emissions, natural and anthropogenic.

2. Comparisons with bottom-up and local estimates

The magnitude of global geo-CH_4 emission values estimated by Hmiel et al. is hardly reconcilable with emission factors experimentally determined from more than 3,000 onshore gas–oil seeps, >740 mud volcanoes, >40 regions with active seabed seeps, diffuse degassing over petroleum basins, and exhalations from >2,300 geothermal-volcanic systems (Etiope et al., 2019). A comparison with global emissions from all these geologic sources was discussed in Etiope and Schwietzke (2019), and it is summarized in **Figure 2a**. These geo-CH_4 emission estimates do

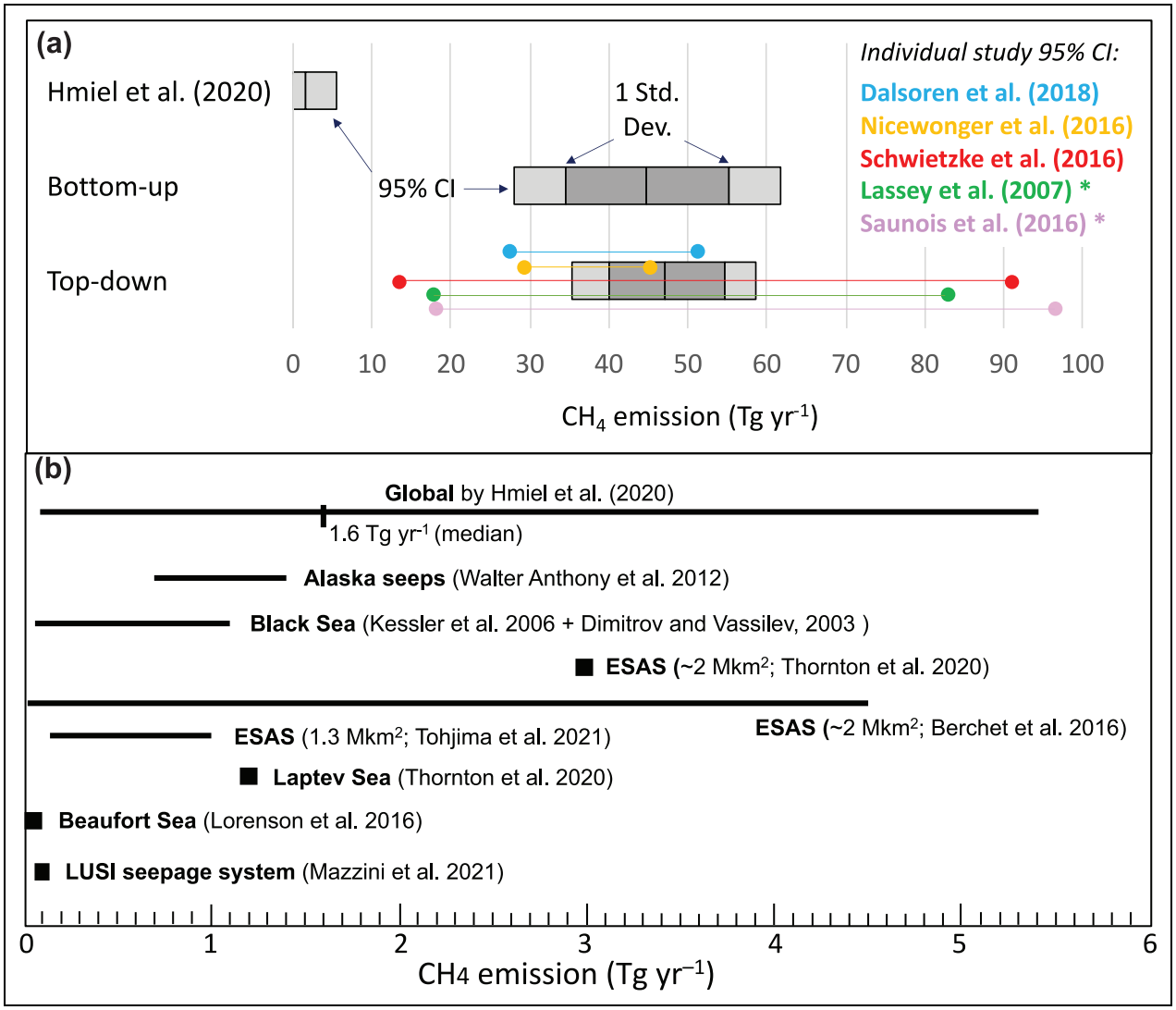

Figure 2. Geo-CH_4 emission estimates based on late preindustrial-era ice-core 14 measurements (Hmiel et al., 2020) and other works. (a) Comparison with global scale emission estimates. The light grey bars indicate the 95% confidence interval, and the dark grey bars represent one standard deviation when available. Bar central values indicate mean or median values according to each study. For top-down studies, the colored dots and lines indicate the 95% confidence intervals of each individual study within the top-down category, and the bars represent the joint probability distributions. Adapted from Etiope and Schwietzke (2019). See Etiope and Schwietzke (2019) for details on estimating geo-CH_4 emissions based on Saunois et al. (2016) and Lassey et al. (2007) data. (b) Comparison with local or regional scale emission estimates. East Siberian Arctic Shelf (ESAS) estimates derive from various measurement techniques (see text for additional discussion). A further, much larger, estimate of CH_4 emission from ESAS suggested 17 Tg yr^{-1} (as best estimate for the entire ESAS, no error bars) and 9 (2.87–18.46) Tg yr^{-1} from bubbling hot spots alone (Shakhova et al., 2014). DOI: https://doi.org/10.1525/elementa.2021.00031.f2

not include the fluxes of ^{14}C-free CH_4 from serpentinized ultramafic rock systems, discovered in at least 17 countries but whose global emission into the atmosphere is still unquantified (Etiope and Whiticar, 2019).

The bottom-up geo-CH_4 emission estimates essentially follow the upscaling methods recommended by the EMEP/CORINAIR Guidelines (EEA, 2004), which are typically applied in agricultural or natural ecosystems. The uncertainties in this type of extrapolation depend then on the uncertainty, statistically derived, of the emission factors and the activity. These uncertainties are not always quantified in the former works on geologic CH_4 emissions published in the 1990s and 2000s. For example, the first global estimates