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Global gas flux from mud volcanoes: A significant source of fossil methane in the atmosphere and the ocean

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[1] There are yet unidentified sources of fossil methane (CH_4) in the atmosphere. Mud volcanoes (MVs) are a potentially significant but poorly quantified geologic source of fossil hydrocarbon gases and CO_2 to the atmosphere and the ocean not included in the current models of sources and sinks. Our statistical analysis of 36 previous measurements and estimates of gas flux from individual MVs suggests that the global gas flux may be as high as ~33 Tg yr^{-1} (~15.9 Tg yr^{-1} during quiescent periods plus ~17.1 Tg yr^{-1} during eruptions). Onshore and shallow offshore MVs are estimated to contribute ~6 Tg yr^{-1} of greenhouse gases directly to the atmosphere. MVs may contribute ~9% of fossil CH_4 missing in the modern atmospheric CH_4 budget, and ~12% in the preindustrial budget. Large volumes (~27 Tg yr^{-1}) of gas may escape from deep-water MVs, suggesting that global gas flux from the seafloor may be underestimated.

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1. Introduction

[2] Fossil (^{14}C-depleted) methane (CH_4) provides ~110 ± 45 Tg yr^{-1} to the atmosphere that has total CH_4 source ~600 ± 80 Tg yr^{-1} [*Crutzen and Lelieveld*, 2001]. Coal mines (~45 ± 15 Tg yr^{-1}) and natural gas hydrate decomposition (5–10 Tg yr^{-1}) are suggested to be significant sources of CH_4 in the atmosphere [*Lelieveld et al.*, 1998]. However, CH_4 venting from hydrate-bearing sediments is rapidly oxidized and dissolved in the water column [*Judd et al.*, 2002], and CH_4 flux from gas hydrate to the atmosphere may be negligible. Significant but poorly constrained volumes of CH_4 leak from natural gas pipelines and petroleum wells [*Lelieveld et al.*, 1998], but this probably cannot account for ~65 Tg yr^{-1} missing from the modern CH_4 budget. Furthermore, ~50 Tg yr^{-1} may be missing from the preindustrial CH_4 budget [*Quay et al.*, 1991]. Search for yet unidentified or/and not included in the global budget sources of fossil CH_4 is ongoing.

[3] Geological sources of fossil CH_4 in the atmosphere are generally thought to be minor [*Lelieveld et al.*, 1998]. However, *Etiope and Klusman* [2002] and *Judd et al.* [2002] estimate the total geologic CH_4 source to be on the order of 30–70 Tg yr^{-1} and 16–40 Tg yr^{-1}, respectively. These recent studies call for more accurate estimates of CH_4 flux from geologic features since their input to the atmosphere may be comparable with the input from other natural (e.g., termites) and anthropogenic (e.g., animal wastes) sources.

[4] Mud volcanoes (MVs) occur worldwide (Figure 1) and represent constructional features (diameter up to 10 km, relief up to 700 m) from which sediments and fluids (water, dissolved salts, gas, and oil) flow or erupt [*Milkov*, 2000; *Kopf*, 2002; *Dimitrov*, 2002]. Approximately 1,100 MVs are documented onshore and in shallow water on continental shelves [*Dimitrov*, 2002] and 10^3–10^5 MVs may exist on continental slopes and abyssal plains [*Milkov*, 2000]. MVs are most common in areas of rapid deposition and lateral tectonic compression with overpressure. MVs occur over seafloor-piercing shale diapirs or as a consequence of migration of fluidized sediment along active faults [*Milkov*, 2000].

[5] MVs are suggested as an important source of CH_4 with other hydrocarbon and non-hydrocarbon gases to the atmosphere and to the ocean. *Dimitrov* [2002] estimates that 10.2–12.6 Tg yr^{-1} of CH_4 is released from onshore and shallow offshore MVs. *Etiope and Klusman* [2002] argue that at least 1–2 Tg yr^{-1} and as much as 10–20 Tg yr^{-1} may be emitted from onshore MVs. On the other hand, *Kopf* [2002] suggests that all MVs on Earth (both onshore and offshore) release only 0.08–1.41 Tg yr^{-1} of gas. The major reason for such discrepancy in the existing estimates is that they are based on the simplified analysis of limited data. In this study we compiled 36 previous measurements and estimates of gas emission from MVs and used a statistical model to estimate global gas flux from MVs, and to assess their significance as a source of fossil gases in the atmosphere.

