MUD VOLCANOES FROM BANAT REGION (ROMANIA): THEIR ENVIRONMENT IMPACT

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Abstract: An assessment of the natural manifestations of fluids (gases, water) and solid phases (fine-grained sediments) from deep sediment layers of Banat area, in mud volcanoes (MVs) category is given. The total occurrence area of recent or ancient mud volcanoes in Fibiş Valley is about 0.9 km^2. All the craters occur along the Fibiş Valley, along a NNE-SSW direction, at an average altitude of 121m. They occur along an active fault system, acting as conduits for the migration of fluids. The main crater is Forocici Crater (FC), where the gas emissions include CO_2, CH_4, CO and H_2S. Seven secondary craters are recorded, and two are temporarily active, having several seepages with gas and water eruptions. Fluid flows of mud volcanoes of the Fibiş Valley have an important impact on atmospheric gas budget. The craters shape is generally irregular developing at soil level, with the exception of crater C2 (sub-circular shape) and Forocici Crater which has a gryphon type cone. Fe dominates the water samples collected at FC pool. Low concentrations of Cr, Ni and Zn were observed. Analyses of heavy metal concentrations of soil show wide variations. All concentrations of metal are low and below the maximum allowable limit. However, some samples show high concentrations of metals, such as those containing Fe, Pb and Cu. All examined soil and mud samples present a higher content of Fe (2144 mg/kg - 2585 mg/kg). Generally, heavy metal concentrations in soil do not exceed the allowable maximum values, except Pb (27.5 - 38.0 mg/kg d.s.), toxic for animals grazing growing in Fabiş Valley. *Salix cinerea* L. which grows on the soil at the limit between the crater and Forocici Crater cone contains Fe (55.50 -80.00 mg/kg d.s.), Cu (2.74 - 4.50 mg/kg d.s), Ni (0.15 mg/kg d.s), Pb (1.35 mg/kg d.s) and Zn (42.20 mg/kg d.s). The mineralized fluids brought from deep layers control heavy metals concentration. From the substratum of Forocici Crater, we identified six species of diatoms (*Gomphonema minutum* (Agardh) Agardh, *Pinnularia obscura* Krasske, *Geissleria acceptata* (Hustedt) Lange-Bertalot & Metzeltin, *Hippodonta linearis* (Østrup) Lange-Bertalot, Metzeltin & Witkowski, *Stauroneis anceps* Ehrenberg *and Nitzschia solita* Hustedt). These data sets represent a contribution to the understanding of mud volcanoes' output, of their influence on the environment and to the understanding of deep and shallow fluids in mud volcano systems.

Key words: mud volcanoes, bubbling pools, carbon dioxide, heavy metals, soil, plants, diatoms

1. INTRODUCTION

Mud volcanoes (MVs) develop in many regions that have similar geological settings (active fault systems). They are a natural phenomenon that reflect regional geological processes and the most important processes related to the natural seepage in earth's surface (e.g. Mazurenko & Soloviev, 2003; Yusifov & Rabinowitz, 2004; Uruioc et al., 2007). Geologists describe the mud volcanoes as unstables, and they are still arguing about their precise genesis.

The origin, subsurface structures, backscatter characteristics, mechanism of formation, paleo-activity, morphology, distribution and the consequences on the surrounding deep biological and geological environments have been described in many papers (e.g. Guliyev & Feizullayev, 1997; Kopf, 2002; Milkov et al., 2004; Mastalerz et al., 2007; Etiope & Milkov, 2004; Etiope et al., 2004a,b; Etiope et al., 2009a and Etiope et al., 2010). Some hypotheses have been put forward concerning the source, age, and emplacement of the extruded mud, as