proposed for marine seeps, with a "best guess" of 10–30 Tg yr^{-1}, were reported in Kvenvolden et al. (2001) following two different process-based models, but without reporting the uncertainties. This marine seepage estimate was recently reassessed with uncertainties (5–12 Tg yr^{-1}) in Etiope et al. (2019) based on actual local–regional measurements. In the "Gaia's breath" paper by Kvenvolden and Rogers (2005), uncertainties of several estimates are qualitatively discussed, but not quantified.

It is interesting to note, however, that all global estimates starting from Etiope and Klusman (2002), to Judd

(2004), Kvenvolden and Rogers (2005), and Etiope et al. (2008), although following different combinations of emission factors and literature data, converged to a total emission of the same order of magnitude (at least one order higher than the one proposed by Hmiel et al.). The uncertainties were more carefully quantified in the latest bottom-up estimates as summarized in Etiope and Schwietzke (2019) and discussed for the various, specific geologic sources in Etiope et al. (2019). These are recalled in **Figure 2**.

A specific problem in the bottom-up upscaling of geologic CH_4 sources lies in the spatial and temporal heterogeneity of the fluxes, that is, in the representativeness of the "activity," the area of seepage or number of point sources for which a given emission factor should be applied. For example, microseepage, the diffuse CH_4 exhalation in areas over petroleum-bearing rocks, seems to be highly heterogeneous depending on the soil conditions (humidity and methanotrophic activity) and atmospheric parameters (mostly barometric pressure, which can influence the advective flux of gas from soil to atmosphere). To reduce this problem, a large number, statistically significant, of microseepage flux data are necessary. Several research groups are working in this direction (e.g., Zhao et al., 2021).

2.1. Contrast with regional emission estimates: The case of the East Siberian Arctic Shelf (ESAS) emissions

A contrast exists also between the Hmiel et al. natural geo-CH_4 estimate and bottom-up estimates of single geo-CH_4 seepage regions, where individual natural local emissions appear to be within the global range reported in Hmiel et al. Specifically, the minimum and median global values of Hmiel et al., 0.1 and 1.6 Tg yr^{-1} are exceeded by regional estimates (based on direct flux measurements and local extrapolations) of only a few individual seepage zones (**Figure 2b**). The sum of maximum values for only three regional seepage zones, discussed below, exceeds the global upper limit of Hmiel et al. (5.4 Tg yr^{-1}).