2. Global Gas Flux Composition

[6] MVs emit gases during long quiescent periods (years to centuries) and during short eruptive periods (hours to days) [*Guliev and Feizullayev*, 1996]. During the quiescent periods, gas bubbles continuously flow from numerous vents, and dissolved gases diffuse from MV sediment. Periods of eruptions of MVs are characterized by rapid flow and, sometimes, ignition of large volumes of hydrocarbon gases [*Bagirov et al.*, 1996]. To constrain the composition of global gas flux from MVs, its "average" geochemical properties are estimated here from gas compositions of 161 onshore and offshore MVs from 15 areas (Figures 2 and 3). Concentration of CH_4 in gas from MVs varies from 0% to 99.9%, the mean is

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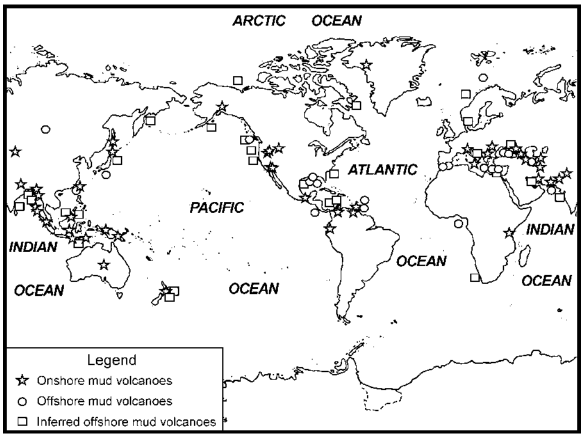

Figure 1. Worldwide distribution of MVs [modified from *Milkov*, 2000]. MVs are documented in 44 onshore and 21 offshore areas, and inferred in 25 offshore areas.

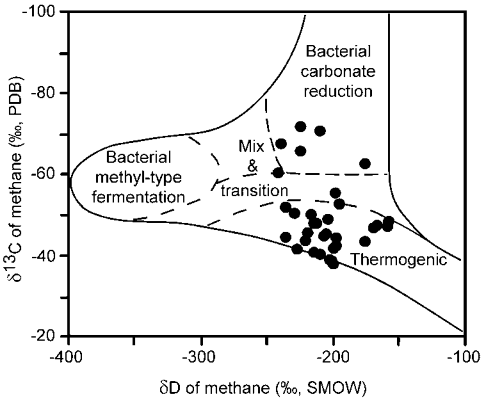

Figure 3. Relationship between $\delta^{13}C$ and δD values for CH_4 emitted from 33 onshore and offshore MVs. Fields of bacterial and thermogenic CH_4 are depicted after *Whiticar* [1999].

86.2%, and the median is 94.1%. Where CH_4 is absent or in low concentration, CO_2 and nitrogen may be major components of the gas. Concentration of CO_2 varies from 0% to 99.9%, the mean is 8.8%, and the median is 2.4%. Concentration of nitrogen varies from 0% to 75.8%, the mean is 4.6%, and the median is 0.6%. MVs whose flux is largely CH_4 often emit C_{2+} hydrocarbon gases (ethane through pentanes) in low relative abundance. Concentration of C_{2+} gases varies from 0% to 15.9%, whereas the mean is 0.5% and the median is 0.1% (Figure 2).

[7] MVs that emit gas in which CH_4 and C_{2+} gases are major components are dominant globally and commonly occur in petroliferous areas [*Guliev and Feizullayev*, 1996]. Molecular and isotopic properties of CH_4 appear to be consistent with thermogenic, bacterial, or are of mixed origin (Figure 3). CH_4 emitted by MVs should be ^{14}C-depleted (i.e., fossil) since most MVs apparently emit thermogenic or mixed gas, although this assumption is poorly constrained by ^{14}C measurements. A few MVs emit gas in which CO_2 and nitrogen are major components, and occur in association with geologically recent magmatic processes [e.g., Copper River basin in Alaska, *Motyka et al.*, 1989].

