Methane emission from mud volcanoes in the Eastern Azerbaijan

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ABSTRACT

Methane (CH_4) flux to the atmosphere was measured from gas vents and, for the first time, from soil microseepage at four quiescent mud volcanoes (MVs) and one everlasting fire in the Eastern Azerbaijan. MVs show different activity of venting craters, gryphons and bubbling pools, with CH_4 fluxes ranging from less than one to hundreds tons per year. Microseepage CH_4 flux is generally on the order of hundreds mg m^{-2} d^{-1}, even far away from the active centers of MVs. The CH_4 flux near the everlasting fires (on the order of 10^5 mg m^{-2} d^{-1}) represents the highest natural CH_4 flux from soil ever measured. The specific CH_4 flux is between 10^2 and 10^3 t km^2 y^{-1}, and is similar to specific flux at other MVs in Europe. At least 1400 tons of CH_4 per year are released from the investigated areas. It is conservatively estimated that all onshore MVs of Azerbaijan may emit into the atmosphere around 0.3-0.9 million tons of CH_4 per year, only during quiescent activity. The new data support recent estimates of global CH_4 emission from MVs and confirm the importance of geologic sources of greenhouse CH_4, although they are not yet considered in the budgets of atmospheric CH_4 sources and sinks.

INTRODUCTION

Mud volcanoes (MVs) have been extensively studied in the last decades, especially for their geodynamic implications and impact on petroleum industry (Milkov, 2000; Kopf, 2002). However, only in the last two-three years the great significance of MVs as natural sources of methane (CH_4) in the atmosphere has been recognized (Dimitrov, 2002; Milkov et al., 2003), mainly as a result of new offshore observations (Milkov, 2000; Holland et al., 2003) and first detailed onshore gas flux measurements (Etiope and Klusman, 2002; Etiope et al. 2002; 2003; 2004).

More than 900 MVs exist on lands and more than 300 on the ocean shelves (Etiope and Milkov, 2004) mainly within the petroliferous basins. More than 1000 MVs may occur in deep-water areas (Milkov, 2000). Recent estimates suggest that onshore and shallow offshore MVs release to the atmosphere at least 6-9 Mt (million tons) of CH_4 per year (Etiope and Milkov, 2004), an output similar to those attributed to gas hydrates and oceans (IPCC, 2001). However, this estimate suffers from the lack of detailed data from Azerbaijan, which is known as the "World capital of mud volcanism". Azerbaijan hosts the world's largest MVs and has the densest MV population, with about 200 structures onshore and 160 in the Caspian Sea. Limited gas flux data for several onshore MVs were presented in the Former Soviet Union literature (Pankov, 1940; Dadashev, 1963; Jakubov et al., 1971) but the methodology of measurements and the significance of the gas flux were not clearly described. Moreover, previous gas flux data do not include the pervasive leakage of gas from soil, known as microseepage (Klusman and Jakel, 1998; Klusman et al., 2000; Etiope and Klusman, 2002), which was recently recognised as an important component of degassing at European MVs (Etiope et al., 2002; 2003; 2004). We report here, for the first time, CH_4 flux data including microseepage from four large MVs (Lokbatan, Kechaldag, Dashgil and Bakhar) and one everlasting fire (Yanardag) in the eastern Azerbaijan. Our results provide critical data that cover the main gap in global data set of CH_4 flux from geologic sources (Morner and Etiope, 2002; Etiope and Klusman, 2002).

GEOLOGICAL SETTING AND DESCRIPTION OF MUD VOLCANOES

MVs of Azerbaijan are located mainly on land in the eastern part of the country and within the central and southern sectors of the Caspian Sea. The investigated MVs occur in the Gobustan region and Absheron peninsula (Fig. 1). This area corresponds to the western flank of the South Caspian depression (Abrams and Narimanov, 1997), and most MVs overlay the faulted and hydrocarbon-bearing anticlines. Geological and morphological details are reported in the numerous works of the Geology Institute of Azerbaijan (e.g., Jakubov et al., 1971; Guliyev and Feizullayev, 1997; Aliyev et al, 2002). According to Aliyev et al. (2002), Azerbaijan MVs may be classified into two main groups: MVs with intense and continuous gryphon-salse activity and weak eruptions, such as the Dashgil MV, and MVs with low or absent venting activity and higher eruptive potential, such as the Lokbatan, Kechaldag and Bakhar MVs.