A major example of this contrast is found with the recent estimates of CH_4 emissions from the ESAS (the Laptev, East Siberian, and part of the Chukchi seas; **Figure 2b**). ESAS emissions of approximately 3 Tg yr^{-1} CH_4 (Thornton et al., 2020) include ^{14}C-depleted Pleistocene microbial and older thermogenic gas (Cramer and Franke, 2005; Berchet et al., 2020; Steinbach et al., 2021). The gas seeps within the Laptev Sea have been suggested to stem from an active, subsurface petroleum system based on measurements of CH_4, ethane, and propane adsorbed on the surface sediments in the seep regions, along with both geochemical (isotopic compatibility with source rock maturity) and geophysical (seismic reflection) data (Cramer and Franke, 2005). It has been established that the gas seeps in the ESAS are not new, recently-formed phenomena, by the presence of triterpenoid biomarkers (Grinko et al., 2020) and substantial CH_4-derived authigenic carbonates (MDAC; Ruban et al., 2020), which also supports a thermogenic CH_4 origin.

Multiple independent methods and studies led to the approximately 3 Tg CH_4 yr^{-1} emission estimate for the ESAS (Thornton et al., 2020 and references therein). However, a fairly wide range of ESAS annual CH_4 emissions have been reported in the literature, some are shown in **Figure 2b**. The underlying measurements behind these estimates (atmospheric and surface water CH_4) are extremely precise, such that errors in the measurements themselves do not contribute to any significant error in the integrated estimates for the entire ESAS region. It is differences in flux estimations, extrapolation methods, and differences in underlying observational data sets that have yielded the various ESAS emission estimates, each of which has a high precision. Utilizing Russian studies (e.g., Shakova et al., 2014) to achieve better spatial coverage of shallow portions of the ESAS not studied in Thornton et al. (2016; 2020) leads to an ESAS total CH_4 emission estimate of approximately 4.65 Tg yr^{-1}, a value also reported in Thornton et al. (2020). Shakhova et al. (2014) reported 17 Tg yr^{-1} as their best estimate (no error bars) for the ESAS and 9 (2.87–18.46) Tg yr^{-1} from bubbling hot spots alone. Although obviously far larger than other studies, these have not been directly refuted in the literature. Within the ESAS, emissions of 1.19 Tg yr^{-1} of dominantly thermogenic CH_4 have been estimated for the Laptev Sea alone (Thornton et al., 2020).

The possibility that ESAS CH_4 is not dominantly fossil (e.g., biogenic from recently deposited seabed sediments from terrestrial erosion processes) is very limited and applicable only to a minimal portion of the gas released from the seawater, and even much of this biogenic CH_4 appears derived from near-fossil carbon from old sources. For example, Sapart et al. (2017) measured ESAS sediment CH_4 close to shore in the Buor-Kahya Bay, showing large depletions in $^{13}CH_4$, and CH_3D, not classically "thermogenic," but with radiocarbon ages of 26–39 ka BP. The discrepancy between these "local" emissions from the ESAS and the global estimates of Hmiel et al. is significant and must call into question the Hmiel et al. conclusions.

It is conceivable that the ESAS emissions could have increased since the time of the preindustrial samples reported by Hmiel et al. However, we know of no published evidence (measurements or models) of such a temporal increase; this would be a worthy topic for future study. With present-day geo-CH_4 of 45 Tg yr^{-1}, this would require an increase in geo-CH_4 emissions since late preindustrial times by a factor of 28 (8.3–450) to satisfy Hmiel et al.'s preindustrial geo-CH_4 emission estimate of 1.6 (0.1–5.4) Tg yr^{-1}. The same argument would apply to other potentially increasing sources of geo-CH_4, such as thawing permafrost (Schuur et al., 2015). As described in more detail in Section 5, **Figure 3** includes a ramped geo-CH_4 scenario wherein geo-CH_4 increases fast enough to accommodate both Hmiel et al.'s results and present-day estimates of geo-CH_4. Unfortunately, no postulated or measured increase since preindustrial times is large enough to bridge the difference between Hmiel et al. and other inventories of geo-CH_4 emissions.

