A new estimate of global methane flux from onshore and shallow submarine mud volcanoes to the atmosphere

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Abstract A new estimate of global methane emission into the atmosphere from mud volcanoes (MVs) on land and shallow seafloor is presented. The estimate, considered a lower limit, is based on 1) new direct measurements of flux, including both venting of methane and diffuse microseepage around craters and vents, and 2) a classification of MV sizes in terms of area (km^2) based on a compilation of data from 120 MVs. The methane flux to the atmosphere is conservatively estimated between 6 and 9 Mt y^{-1}. This emission from MVs is 3–6% of the natural methane sources and is comparable with ocean and hydrate sources, officially considered in the atmospheric methane budget. The total geologic source, including MVs, seepage from seafloor, microseepage in hydrocarbon-prone areas and geothermal sources, would amount to 35–45 Mt y^{-1}. The authors believe it is time to add this parameter in the Intergovernmental Panel on Climate Change official tables of atmospheric methane sources.

Keywords Methane · Mud volcanoes · Greenhouse gas · Climate change

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Introduction

Geologic emission of fossil (radiocarbon-free) methane has been recently recognised as an important component of global natural methane sources, being in the range of 13 to 36 Mt y^{-1} (Judd and others 2002) or 30 to 70 Mt y^{-1} (Etiope and Klusman 2002). These estimates imply that the cumulative geologic source of methane is 8–18% of the anthropogenic sources, and is equal to or exceeds many sources and sinks officially considered in the tables of the IPCC (IPCC 2001). Geologic flux may be also considered as a "missing source" of the radiocarbon-free methane budget suggested for the unbalance between the sum of the fossil-fuel sources and the portion of fossil CH_4 in the atmosphere (Crutzen 1991; Lacroix 1993). However, the estimates of the various geologic source strengths, including gas release in hydrocarbon-prone sedimentary basins and geothermal areas, need to be refined. Submarine gas seeps and mud volcanoes on land and the seafloor seem to be the largest geologic methane sources. An updated estimate of global submarine seepage (about 20 Mt y^{-1} to the atmosphere) is reported by Kvenvolden and others (2001). The attention is focused on mud volcanoes (MVs), which can be the most spectacular expressions of natural release of greenhouse gases. Methane is the dominant gas emitted from most MVs (Milkov and others 2003), and the global methane flux from MVs essentially equals the global gas flux. Genesis, geology and geochemistry of MVs have already been described in detail (Milkov 2000; Dimitrov 2002; Kopf 2002; Revil 2002). Direct measurements of methane flux into the atmosphere are still scarce, being available only from subaerial MVs of Italy (Etiope and others 2002; 2003), Romania (Etiope and others 2004) and partially of Azerbaijan (Dadashev 1963; Jakubov and others 1971). To date, six estimates of the global methane flux from MVs have been reported (Dimitrov 2002; 2003; Etiope and Klusman 2002; Kopf, 2002; 2003; Milkov and others 2003). In this work the authors converge and refine the estimates produced by Milkov and others (2003) and Etiope and Klusman (2002) on the basis of new flux data, including diffuse microseepage around craters and vents (not considered by other authors), and a new compilation on worldwide MV size data. The authors do not consider gas flux from deep-water mud volcanoes in this paper because their contribution to methane budget of the atmosphere may be insignificant (Milkov and others 2003).

Previous global MV flux estimates

The six available global estimates of methane flux from MVs are reported in Table 1. Although the estimates were published within a two-year time frame, they were based on different approaches and data sets. The estimate of Kopf (2002) is based mainly on previous rough estimates (no direct measurements) of methane flux from submarine MVs in the Mediterranean Ridge and the calculation procedure is not described. Microseepage from subaerial MV flanks is not mentioned and it is not clear if the methane flux refers to the "true" emission (emission at seafloor from submarine MVs) or to the output into the atmosphere. The final value of ~0.08–1.4 Mt y^{-1} is one to three orders of magnitude lower than the other estimates.