3. Estimate of Global Gas Flux

3.1. Quiescent Periods

[8] For quiescent periods, the data set analyzed here includes 36 measurements and estimates of gas flux from individual MVs in various geological settings (Table 1). The data set analyzed is assumed to be representative of the global population of MVs. Although statistical analysis of the gas flux data appears feasible and worthwhile, several factors introduce uncertainty to the estimate of global gas flux from MVs, and should be mentioned. First, accuracy of measurements and estimates presented in Table 1 varies widely. Gas flux was measured directly only at MVs in Sicily [*Etiope et al.*, 2002] and in Azerbaijan [*Dadashev*, 1963]. Estimates of gas flux from submarine MVs are derived using various approaches. Second, only estimates from Sicily encompass both flux of free gas from vents and diffuse soil degassing [*Etiope et al.*, 2002]. The study of *Etiope et al.* [2002] suggests that total gas from diffuse degassing is an order of magnitude higher than total gas from venting. Estimates from other areas include only emission of free gas and thus are likely to be underestimates. Furthermore, values in Table 1 represent measurements and estimates of gas flux within short time spans. However, gas flux from MVs varies significantly over years and even days [*Guliyev and Feizullayev*, 1996]. Thus, our results should be considered as a first-order approximation because of limited measurements and uncertain estimates. More accurate measurement of free gas *and* diffuse flux from specific sites over long time spans are required to better constrain the range and the statistical distribution of gas fluxes.

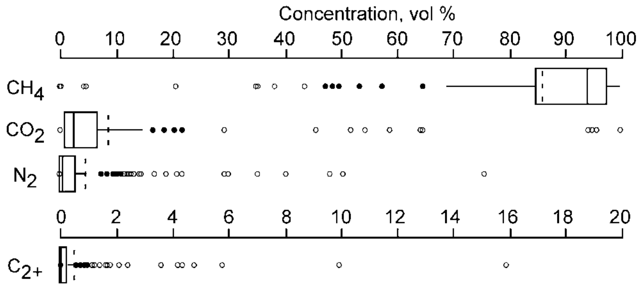

Figure 2. Box plots illustrating composition of gas flux from 161 onshore and offshore MVs.

[9] Gas flux from individual MVs during quiescent periods varies from 10^2 to 1.5×10^7 m^3 yr^{-1}. Log-transformed values of gas flux appear in reasonable agreement with a normal distribution ($p = 0.4843$ from Shapiro-Wilk test; Figure 4). Distribution parameters are estimated using standard methods [*Shao*, 1999] assuming data are log-normally distributed. Mean gas flux from MVs (3.6×10^6 m^3 yr^{-1}) is calculated as a function of estimated parameters.

[10] Using the mean gas flux, we estimate the total global gas flux from 1,100 MVs onshore and in shallow water to be 3.96×10^9 m^3 yr^{-1} (2.9 Tg yr^{-1}) during quiescent periods. *Dimitrov* [2002] used a more limited set of data, and estimated CH_4 flux from onshore and shallow MVs to be 0.33–2.64 Tg yr^{-1}. Our statistical model may provide a better-constrained result that lies near the upper range of the previous estimate. Furthermore, our result is similar to the conclusion of *Etiope and Klusman* [2002] who suggest that

Table 1. Measurements and Estimates of Gas Flux From MVs

Region	Mud volcano	Gas flux ($m^3\ yr^{-1}$)
Sicily	Maccalube[a]	5.8×10^5
	Bissana[a]	4×10^3
	Fuoco di Censo[a]	9×10^3
	Salinelle Stadio[a]	7.3×10^2
	Salinelle S. Biagio[a]	3.1×10^3
Azerbaijan	Nabur[b]	5.5×10^5
	Dashgil[b]	1.5×10^7
	Kyurovdag[c]	1×10^4
	Mishovdag NW[c]	9.9×10^3
	Mishovdag SE[c]	6.4×10^3
	Dourovdag[c]	1.3×10^5
	Douzdag[c]	6.9×10^3
	Pil'pilya[c]	1×10^2
	Neftechal. Sopka[c]	4.8×10^3
	Gamma[c]	4×10^3
	Byandovan[c]	6.9×10^3
	Kalmas[c]	1.3×10^3
	Kotourdag[c]	3.2×10^4
	Airantekyan[c]	5.5×10^4
	Zotova[c]	2.6×10^4
	Arzani[c]	2.5×10^4
	Gotourlik[c]	6.7×10^3
	Yurkini Sal'zi[c]	1×10^5
	Oumbaki[c]	4.2×10^5
	Ahtarma[c]	7.3×10^5
	Otmanbozdag[c]	4.4×10^4
	Outal'gi[c]	1.4×10^5
	Dashgil[d]	$>8 \times 10^2$
Sakhalin	Pugachevskiy[e]	1.1×10^7
Trinidad	Lagon Bouffe[f]	5.3×10^6
	Palo Seco[f]	5.3×10^6
Mediterranean Sea	Milano[g]	$(1.4–6.2) \times 10^5$
	Napoli[g]	$(2.7–10.1) \times 10^5$
Norwegian Sea	Haakon Mosby[h]	1.5×10^5
Offshore Barbados	Atalante[i]	4.3×10^6
	Cyclope[i]	3.2×10^5