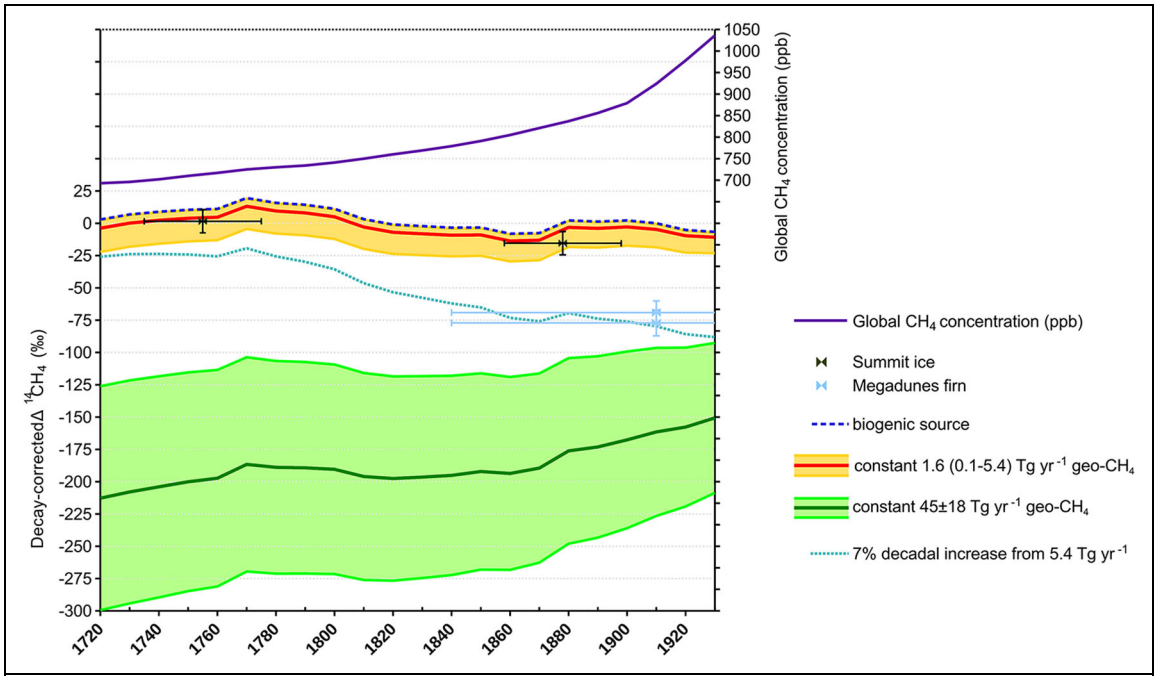

Figure 3. Geo-CH_4 emission scenarios effect on $\Delta^{14}CH_4$ for late preindustrial-early industrial period. Dark purple line is total global CH_4 emissions based on Etheridge et al. (1998). Dashed blue line is the biogenic source $\Delta^{14}CH_4$ (‰) from the IntCal20 radiocarbon age calibration curve (Reimer et al., 2020), offset by 6 years to account for CO_2 uptake and CH_4 release. The Megadunes, Antarctica firn air samples are reported in Severinghaus et al. (2010). Note that the year uncertainty on the Megadunes samples is $\pm$70 years, extending beyond the right side of the chart as drawn. The Summit Ice samples are reported in Hmiel et al. (2020). The red and green curves represent modeled scenarios for global geo-CH_4 emissions effect on global $\Delta^{14}CH_4$. Light green represents a constant 45 ± 18 Tg yr^{-1} geo-CH_4 as in Etiope and Schwietzke (2019). The red curve represents the assumption of constant 1.6 Tg yr^{-1} geo-CH_4 in Hmiel et al. (2020). Finally, the light blue curve assumes geo-CH_4 emission starts at 5.4 Tg yr^{-1} in 1700 (upper limit of Hmiel et al.) and increases at 7% per decade, reaching 47 Tg yr^{-1} in 2000 (not shown on chart). DOI: https://doi.org/10.1525/elementa.2021.00031.f3