One year later Kopf (2003) proposes a wide range of 0.2 to 123 Mt y^{-1}, for total flux to the ocean and to the atmosphere, with the upper value 3 orders of magnitude higher than the previous value reported in Kopf (2002). Such a rough estimate results, however, from calculation errors in Table 3 of Kopf (2003) and is anyway derived considering flux data of different nature and significance, direct measurements (e.g., Etiope and others 2002) and indirect estimates from offshore MVs (e.g. Henry and others 1996) or MV eruptions (e.g., Guliyev 1992), without any filtering or data quality check. From the data reported in Table 3 of Kopf (2003) it appears that on average 99.8% of the global flux is from offshore MVs, and the onshore MVs emit globally only from 46.6 t y^{-1} to 0.33 Mt y^{-1}. The lower value is clearly illogical: it is 1/10 of the gas amount typically produced just by one MV, such as those in Sicily and Eastern Romania (Etiope and others 2002; 2004). The upper value is one order of magnitude lower than the other estimates of Dimitrov (2003), Etiope and Klusman (2002) and Milkov and others (2003) (see Table 1). It also should be noted that Kopf (2003) misquoted the results presented in Milkov and others (2003) confusing the values of global gas flux from onshore and offshore MVs (~33 Mt y^{-1}) with the gas flux to the atmosphere (~6 Mt y^{-1}).

The estimate of Dimitrov (2002) is based on an extensive Russian literature review from which a mean flux of 0.3 to 2.4 Gg CH$_4$ y^{-1} is derived for an "average" single MV. The product of this flux by a total of 1,100 MVs located onshore and on the shelves offshore (without considering the water depth and the related dissolution in water) leads to the global quiescent flux of 0.33–2.64 Mt y^{-1}. Estimations of eruptive flux (0.25 Mt per event) and eruption periodicity are considered to obtain a total output of 10.3 to 12.6 Mt y^{-1}. These results imply that the global degassing during eruptions is from 5 to 30 times greater than degassing during quiescent periods. Dimitrov (2002) further states that this estimate is very speculative and must be regarded with great skepticism.

One year later Dimitrov (2003) assumes an average quiescent flux of 2.4–2.6 Gg CH$_4$ y^{-1} (the upper value is from Milkov and others, 2003) and a similar eruptive flux (i.e., now the quiescent-eruptive flux ratio is 1:1), obtaining a global flux of about 5 Mt y^{-1}. No new data are considered with respect to the previous estimate.

The estimate of Etiope and Klusman (2002) refers to onshore MVs only and is based on the first experimental flux data (hundreds of direct measurements of vents and soil microseepage) from European MVs (Etiope and others 2002; 2003) and an unpublished data-set from Azerbaijan (provided by A. Feizullaiyev, Geological Institute of Azerbaijan). The authors for the first time distinguish the methane flux from vents from the diffuse emission from soil (microseepage) that has been discovered to be significant and occurring throughout the MV area (Etiope and others 2002; 2003; 2004). In total 800 onshore MVs (Guliyev and Feizullayev 1997) were considered as a lower limit. For a range of 10–1000 t y^{-1} from each MV, 600 MVs are estimated to emit between 10^3–10^6 t y^{-1} in quiescent periods. Adding the regional Azerbaijan (about 200 MVs) estimates, all MVs would conservatively emit at least 1–2 Mt CH$_4$ y^{-1} from vents only, and potentially reach 10 Mt y^{-1} including microseepage from flanks and surrounding soil, and eruptions.

The estimate of Milkov and others (2003) is based on 36 flux data from individual MVs (including the direct measurements of Etiope and others 2002) and the assumption that the estimates are log-normally distributed. The authors considered the same total number of MVs (1,100) as reported by Dimitrov (2002) and derived a global quiescent gas flux to the atmosphere of 2.9 Mt y^{-1}. Regarding the eruptions, a periodicity of 17 events per year worldwide and an average eruption output of 0.18 Mt (Dadashev 1963) were considered. The

Table 1
Estimates of global methane flux from MVs (Mt y^{-1})