Gas emitted by MVs of Gobustan and Absheron peninsula is composed mainly of CH_4 (90-99.8 vol.%) and CO_2 (0.2-8.6 vol.%) (Aliyev et al. 2002). Relative abundance of C2+ hydrocarbon gases, carbon isotopic composition of CH_4 (äC^{13} values are between –44 and -37 ‰ relative to the PDB standard; Dadashev and Guliyev, 1989), and associated oil seepage suggest the thermogenic origin of emitted gases. It has been suggested that hydrocarbon gases were generated in Miocene to Pliocene deposits from terrestrial and marine kerogene with abundant humic component and limited bacterial contribution (Abrams and Narimanov, 1997).

The Dashgil and Bakhar MVs are located on the opposite pereclines of the same NE-SW stretched anticline (Fig. 1), about 66 km SW from Baku. The Dashgil MV has more than 60 gryphons (Fig. 2) and large pools and its structural and morphological features are described in detail by Hovland et al. (1997). It erupted at least six times since 1882 (Aliyev et al., 2002).

The Bakhar MV is located on the eastern tip of the Alyat cape (Zotov mountain). The MV has ~30 gryphons and salses that are clustered in two groups separated by ~600-700 m. The mud cones are 8-10 m high and have base diameters of 25-30 m. The MV hosts one salse (pool) that has a diameter ~100 m. The Bakhar MV erupted at least 11 times since 1853 (Aliyev et al., 2002). During the last eruption (October 1992), a large amount of mud was ejected and a flame column rose into the air for about 150 m, brightly illuminating the near villages (Aliyev et al., 2002).

The Lokbatan MV, located on an anticline about 60 km SW of Baku city, is the most frequently erupting MV in Azerbaijan. It erupted at least 23 times since 1829 (Aliyev et al., 2002). The largest volumes of the mud breccia are ejected on the western side of the MV field, where several domes resulted from the last eruptions in October 2001. Mud breccia contains rocks fragments from the Upper Cretaceous-Tertiary sequences. The geologic structure is build up by the deposits of Absheron (Quaternary) suite, Akchagyl (Upper Pliocene) suite, and the hydrocarbon-bearing Lower Pliocene productive series.

The Kechaldag MV is located ~22 km NE from Baku, within the bounds of Western Absheron tectonic belt and in the Southern-Eastern part of the Binagadi-Kechaldag structure over Paleogene-Miocene rocks. This MV is not widely described in the literature because it was assumed to be dormant. However, the MV suddenly erupted in 2000 and ejected large volumes of mud breccia that flowed into pond supplying water to nearby cities and villages (Aliyev et al., 2002).

Yanardag village hosts one of the most fascinating everlasting fires in Azerbaijan, that served as traditional symbols of the ancient Zoroastrian religion. Intensive flames, up to 1 m high, develop for ~15 m along the base of a 2-4 m high and ~200 m long tectonic scarp (Fig. 4). This fault is a part of the huge Balakhan-Fatmai structure on the Absheron peninsula.