2.2. Other local CH_4 seepages

Local CH_4 seepage emission estimates from other regions (**Figure 2b**) are also of the same order of magnitude as the minimum and median global values of Hmiel et al. (0.1 and 1.6 Tg yr^{-1}). For example, based on radiocarbon analyses, Kessler et al. (2006) calculated that 0.05–0.21 Tg CH_4 yr^{-1} escape to the atmosphere from seafloor gas seepage in the Black Sea basin. This value must be added to the shallow water bubbling seeps spread along the coasts of Turkey, Georgia, Bulgaria, Russia, and Ukraine, where CH_4 is passing through the entire water column; this would add to the atmosphere, according to emission factors based on direct measurements, roughly 0.9 Tg CH_4 yr^{-1} (Dimitrov and Vassilev, 2003). Walter Anthony et al. (2012) estimated that geologic seepage in Alaska amounts to about 0.7–1.4 Tg yr^{-1}, of which about 0.25 Tg yr^{-1} comes from measured, localized seeps. The Beaufort Sea shelf (a world-class petroleum province with thermogenic gas and oil) releases into the atmosphere about 0.05 Tg yr^{-1} (Lorenson et al., 2016), half of the global minimum reported in Hmiel et al.

The attempts by Hmiel et al. to reconcile their estimate with the existing literature fall short in several respects. With regard to submarine emissions, rather than the Hmiel et al. statement, "CH_4 emissions to the atmosphere are probably very low . . . [.]." (p. 411), we believe it would have been more rigorous to acknowledge the published estimates of geo-CH_4 emissions to the atmosphere, such as those described above and reviewed in Etiope et al. (2019). We emphasize that summing available data from just 15 submarine near-shore areas (Etiope et al., 2019) results in total emission to the atmosphere of 3.9 Tg yr^{-1}, which is more than twice the median global value of 1.6 Tg yr^{-1} proposed by Hmiel et al. (we note that, for ESAS, Etiope et al., 2019, considered the previously published average of 2 Tg yr^{-1} by Berchet et al., 2016, not the 3 Tg yr^{-1} proposed subsequently by Thornton et al., 2020).

It is worth discussing here what constitutes a bottom-up extrapolation. In a vastly oversimplified way, one could make a single measurement of CH_4 above a single gas seep or source and multiply it by the number of gas seeps in the world to obtain the global emission. For example, with regard to onshore seepages, Hmiel et al. argue that the extrapolation of source-level measurements to global scale could contribute to a difference of one order of magnitude or more: "To provide a sense of scale for the extrapolation in the case of mud volcanoes, approximately 0.0026 Tg CH_4 yr^{-1} of measured CH_4 emissions are scaled up to 6.1 Tg CH_4 yr^{-1}" (p. 411). This argument that such extrapolations are biased simply because they are too

large is misleading because it misjudges the actual effect of extrapolations in other sectors of the global CH_4 cycle. Actual extrapolation methods based on "emission factor" (statistically derived mean flux for each specific type of source) and "activity" (area of diffuse emission or number of emission points) are widely used for other natural and anthropogenic sources (e.g., wetlands, agriculture), yet their difference to top-down methods is nowhere close to a factor of 10 or more, in fact, it is less than a factor of two (Saunois et al., 2016).

A recent study used satellite retrievals over the Lusi hydrothermal system in Indonesia to determine an annual geo-CH_4 emission from this single source to be approximately 0.1 (0.140 $\pm$ 0.087) Tg yr^{-1} (Mazzini et al., 2021). This satellite retrieval is thus a regional estimate that does not rely on upscaling of surface point measurements. Nonetheless, ground-based measurements at the Lusi system also yielded an emission rate of approximately 0.1 Tg yr^{-1}. Thus, two separate estimates of the Lusi CH_4 emissions alone are more than half the 0.16 Tg yr^{-1} global minimum of Hmiel et al. In addition, the CH_4 emission factor (total emission divided by seepage area) of Lusi is consistent with the one measured in other seeps. Some controversy remains over whether the Lusi eruption was triggered by drilling or a nearby earthquake. Nevertheless, the Mazzini et al. (2021) study confirms that the statistically derived emission factors attributed to large seeps and mud volcanoes for the global bottom-up emission estimate (Etiope et al., 2019) are not overestimated by an order of magnitude, demonstrating that such extrapolations can be valid. The "big emitters" of the seep inventory in Etiope et al. (2019) include 27 mud volcanoes or mud volcano clusters, mostly in Azerbaijan, similar to or exceeding the emissions from Lusi. Each of these natural emitters may release CH_4 amounts of the same order of magnitude of those degassed by Lusi.