	Flux to the atmosphere		Flux to atmosphere and oceans
	Quiescent degassing	Quiescent and eruptions	
Dimitrov (2002)	0.33–2.64	10.3–12.6	–
Etiope and Klusman (2002)[a]	>1–2[b]	5–10[c]	–
Kopf (2002)	–	–	0.08–1.4
Milkov and others (2003)	2.9	6	33
Kopf (2003)[d]		0.00005–0.328	0.2–123
Dimitrov (2003)		5	
This study	>3–4.5	>6–9	

[a] onshore MV only
[b] flux from vents
[c] flux from vents and flank microseepage
[d] estimate not reliable; significant arithmetic mistakes are made during calculations and data manipulations

total flux to the atmosphere is ∼6 Mt y^{-1}, almost half of which is from degassing during eruptions. Milkov and others (2003) further suggested that deep-water MVs may emit ∼27 Mt y^{-1} of gas but their contribution to the atmospheric methane source may be insignificant due to methane consumption and sequestration at the seafloor and in the water column.

Basically the estimates of Etiope and Klusman (2002), Milkov and others (2003) and Dimitrov (2003) are quite similar.

Data review and elaboration

The procedure followed for a new global flux estimate is presented in Table 2. The procedure is based on the following steps:

- determination of the number of MVs and classification of MVs according to their size (area);
- calculation, for each MV class, of global onshore flux based on field measurements of specific flux (the amount of gas emitted per unit time per unit area), including emissions from vents and diffuse microseepage;
- calculation, for each MV class, of global offshore flux based on estimates of emission from vents and bubble-dissolution models (i.e. taking water depth into account).

MV classes and number

Dimitrov (2002) reports a total of at least 926 onshore and 270–570 offshore MVs. In general, the number of MVs reported for a given area, however, is not based on a sufficient structural and morphological examination. As a result, in many documents, including Dimitrov (2002), “mud volcanoes” may refer to single edifices, groups of vents (craters or gryphons), or mud volcano clusters (Etiope and others 2003). The authors tried to select data on MV areas referring clearly to structures (edifices) that have a main central crater and satellite vents, or clusters of cones forming homogeneously a single muddy area morphologically elevated, and distinct from the sur-

Table 2
Procedure for a new global estimate of methane flux from mud volcanoes

	Small-size	Medium-size	Large-size		Total
			Large	Giant	
ON-SHORE					
Number[a]	417	417	86	6	926
Single MV area[b] (km^2) mean (range)	0.18 (0.0002–0.5)	3.5 (0.5–9)	13 (9–17)	38 (20–58)	–
Single seepage area[c] (km^2)	0.75	5.3	16.4	40	–
Total area[d] (km^2)	75	1459	1118	228	2880
Total seepage area[e] (km^2)	313	2210	1410	240	4173
Specific methane flux[f](t km^{-2} y^{-1})	100–500	500–1000	≈1000		–
Total flux[g](Mt/y)					
quiescent degassing	0.03–0.16	1.1–2.2	1.65		2.8–4.0
quiescent+eruption	0.06–0.32	2.2–4.4	3.30		5.6–8.0
SHALLOW OFFSHORE					
Number[h]	135	135	30		300
Venting flux per MV[i] (t/y)	10–200	100–3000	10000		–
Total quiescent flux at seabed[j] (t/y)	1350–27000	13500–405000	300000		315000–732000
Total flux to the atmosphere[k] (Mt/y)					
quiescent degassing	0.002–0.02	0.009–0.26	0.2		0.21–0.48
quiescent+eruption	0.004–0.04	0.02–0.52	0.4		0.42–0.96
TOTAL CH_4 FLUX (Mt/y)					6–9

[a] all numbers are lower limits; based on total number of MVs by Dimitrov (2002) and class percentages from Fig. 1

[b] from Fig. 1

[c] assuming the radius of seepage area outside of a MV edifice to be 250 m greater than the size of the MV itself (see text and Fig. 2)

[d] a×b

[e] a×c

[f] from Etiope and Klusman (2002), Etiope and others (2002; 2003, 2004); for large MVs the mean flux is derived from Milkov and others (2003) and Jakubov and others (1971)