METHODOLOGY

The output of CH_4 from vents was measured by inverted funnel (Etiope et al., 2002) and single tube flow-meters (accuracy 5%) in 24 accessible gryphons and bubbling pools. In addition, for vents not accessible for direct measurements, the output was estimated based on visual observations and a theoretical relationship between the size of bubbles, the frequency of their appearance from a vent and the CH_4 flux (Etiope et al, 2004). Microseepage and low-flux venting was measured by the closed-chamber method, similar to the Crill and Savage systems (Norman et al., 1997). Gas accumulated in the chambers was collected two or three times into syringes at time intervals varying from 10 seconds to 10 minutes after the deployment of the chamber. Methane concentration was analyzed in duplicate by gas chromatograph with FID detector (Autofim II, Telegan; detection limit 0.1 ppm, accuracy 4-5%). This technique has been widely employed in microseepage studies in petroliferous areas (e.g., Klusman et al, 2000; Etiope et al, 2002) and in the study of biological fluxes of CH_4 in wetlands and drylands (e.g., Scharffe et al., 1990). In total 87 microseepage measurements were homogeneously distributed throughout the MV areas, covering all sectors with different vent density and soil conditions. For example in the Dashgil MV microseepage measurements were carried out following homogeneous sampling grids throughout the MV plateau within the “gryphon zone”, around the largest bubbling pool, in the “fore zone” and “back zone” (Fig.2). The total MV output is thus derived by summing up the outputs from all vents and sectors. Similar strategies were extensively used to derive the gas output from MV and geothermal areas (e.g., Etiope et al., 2004; Chiodini et al., 1998). MV areas were derived by GPS (Garmin eTrex Legend) and Azerbaijan 1:100,000 topographic maps (University of California Library; www.lib.berkeley.edu).

RESULTS

Table 1 shows the mean and standard deviation of the microseepage and vent flux measurements performed in the different MV sectors; table 2 summarises the flux data and the total output for the measured and total MV areas, as described hereafter.

The Lokbatan MV did not have active gryphons or other gas vents during the survey. The microseepage ranges from 110 to 603 mg m^{-2} d^{-1} (mean of 307 mg m^{-2} d^{-1}) over an area of 0.1 km^2 and it is on the order of 10^3-10^5 mg m^{-2} d^{-1} near small (0.5 m high) natural flames located in the central crater. The high degassing zone has area ~60 m^2. The microseepage rapidly decreases outside of the flame area and is on the order of 10^2 mg m^{-2} d^{-1} at a distance 40-50 m away from the flames. This microseepage level is also typical for soils near the boundaries of the Lokbatan MV, outside of the recent mud flows.

Four main gryphons are active at the Kechaldag MV, and they produce from 0.3 to 2.5 t/y of CH_4. The rates of microseepage range from 80 to 820 mg m^{-2} d^{-1}. The highest microseepage values are measured along the margin of the central crater bound, where radial faults (outcropping at the surface) developed after the crater erupted and subsequently collapsed in October 2000. Lower microseepage rates are measured at the flanks of the Kechaldag MV. The CH_4 flux remains positive (80-210 mg m^{-2} d^{-1}) even outside of the recent mud flows.

The Dashgil MV is characterized by high venting activity and at the time of measurements had over 60 active gryphons (Figs. 2 and 3) and bubbling pools (salses). Three main different degassing zones were recognised (Fig. 2): 1) fore zone (western side of the MV) with microseepage from 55 to 340 mg m^{-2} d^{-1}, 2) gryphon zone (central axis) with microseepage from 220 to 1469 mg m^{-2} d^{-1}, and 3) the pool zone on the eastern side with microseepage from 840 to 27,200 mg m^{-2} d^{-1}. The measured individual gryphons produce from 0.8 to 12 t/y of CH_4. The largest pool occupies the area ~4500 m^2 (level pool as designated by Hovland et al., 1997) and releases a bubbling plume extending about 10 m^2. Based on photo and video images, we estimated that ~4600 bubble trains occur in the pool producing a total CH_4 output of ~500 t/y.

The Bakhar MV is characterized by the lowest degassing activity among the investigated MVs. Two main groups of gryphons and small salses occur. The measured gryphons produce from 0.1 to 0.5 t/y of CH_4. The microseepage ranges from 65 to 570 mg m^{-2} d^{-1} along the flanks, and from 300 to 720 mg m^{-2} d^{-1} on the summit area near the gryphons.