3. Comparison with top-down geo-CH_4 estimates

Near-zero geo-CH_4 estimates of Hmiel et al. also contest some non-$^{14}CH_4$ based top-down emission estimates (**Figure 2a**). In particular, ice-core $\delta^{13}C$–CH_4 measurement-based geo-CH_4 flux estimates (Schwietzke et al., 2016) were suggested by Hmiel et al. to be overestimates and to not provide a strong constraint on preindustrial-era geo-CH_4 fluxes because of uncertainties in the magnitude of CH_4 emissions from biomass burning. The uncertain parameter, they argue, is whether preindustrial biomass burning CH_4 emissions were comparable to the present-day biomass burning CH_4 emissions.

It is curious that in a concurrently published paper (Dyonisius et al., 2020), however, the same authors use $\delta^{13}C$–CH_4 and δD–CH_4 data in a box model to find that "biomass burning in the preindustrial Holocene were 22 to 56 teragrams of methane per year" (p. 907), a range which is comparable to today and consistent with the assumptions in Schwietzke et al. (2016). Much higher preindustrial biomass burning CH_4 emissions would be required to explain the near-zero geologic CH_4 emissions in Hmiel et al. Both scenarios cannot be true; and Hmiel et al.'s dismissal of $\delta^{13}C$–CH_4 ice-core studies would seem to preclude the use of such studies in Dyonisius et al., so the argument in Hmiel et al. of "not a strong constraint" is not valid for the purpose of this article.

Similarly, Hmiel et al. dismissed the results of Nicewonger et al. (2016), a paper which estimated preindustrial geo-CH_4 from ice-core ethane measurements. The reason again was due to assumed unconstrained preindustrial biomass burning levels, yet again, the constraints in the Dionysius et al. paper would suggest this dismissal to be unsupported.

4. Did natural seepage increase due to the fossil-fuel industry?

As a possible explanation of the discrepancy between late preindustrial ice-core-based fossil CH_4 emission and present-day data, Hmiel et al. proposed the idea that seepage may have increased over time due to petroleum extraction and ground-water aquifer depletion. To our knowledge, however, seepage data suggest the opposite: many historical seeps disappeared or their fluid activity has been strongly reduced due to gas and/or oil production (e.g., Quigley et al., 1999; Schimmelmann et al., 2018). The decrease of seepage is a result of the decrease in reservoir pore fluid pressures, which is well-known in petroleum reservoir engineering (fluid overpressure is the driving force that brings petroleum fluids to the surface; a decrease in pore fluid pressure is therefore an inevitable consequence of petroleum production) and in seepage studies (e.g., Hunt, 1996; Klusman, 2011). Moreover, geothermal CH_4 emissions, which do not occur in petroliferous basins and are not affected by the fossil fuel industry, are also estimated to emit enough CH_4 to be inconsistent with a near-zero natural geo-CH_4 estimate (global emission in a range of approximately 2–9 Tg yr^{-1}; Lacroix, 1993; Etiope and Klusman, 2002; Etiope et al., 2008). Theoretically, seepage could locally increase due to ground-water aquifer depletion, as Hmiel et al. postulated, but ground-water depletion is hardly plausible as the reason for a global emission increase of an order of magnitude higher than the late preindustrial estimates.

Geologic evidence indicates that petroleum fluids have been migrating to the surface (land or seabed) for extended periods of time. For example, Knies et al. (2018) suggested that seepage west of Svalbard has persisted since the early Pleistocene. MDAC, formed because of the anaerobic oxidation of CH_4 (Knittel and Boetius, 2009), is a ubiquitous feature of CH_4 seeps (Aloisi et al., 2000; Naehr et al., 2007; Feng et al., 2010, 2014; Ruban et al., 2020; and many others). Uranium-thorium (U-Th) dating of MDAC aragonite indicates ages of between 1.61 ($\pm$0.02) and 4.32 ($\pm$0.08) ka BP from Hola Trough, off Northern Norway (220 m water depth; Sauer et al. 2017), and since approximately 160 ka BP from Vestnesa Ridge, west of Svalbard (approximately 1,200-m water depth; Himmler et al., 2019). A shallower water example, more relevant to fossil emissions to the atmosphere, is the Croker Carbonate Slabs active seep area in the Irish Sea where MDAC, present over an area of 20 km^2 (possibly 57 km^2) and up to 6-m thick, provided U-Th ages indicating

continuous seepage since 17,000 (±5,500) yr BP (Judd et al., 2019).