[g] (e×f/10^6); eruption output equivalent to quiescent degassing as suggested by Milkov and others (2003)

[h] conservative number of shallow submarine MVs within 200 m depth (shelf); class percentage from Fig. 1

[i] mean fluxes of the 3 MV classes from Etiope and others (2002; 2003, 2004), and Milkov and others (2003; Table 1)

[j] h×i

[k] based on 20% (for 150 MVs) and 50% (for 150 MVs) of dissolution as suggested by bubbling-dissolution models (Leifer and Patro 2002)

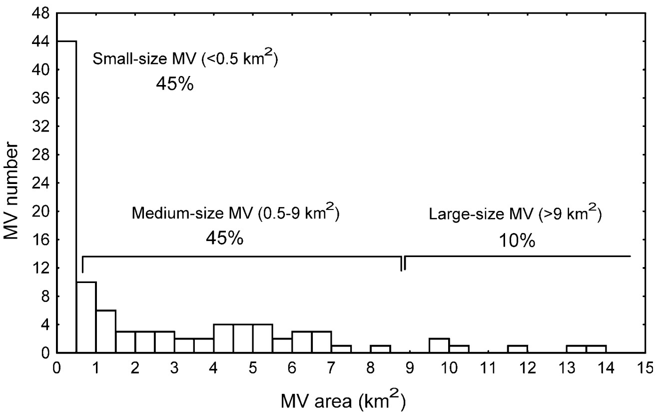

Fig. 1
Distribution and classification of MV size from a data set of 120 MVs

rounding soil and vegetation type. Data (area in km^2) from 120 individual onshore and shallow submarine MVs were compiled, on the basis of available scientific literature, personal communications, and web sites. The 120 MVs are from Azerbaijan (Jakubov and others 1971), Sakhalin, Russia (Gurieva and Sharkov 1987), Panama (Reed and others 1990), Adriatic Sea (Hovland and Curzi 1989), Italy (Etiope and others 2002; 2003; Martinelli 1999), Mexico (Humphrey 1963), Romania (Etiope and others 2004), Irian-Jaya/Papua N.G. (Kopf 2002), Ukraine (Solecki personal communication), Pakistan (Delisle and others 2002), Turkmenistan (Kopf 2002), Venezuela (Aslan and others 2001), Taiwan, Alaska, India (web sites), Trinidad (Dia and others 1999 and web sites). On the basis of the resulting histogram (Fig. 1) the following distinction of MVs in three classes based on their area was proposed:

- small-size MV: <0.5 km^2(45%)
- medium-size MV: 0.5–9 km^2(45%)
- large-size MV: >9 km^2(10%)

Large-size class includes at least 6 MVs exceeding 20 km^2: the authors call them “giant” MVs. The authors assume that the data set analysed, and consequently the distribution of the MV categories as summarized in Figure 1, are representative of the global population of MVs including shallow submarine MVs.

Consequently, the number of MVs for each class is estimated to be:

	onshore	shallow offshore
– total MVs (1226)	926	300
– small size MVs	417	135
– medium size MVs	417	135
– large size MVs	92	30

It is emphasized again that these estimates have some uncertainties due to the variable definition of MVs (Etiope and others, 2003), but they are considered as lower limits. New mud volcanoes may be found on the shelves as exemplified by recent discoveries offshore Sicily (Holland and others, 2003) and in the East China Sea (Yin and others, 2003).