The microseepage of CH_4 around the Yanardag everlasting fire ranges from 6700 to >560,000 mg m^{-2} d^{-1}. To our knowledge, it represents the highest rate of microseepage from soil ever measured. The only other everlasting fires measured so far (in Sicily and Romania) displayed CH_4 fluxes on the order of 10^4-10^5 mg m^{-2} d^{-1}, but the maximum value was 245000 mg m^{-2} d^{-1} (Andreiasu fire in Eastern Romania; Etiope et al., 2004). At the Yanardag, the greatest flux was measured on the upper side of the fault scarp along which the flames are released (Fig. 4). Microseepage on the order of 10^3 mg m^{-2} d^{-1} occurs at ~30 m from the fire, on the upper side of the study area. The total degassing area is clearly larger than the measured area, and it is very likely that the microseepage is pervasive along the fault zone which is manifested at the surface as a >200 m-long scarp.

DISCUSSION AND CONCLUSIONS

Our surveys confirm that the specific flux from MVs is between 10^2 and 10^3 t km^2 y^{-1}, as previously found in the main European MVs (Etiope et al., 2002; Etiope et al., 2004). Degassing from the active Dashgil MV is relatively high and similar to Paclele Mari MV of Eastern Romania, both in terms of microseepage and vent activity. We define that the Dashgil MV is relatively high-degassing (HDMV). In contrast, the Lokbatan, Kechaldag and Bakhar MVs have relatively low (< 250 t km^2 y^{-1}) specific CH_4 fluxes (i.e., LDMVs), even lower than those measured in Sicily and Romania (Etiope et al., 2002; 2004).

The CH_4 flux values from the vents of the Dashgil MV are in a good agreement with those reported by Pankov (1940), who measured that individual gryphons emit from 0.2 to 9 t/y of CH_4. Hovland et al. (1997) visually estimated a total output of 0.5 t/y of gas for the whole Dashgil MV; this value is clearly erroneous. Venting fluxes from the Bakhar MV are similar to those reported by Dadashev (1963), who estimated that the flux from individual gryphons ranges from 0.015 to 2.1 t/y of gas.

The microseepage data suggest that soil degassing is relatively high and pervasive even far away from the active centers of MVs. Previous studies suggested that microseepage in tectonically and seismically active and hydrocarbon-bearing areas far away from individual MVs (Great Caucasus, Lesser Caucasus, Kura depression) ranges from 8 to about 400 mg m^{-2} d^{-1} (Balakin et al., 1981; Sokolov, 1971). In the areas of Azerbaijan not populated by MVs, the microseepage was estimated to be on the order of a few tens of mg m^{-2} d^{-1} (below 200 mg m^{-2} d^{-1}) (Voitov, 1975). The higher microseepage values were measured over active faults that apparently serve as migration pathways for petroleum fluids. These values are coherent with microseepage rates measured in other non-MV hydrocarbon-bearing areas of the United States (e.g., up to 43 mg m^{-2} d^{-1} in Denver-Julesburg basin and up to 200 mg m^{-2} d^{-1} at Music Mt., Pennsylvania, Etiope and Klusman, 2002 and references therein). Microseepage in Azerbaijan may exceed the microbial consumption in the soil throughout vast areas, and therefore may represent a significant regional source of CH_4 in the atmosphere.

The total CH_4 output from the four investigated MVs and the everlasting fire (total area of 5.9 km^2) is conservatively estimated to be ~1400 t/y; the mean specific flux (excluding fire) is about 450 t km^2 y^{-1} for the area surveyed and 225 t km^2 y^{-1} if extrapolated to the whole MV areas. Based on the regional data (Jakubov et al., 1971), we estimate that the total area covered by onshore MVs in Azerbaijan is between 1200 and 2000 km^2. We further estimate that ~720-1200 km^2 (60% of the total area of MVs) is occupied by the LDMVs, and ~480-800 km^2 (40% of the total area of MVs) is occupied by the HDMVs. Assuming a specific flux of 400-800 t km^2 y^{-1} for the HDMVs and 100-200 t km^2 y^{-1} for the LDMVs, we conservatively estimate that all onshore MVs of Azerbaijan may emit into the atmosphere ~0.3-0.9 Mt of CH_4 per year only during quiescent activity.