[a]*Etiope et al.* [2002], [b]*Jakubov et al.* [1971], [c]*Dadashev* [1963], [d]*Hovland et al.* [1997], [e]*Vereschagin and Kovtunovich* [1970], [f]*Deville et al* [2001], [g]*Kopf and Behrman* [2000], [h]*Ginsburg et al.* [1999], [i]*Henry et al.* [1996].

the minimum gas flux from onshore MVs during quiescent periods is 1–2 Tg yr^{-1}.

[11] It is more difficult to estimate gas flux from deep-water MVs since their total number is poorly constrained. *Milkov* [2000] suggests that 10^3–10^5 MVs may be present on continental slopes and abyssal planes. This assumption is consistent with the recently confirmed wide distribution of MVs in the ocean, especially on active continental margins [*Kopf*, 2002]. For the purpose of this study we conservatively assume the occurrence of 5,000 deep-water MVs. Then the global gas flux from deep-water areas during quiescent periods is $18 \times 10^9\ m^3\ yr^{-1}$ (13 Tg yr^{-1}).

3.2. Eruptive Periods

[12] No direct measurements of gas flux exist during eruptive periods at MVs. A few estimates imply that high gas flux on the order of 10^7–$10^{10}\ m^3$ over periods of days may be possible [*Dadashev*, 1963; *Guliev and Feizullayev*, 1996; *Dimitrov*, 2002]. In Azerbaijan, 220 MVs are known, of which 60 erupted 250 times over 185 years [*Guliyev and Feizullayev*, 1996]. Thus, average frequency of recorded eruptions in Azerbaijan is 1.35 per year. However, many eruptions of MVs were not recorded during initial observations in the 19th and 20th centuries [*Bagirov et al.*, 1996]. Remote and short-term eruptions not associated with flames are most likely to be absent from the historical record. Based on statistical analysis of historical data, *Bagirov et al.* [1996] calculate that in Azerbaijan the average frequency of eruptions is ~9.7 per year, and strong eruptions occur at a frequency of 3.4 per year.

[13] Assuming similar proportions of eruptive MVs (~27%) and similar frequency of strong eruptions as in Azerbaijan, we estimate that 1,100 onshore and shallow offshore MVs erupt ~17 times per year. Average gas flux during recorded strong eruptions of MVs in Azerbaijan is estimated to be $2.5\times10^8\ m^3$ [*Dadashev*, 1963]. If we assume the same average gas flux for other eruptive MVs, $4.22 \times 10^9\ m^3\ yr^{-1}$ (3.1 Tg yr^{-1}) of gas may be emitted during eruptions onshore and in shallow water offshore. Similar calculations for 5,000 deep-water MVs suggest that ~77 eruptions occur every year on continental slopes and abyssal plains, and $19.2 \times 10^9\ m^3\ yr^{-1}$ (14 Tg yr^{-1}) of gas may be emitted. However, this estimate is uncertain because there are few measurements of eruptive activity of MVs in deep water.