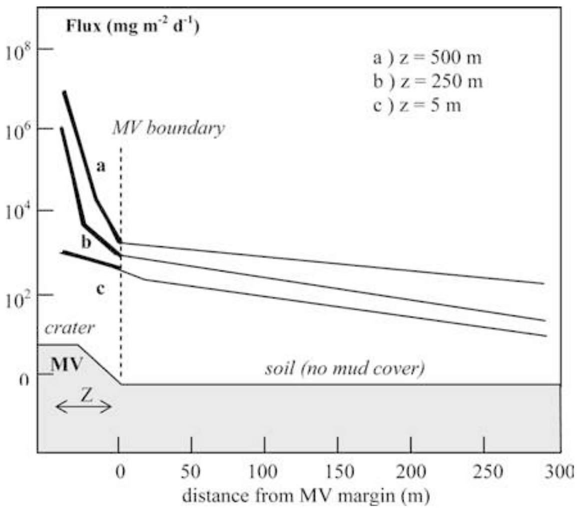

Fig. 2
Profile illustrating methane flux decreasing from the MV boundary toward the surrounding soil. Z is the mean distance between the central crater and the MV boundary. a) Paclele Mari and Paclele Mici MVs, Eastern Romania (mean of 3 profiles elaborated from data in Etiope and others, 2004); b) Maccalube MV, Sicily (mean of 2 profiles; Etiope and others 2002); c) Pineto MV, central Italy (mean of 2 profiles elaborated from data in Etiope and others 2003). The average output at the vents (0.001, 1 and 10 kg/d) is equivalent to about 10^3, 10^6 and 10^7 mg m^{-2} d^{-1}, respectively. In all cases positive diffuse CH_4 flux from the ground occurs up to 500–1000 m out of the muddy area

Flux from onshore MVs

The field surveys carried out in small- and medium-size MVs in Italy and Romania (Etiope and others 2002; 2003; 2004) indicated that MVs typically emit a specific methane flux in the range from 10^2 to 10^3 t km^{-2} y^{-1}and that the diffuse and pervasive microseepage around MV craters, gryphons and bubbling pools is a fundamental component of the total methane output. The soil CH_4 positive flux (on the order of 10^2–10^5 mg m^{-2} d^{-1}) is significantly high even at large distances (more than 1 km) from the MV edifices, suggesting that microseepage exists over wide areas. Figure 2 shows typical flux patterns towards the external area of the MVs investigated. Microseepage is measured to be on the order of 10^2 mg m^{-2} d^{-1}at distance about 250–300 m from the muddy cover. Accordingly, a seepage area is defined as a combination of MV area and a peripheral bound with a radius of 250 m from the MV boundary. Then, a global seepage area was estimated and multiplied by the specific flux experimentally measured in quiescent periods in small- and medium-size MVs (Etiope and Klusman 2002; Etiope and others 2002; 2003; 2004). For large MVs a mean ratio flux/area is derived from data reported by Milkov and others (2003) and Jakubov and others (1971).

Conservatively the authors assumed that eruptive degassing equals the quiescent output, as in Milkov and others (2003), although Dimitrov (2002) implies that gas flux from quiescent periods is significantly (by a factor up to 30) less than the flux during eruptions.

The authors obtain conservatively 2.8–4 Mt y^{-1} for onshore quiescent degassing and assume 5.6–8 Mt y^{-1} including eruptions.

Flux from shallow offshore MVs

Shallow offshore MVs were defined as the MVs located on the shelf at depths less than 200 m. From the main references on the Caspian, Azov, Black, Caribbean, Mediterranean and Yellow Seas (Dimitrov 2002; Kopf 2002, Holland and others 2003; Yin and others 2003), the authors estimate conservatively that at least 300 shallow MVs exist, and that at least 150 are at a depth of <50 m. Considering the recent bubble-dissolution models of Leifer and Patro (2002), for a mean bubble radius of 1–2 cm, the authors calculate that for depths ≤ 50 m and ≤ 100 m (average shelf depth) about 80% and 50% of methane enters the atmosphere, respectively. Only flux from vents (bubbling plumes) were considered as diffuse microseepage for offshore MVs is likely to be insignificant due to oxidation and dissolution processes. The authors obtain a global methane flux from shallow offshore MVs to the atmosphere of at least 0.4–0.9 Mt y^{-1}.