A recent estimate suggests that the global CH_4 flux from onshore and shallow offshore MVs during their quiescent activity may be ~2.8-4 Mt/y (Etiope and Milkov, 2003). Azerbaijan MVs may therefore contribute ~20% of this flux. However, it appears that the Azerbaijan MVs may be characterized by greater eruptive activity relative to other MVs in the world (Aliyev et al., 2002). In their global estimation of gas flux from mud volcanoes, Milkov et al. (2003) concluded that the global eruptive degassing may be approximately equal to the global quiescent degassing. In contrast, Dimitrov (2002) suggests that gas flux from quiescent periods is significantly (by a factor of up to 30) less than the gas flux during eruptions. Detailed studies and measurements of gas flux during eruption, and direct measurements of gas flux from submarine mud volcanoes appear to be critical to further constrain the global gas flux from MVs.

According to the available estimates, onshore and shallow offshore MVs contribute from 15 to 25% to the global geologic CH_4 flux, which is conservatively estimated to be 35-45 Mt y^{-1} (Etiope and Milkov, 2004). Our direct measurements of CH_4 flux in the Eastern Azerbaijan confirm that MVs are important sources of CH_4 in the atmosphere. It appears that the significance of geological sources of CH_4 may be even greater because direct data for several still underestimated components of the CH_4 flux (e.g., MV eruptions, offshore MVs, microseepage in sedimentary basins, geothermal flux, etc.; Etiope and Klusman, 2002) are yet to be collected. However, it is already clear that the geologic sources of CH_4 cannot be longer omitted from the atmospheric budgets because they are comparable to or higher than several other sources and sinks (e.g., termites, biomass burning, soil uptake) officially considered in the IPCC tables (IPCC, 2001).

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TABLE 1. DESCRIPTIVE STATISTICS OF VENTING AND MICROSEEPAGE FLUX DATA

MV	Sector	Vents*			Microseepage		
		N.	Mean	St.dev.	N.	Mean	St.dev.
			($t\ y^{-1}$)			($mg\ m^{-2}\ d^{-1}$)	
Lokbatan	Flanks				11	307	157
	Crater		N.A.		3	2800	872
	Fire zone				3	366,000	118,000
Kechaldag	Flanks				6	163	80
	Crater (central axis)	4	1.2	1	10	417	222
Dashgil	Fore zone				11	188	100
	Gryphons zone	15	4.3	3.1	11	593	364
	Back zone				4	674	107
	Pool zone				7	9450	11100
Bakhar	Flanks				9	171	159
	Vents zone	5	0.6	0.4	3	490	212
Yanardag	Fire zone		N.A.		5	247,600	198,000
	grassland				4	9445	4150

* Vents measured directly. For all the other vents (about 70 at Dashgil and Bakhar) flux was derived by theoretical relationship between bubble size, bursting frequency and gas flux (Etiope et al, 2004).
N.A. Not Applicable

TABLE 2. SUMMARY OF MUD VOLCANO FLUX DATA

MV	Measured and total MV area (km^2)	Mean microseepage ($mg\ m^{-2}\ d^{-1}$)	Microseepage output ($t\ y^{-1}$)	Vent output ($t\ y^{-1}$)	Total output ($t\ y^{-1}$)	Specific flux ($t\ km^{-2}\ y^{-1}$)
Lokbatan	0.1	307 *	8 † + 11.2	N.A.	19.2	192
	2.98		139		342	115
Kechaldag	0.05	320	5.8	4	9.8	196
	0.77		90		94	122
Dashgil	0.6	410§	104	623	727	1200
	1.4		220		843	600
Bakhar	0.05	250	5.5	8.4	14	230
	0.7		36.5		45	64
Yanardag	0.001	63200	27	N.A.	40	40000
	> 0.01?		233		68	6800
Total	0.8	322 ‡	161.5	635	810	454 ‡
	5.86		718		1392	225 ‡