4. Discussion and Conclusions

[14] Global gas flux from MVs is estimated to be ~15.9 Tg yr^{-1} during quiescent periods, and ~17.1 Tg yr^{-1} during eruptive periods. The uncertainties associated with the data (see above), poorly constrained numbers of deep-water MVs, and the lack of measurements of gas flux during eruptive periods suggest that results are quite approximate. However, our study implies that the global gas flux from MVs to the ocean and atmosphere (estimated here at ~33 Tg yr^{-1} as the sum of emissions during quiescent and eruptive periods) may significantly exceed gas emission from some other natural sources (e.g., termites and natural animals) and some anthropogenic sources (e.g., domestic sewage and animal waste) [*Crutzen and Lelieveld*, 2001]. Furthermore, our estimate is two-four orders of magnitude greater than the previous estimate of global gas flux from MVs [0.08–1.41 Tg yr^{-1}, *Kopf*, 2002].

[15] Onshore MVs emit CH_4 and CO_2 greenhouse gases directly to the atmosphere. In the case of shallow water MVs (<75 m), solution or microbial oxidation of vent gas is not quantitatively significant and much of the gas enters the

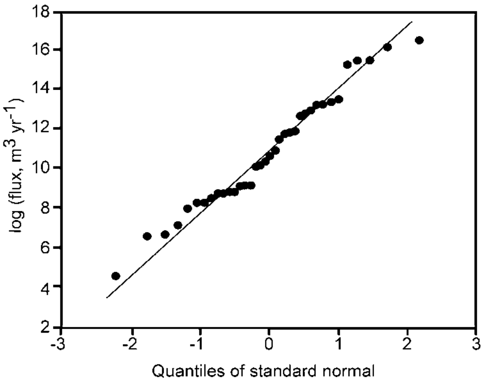

Figure 4. Normal probability plot for gas flux data from Table 1. The approximate straight-line fit indicates underlying normality.

atmosphere [*Judd et al.*, 2002]. Some fraction of hydrocarbon gases from MVs is converted to CO_2 as a consequence of combustion during eruptions, but such violent eruptions are rare [*Guliyev and Feizullayev*, 1996]. Onshore and shallow MVs [total number is assumed to be 1,100; *Dimitrov*, 2002] are estimated to contribute $\sim$6 Tg yr^{-1} of gas to the atmosphere during both quiescent and eruptive periods. New mud volcano areas may be discovered on shelves in the future, and the total gas flux may be higher. On the other hand, a portion of vent gases may be oxidized and dissolved even in shallow water [*Judd et al.*, 2002], potentially decreasing the flux to the atmosphere. CH_4 is the major component of gas flux. CH_4 from MVs appears a minor atmospheric source ($\sim$1% of total sources). However, MVs may contribute $\sim$9% of the missing fossil CH_4 to the modern CH_4 budget, and $\sim$12% to the preindustrial budget. Thus, budgets of atmospheric CH_4 sources and sinks may be improved by inclusion of CH_4 from MVs. Moreover, CH_4 and CO_2 are important greenhouse gases, and climate change models may be improved by inclusion of gas flux from MVs.

[16] It is estimated here that large, although uncertain, volumes of gases ($\sim$27 Tg yr^{-1}) are emitted from deep-water MVs to the water column. It appears that the contribution of deep-water MVs to the atmospheric sources budget may not be significant. In sediment, some fraction of hydrocarbon gases from MVs is sequestered as gas hydrate, is oxidized by bacteria and archaea, and ultimately is sequestered as authigenic carbonate rock [*Judd et al.*, 2002]. In a thick water column, hydrocarbon gases often may be lost to solution and biodegradation [*Judd et al.*, 2002]. However, large gas bubbles from high flux vent sites in deep water transfer to the atmosphere if lined with crude oil [*Sassen et al.*, 2001].

[17] Although not a major source of atmospheric CH_4, gas flux from deep-water MVs may contribute to the oceanic CH_4 and carbon budgets. The current view is that seepage on the continental shelves [18–48 Tg yr^{-1}; *Hornafius et al.*, 1999] accounts for the bulk of global CH_4 flux from the seafloor [30–50 Tg yr^{-1}; *Kvenvolden et al.*, 2001]. Our results suggest that this may not be a reasonable estimate since only deep-water MVs may emit $\sim$27 Tg yr^{-1} of gas. In addition to MVs, fault and salt-related seeps and vents occur on continental slopes and abyssal plains [*Judd et al.*, 2002]. Hydrocarbon seepage from deep-water areas of continental margins may be significantly underestimated. Seeps and vents emit large volumes of ^{13}C-depleted gases, affecting the mass and the isotopic composition of the oceanic carbon pool to an unknown extent.

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