Discussion and conclusions

The global methane flux from onshore and shallow submarine MVs is estimated conservatively between 6 and 9 Mt y^{-1}. This is the first estimate which considers accurately direct flux measurements including microseepage from MV flanks and surrounding zones, that are significant components of the MV gas output. The procedure followed did not distinguish vent-flux and microseepage in onshore MVs, as the total specific flux was considered. The authors may test the global estimate considering separately the two flux components. The small- and medium-size MVs of Italy and Romania (excluding everlasting fires and restricted zones of enhanced soil degassing) show a microseepage specific flux between 280 and 600 t km^{-2} y^{-1}, with a mean of 400 t km^{-2} y^{-1}. Applying this range to the 120 MVs (total area of 516 km^2) yelds a microseepage flux between 144,000 and 310,000 t y^{-1}. Assuming that the 120 data are representative of the global population of onshore MVs (926), the total microseepage flux would be 1–2.4 Mt y^{-1}. The measured MVs (excluding everlasting fires) show a microseepage/vent-flux ratio from 0.7 to 10. Applying this range to all MVs, considering the previous microseepage estimate, yields a total vent-flux from 0.1 to 3.4 Mt y^{-1}. Therefore, the total vent and microseepage quiescent flux from onshore MVs would be between 1.1 and 5.8 Mt y^{-1}. The previous estimate (excluding eruptions) lies within this range.
The authors outline again that the uncertainties are related to:

- limited quiescent flux data-set and the lack of measurements during eruptive events;
- poorly constrained numbers of MVs and their areas;
- the short-distance spatial variability, and short-term temporal variability in flux rates.

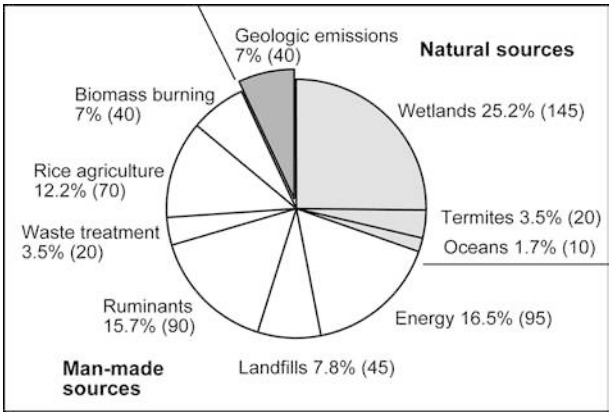

Fig. 3
Sources of atmospheric methane including geologic emissions (average fluxes in Mt y^{-1} in brackets). Non-geologic sources are from IPCC (2001; average of seven published estimates). See for comparison Judd and others (2002). Geologic emissions are due to mud volcanoes, microseepage, geothermal fluxes, and marine seepage, including flux from hydrates (which are the only geologic source considered by IPCC)

The total identified source of methane (Lelieveld and others 1998; IPCC 2001) is presently estimated, on average, at 600 Mt y^{-1}, of which about 32% is from natural sources (wetlands ~145 Mt y^{-1}, termites ~20 Mt y^{-1}, oceans ~10 Mt y^{-1}, hydrates 5–10 Mt y^{-1}) and 68% from anthropogenic emissions (including rice paddies ~70 Mt y^{-1}, biomass burning ~40 Mt y^{-1}, enteric fermentation ~90 Mt y^{-1}, fossil methane from the coal mining and oil industry 95–110 Mt y^{-1}). The new estimate of global emission from MVs represents 3–5% of the natural methane sources in the atmosphere and it is at the same level of ocean and gas hydrate sources. The total geologic source, including MVs (6–9 Mt y^{-1}; this work), submarine seepage (20 Mt y^{-1}; Kvenvolden and others 2001), microseepage in hydrocarbon-prone areas and geothermal sources (at least 7 and 2.5–6.3 Mt y^{-1} respectively; Etiope and Klusman 2002), would amount to around 35–45 Mt y^{-1}. This value is comparable to or is higher than several other sources (Fig. 3) and sinks considered by the IPCC (e.g., termites, biomass burning, landfills, soil uptake), and it is 30–40% of the fossil (radiocarbon free) methane source (which is around 20% of the total source; Lelieveld and others 1998). The authors believe it is time to add this parameter into the official IPCC tables of the atmospheric methane budget. To progress beyond these calculations, further data from field measurements are needed on the major MVs of the world; for submarine MVs, technological solutions must be developed to perform reliable measurements in coastal and deep-water areas.

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