5. Global CH_4 emissions and potential effects of nonconstant geo-CH_4 during the preindustrial era

The key untested assumption in Hmiel et al. behind their title claim of underestimation of present-day fossil fuel-related CH_4 emissions is that geo-CH_4 has been constant from the late preindustrial to the present. To further examine this, we have developed a thought experiment by calculating a decay-corrected $\Delta^{14}CH_4$ (‰) time series, which would be expected for a selection of potential geo-CH_4 emission rates in the late preindustrial period (**Figure 3**). As described below, we calculated time series for (i) low constant, (ii) high constant, and (iii) temporally increasing geo-CH_4 emissions. The model accounts for the fact that total atmospheric CH_4 increased during the 1755–1880 period of the two late preindustrial samples in Hmiel et al., from approximately 730 to approximately 840 ppb (Etheridge et al., 1998). We note that Hmiel et al. report a median of 214.8 Tg yr^{-1} for preindustrial global emissions (a value about 15%–25% lower than other published estimates for the late preindustrial), and they did not include explicitly increasing global CH_4 emissions. The average and median in Hmiel et al. are both well below the approximately 250 Tg yr^{-1} for the late preindustrial Holocene (years 1000–1800) and further below the estimated 1880 emissions (approximately 300–360 Tg yr^{-1}) (Etheridge et al., 1998; Prather et al., 2012; Meinshausen et al., 2017).

We calculated the biogenic source $\Delta^{14}CH_4$ based on the IntCal20 database reported in Reimer et al. (2020). This baseline was combined with the $\Delta^{14}CH_4$ shifts for the two constant geo-CH_4 emission rates seen in **Figure 3**: 1.6 and 45 Tg yr^{-1}; 1.6 (0.1–5.4) Tg yr^{-1} is the median Hmiel et al. preindustrial geo-CH_4 result; 45 ± 18 Tg yr^{-1} is the present-day geo-CH_4 from Etiope and Schwietzke (2019). A positive shift in $\Delta^{14}CH_4$ is clear for the 45 Tg yr^{-1} scenario, though not nearly enough to close the gap between the two scenarios. As expected, with only 1.6 Tg yr^{-1}, global $\Delta^{14}CH_4$ is dominated by the biogenic source's $\Delta^{14}CH_4$. Finally, we note that the lower bound of the 45 Tg yr^{-1} scenario is close to compatible with the Severinghaus et al. (2010) Megadunes, Antarctica firn sample results.

We show one additional scenario in **Figure 3,** a sufficient increase of geo-CH_4 from the preindustrial times to the present to accommodate both the upper bound of Hmiel et al. (5.4 Tg yr^{-1}) and the Etiope and Schwietzke 45 ± 18 Tg yr^{-1}. To do this, we assumed a starting geo-CH_4 of 5.4 Tg yr^{-1} in 1700, increasing by 7% per decade until the present day. This rate of increase results in 47 Tg yr^{-1} geo-CH_4 in the year 2000. A sustained increase in geo-CH_4 of slightly more than 5% per decade would reach the lower bound of Etiope and Schwietzke (27 Tg yr^{-1}). Unfortunately, we know of no publications describing processes that would account for such rates of increase in geo-CH_4 emissions. In particular, the change in geo-CH_4 may have been in a downward direction, as discussed in the Section 4. Additionally, the hypothetical time series of the temporal increase tested in this thought experiment is inconsistent with the younger of the two Summit samples in Hmiel et al. Thus, in order to reconcile Hmiel et al. with present-day geo-CH_4 estimates, an even steeper increase in geo-CH_4 emissions than the 7% per decade would be needed after 1880.