* Excluding fire
† Fire zone
§ Excluding pool zone
‡ Excluding Yanardag fire
N.A. Not Applicable

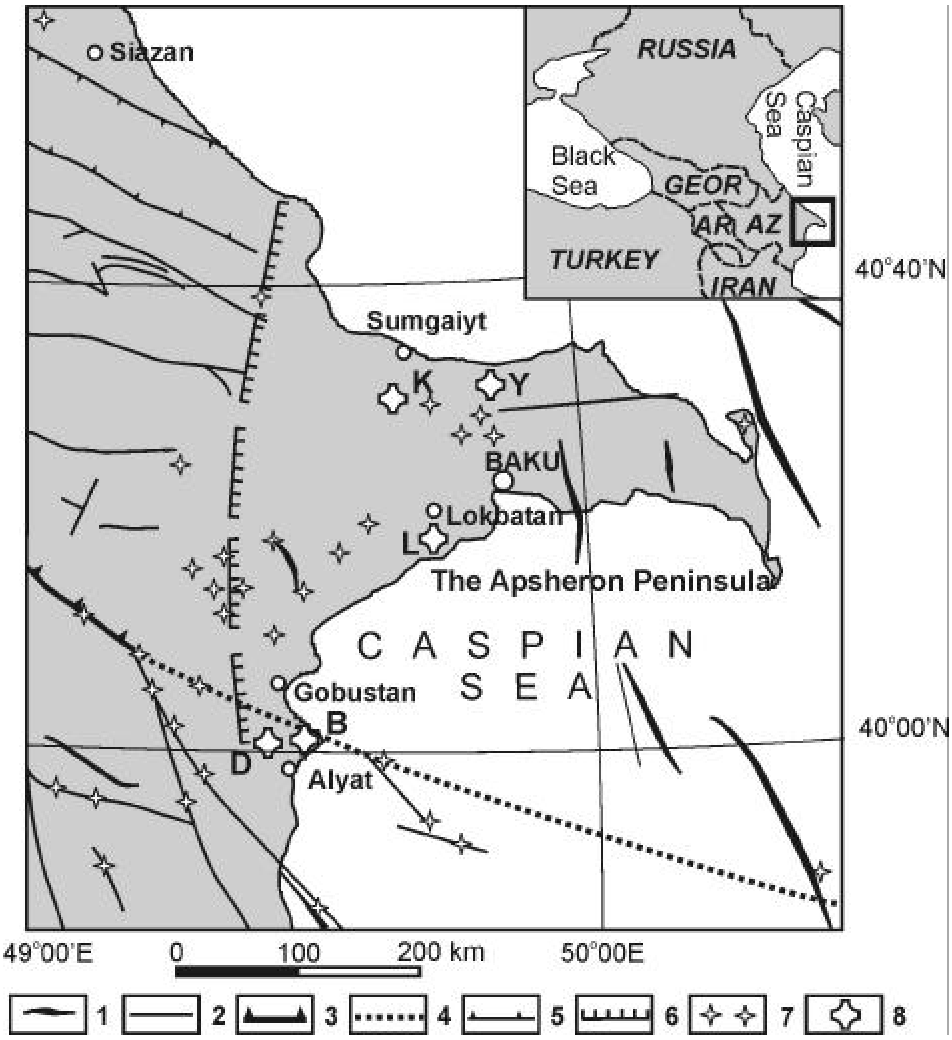
FIGURE CAPTIONS

Figure 1. Structural map of Eastern Azerbaijan and mud volcano locations (modified after Kadirov, 2000). 1– axes of large anticlinal folds; 2 – normal faults, hundreds to thousands of meters of amplitude; 3 – crustal faults manifested at the surface by thrusts and overfaults; 4 – buried crustal faults; 5 – thrusts and overfaults with thousands of meters of amplitude; 6 – buried flexure; 7 – mud volcanoes; 8 – investigated sites: B – Bakhar, D – Dashgil, K – Kechaldag, L – Lokbatan, Y – Yanardag.