6. Conclusion

The top-down and bottom-up results described above are representative of those from a wide community of scholars who independently assessed regional and global atmospheric emissions of geo-CH_4 and whose individual conclusions are broadly consistent with each other (e.g., Lacroix, 1993; Klusman et al., 1998; Dimitrov, 2002; Judd et al., 2002; Milkov et al., 2003; Judd, 2004; Kvenvolden and Rogers, 2005; Etiope et al., 2008; Nicewonger et al., 2016; Schwietzke et al., 2016; Dalsøren et al., 2018; Etiope et al., 2019; Thornton et al., 2020). Thus, if the results in Hmiel et al. represent the true present-day geo-CH_4 magnitude, all these other studies must have overestimated the emissions by an order of magnitude or more, despite using different data sets and different methods in different regions. Judging by the success of extrapolations in other global CH_4 source analyses (e.g., wetlands and agriculture in Saunois et al., 2020), such large discrepancies are unlikely to be caused by extrapolation errors. This is a major stumbling block in reconciling the diverging global geo-CH_4 estimates.

In our opinion, a key point is the fact that the global lower range of Hmiel et al., from 0.1 to 1.6 Tg yr^{-1}, certainly is not representative of present-day geo-CH_4 emissions, as it is comparable with, or exceeded by, numerous independent local and small regional estimates. Can any errors in the method established by Petrenko et al. (2017) and Hmiel et al. (2020) be ruled out at this point (such as incomplete analyte recovery as well as biases in the inverse modeling used to calculate the geologic source)? Can processes be ruled out that might make the abundance of ^{14}C-free CH_4 in the investigated ice-core sites not representative of the global abundance? If the ice-core data (or their elaboration) resulting in global emission <1 Tg yr^{-1} are erroneous, then a thorough review of the full methodology (including assumptions used in the modeling) as well as independent reproduction of the analysis may be warranted.

Overall, the Hmiel et al. statements that geo-CH_4 emissions "[. . .] *were* about 1.6 teragrams CH_4 per year, with a maximum of 5.4 teragrams CH_4 per year" and that these estimates "provide a *firm target* (emphasis added) for inventories of the global CH_4 budget [. . .]" (p. 409) are clearly overconfident. Recalling Jackson et al. (2020): "A number as small as 5 Tg CH_4 per year for all natural geologic emissions (Hmiel et al. 2020) seems difficult to reconcile with the results of Thornton et al. (2020), the work of other researchers more broadly, and with bottom-up approaches generally."

These potential issues with data collection, analysis, and interpretation, and other questions, should be examined by a wide group of experts before we reset, on the basis of a single method and two samples, the global

estimates of geologic and anthropogenic CH_4 emissions. For the bottom-up emission estimates, future work might include the improvement of the emission factors and related activity (area or number of point sources). Some improvements can be obtained by satellite-based measurements but, considering the detection limits of presently available satellites (e.g., TROPOMI, PRISMA, Sentinel), this is possible only for large and very active macroseeps. Drone-based flux measurements may help for most seeps, offshore and onshore. Ground-based surveys (remote sensing from vehicles, e.g., Leifer et al., 2018, or punctual closed-chamber measurements) remain the best option for the geologic sources with lower fluxes, such as microseepage. Continuous analyses of sea surface air in the vicinity of, and down-current from, offshore seep areas acquired during ship-borne surveys provide indications of sea-air CH_4 flux (Judd, 2015).

A wide gap exists between the geo-CH_4 estimate of Hmiel et al. and numerous previous studies utilizing a wide variety of methodologies for geo-CH_4 determination. We show that while very large temporal geo-CH_4 increases since early industrial times could in theory reconcile Hmiel et al. with present-day geo-CH_4 estimates, there is no known literature describing the potential mechanisms of such a trend. In fact, only mechanisms for the opposite trend are currently known. We hope that the top-down and bottom-up comparisons in this article as well as a previous review (Etiope and Schwietzke, 2019) will encourage the wider CH_4 science community to further improve the data collection, analysis, and interpretation of top-down and bottom-up methods rather than simply accepting the stark divergence in global geo-CH_4 estimates recently suggested by the Hmiel et al. paper. The potential importance of the Hmiel et al. result to our understanding of anthropogenic CH_4 emissions demands such close attention before we dramatically revise our understanding of Earth's natural CH_4 emissions on the basis of a single method and two samples.

Data accessibility statement

All data discussed in this article are available in the related literature references provided below.

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Competing interests

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Author contributions

All coauthors participated in conceiving, drafting, and revising the article and approved the submitted version.

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