Figure 2. Gas flux sampling sectors on the plateau of the Dashgil mud volcano.

Figure 3. Gas eruption from a gryphon of Dashgil. Crater is 3 m wide and the gas-mud bubble has a diameter of about 70 cm. This type of eruption occurs every 5-10 minutes.

Figure 4. Gas flux sampling at the Yanardag everlasting fire.



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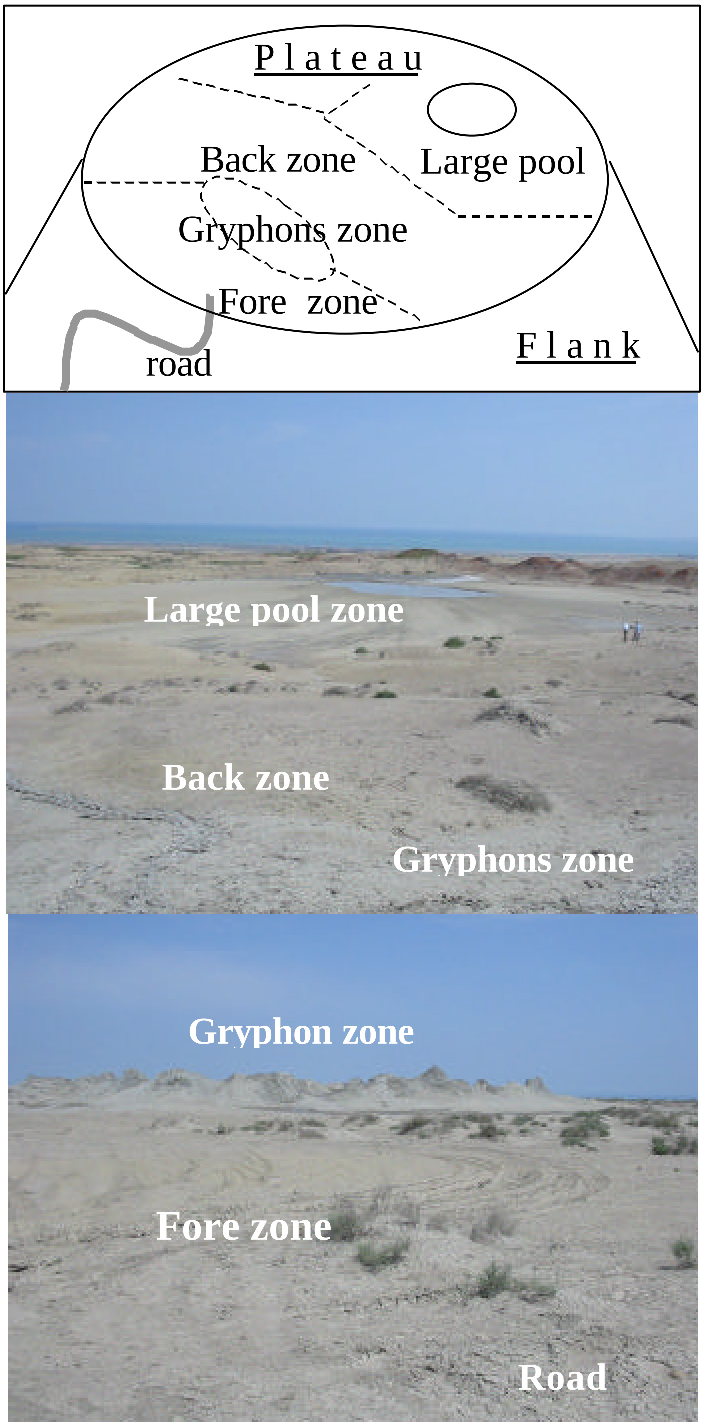

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