well as the various mechanisms leading to its extrusion (e.g. Huguen et al., 2005). According to Higgins & Saunder (1974), Fowler et al., (2000), Kopf & Behrmann (2000), Milkov (2000) and Kopf (2002), tectonic processes form mud volcanoes, such as overpressure buildup in compressional settings, or the maturation and degassing of rapidly buried organic-rich sediments. The mud and fluids have a deep origin, but they are sometimes stored in intermediate-depth mud chambers (Planke et al., 2003). The mud volcanoes are a consequence of the transfer of semi-liquid material and gas from depth towards the surface (Deville & Guerlais, 2009). Therefore, they are an important source of information about subsurface sediments and conditions (e.g. Yusifov & Rabinowitz, 2004). The mud volcanoes are usually cone-shaped edifices of variable dimensions from one to two meters, to several hundred meters in height, constructed by the emission of argillaceous material transported in the same time with water and gases (Dimitrov, 2002; Kopf, 2002; Accaino et al., 2007). These features are recorded in different tectonic settings. Eruptions of mud volcanoes can be explosive or effusive, and may show a variable degree of activity. The identification of their characteristics (nature, geometry and size) may be difficult for historical eruptions (Bonini, 2009a). Also, MVs typically include gas phases, composed primarily of methane and carbon dioxide, which may play a role in driving eruptions (Manga et al., 2009). The normal activity of mud volcanoes consists of gradual and progressive outflows of liquefied sediments (Accaino et al., 2007). MVs activity manifested through mud, water and gas eruptions determines at the surface of terrestrial crust specific landscapes known in the specialty literature under the name of gryphons (mud volcanoes with cone height lower than 3.00 meters and the external slope with small inclination, that permits mud exterior seepage), mud cones (cone height smaller than 10.00 meters, through which mud and rock fragments are expelled), salses (mud volcanoes who's crater fills up with water forming a lake, through which gas emissions break in), springs (cones smaller than 0.50 meters, through which water gets out under pressure and dominates over gases) (Hovland et al., 1997; Guliyev & Feizullyev, 1997; Aliyev et al., 2002). Liberated gases can be a mixture of hydrocarbons (70-90% CH_4) or CO_2, accompanied by small quantities of N_2, H_2S, and NH_3. Also, noble gases can occur such as He, Ar, Rn, Ne and Kr (Lynch & Hudnut, 2008). The significance of mud volcanoes as natural sources of atmospheric methane (CH_4) is already recognized (Etiope, 2009; Etiope et al., 2002, 2004a,b, 2006, 2009a; Etiope & Milkov, 2004;

Marinaro et al., 2006; Baciu et al., 2007). They contribute to the global atmospheric methane budget and have an impact on the earth's climate (Kopf, 2003).

Mud pots are of interest due to their uncommon gaseous emissions and to their potential role in influencing the atmosphere and climate (Lynch & Hudnut, 2008). MVs are not only a source of direct information through the fluid material carried by water, gases, and mineral suspense brought from profound areas of the terrestrial crust, but also a touring area (Uruioc et al., 2011).

In Romania, MVs and dry macro-seeps were identified in the Carpathian Foredeep (where four mud volcano fields are active: Pâclele Mari, Pâclele Mici, Fierbători and Beciu), in the Transylvanian Depression (Sărmăşel, Homorod, Bazna), as well as on the Moldavian Platform (Baciu et al., 2007). In the East of Romania, a research over MVs from Berca and Andreiaşu marks out CH_4, CO_2, and N_2 emissions with crustal origin (Etiope et al., 2004a). In southwest Romania, MVs studies from Banat region were undertaken by Bizerea (1965) and Uruioc et al., (2007, 2011).

In this paper we overview the main features of the mud volcanoes in Fibiş Valley expand the data set regarding heavy metals content from water, soil and vegetation, investigated for the first time in 2005 (Uruioc et al., 2007, 2011). Also, we show a model for the migration pathways of fluids into the Forocici Crater. The aim of this paper is the biodiversity of diatoms flora found in the Forocici pool.

2. GEOTECTONIC SETTING AND SITE CHARACTERIZATION

2.1. Regional geology

The structures of southwestern part of Romania include the complex tectonic contact area between the Carpathian orogen (South Carpathians) and the subsidence structures of the southeastern border of Pannonian Depression (Oros, 2007; Grecea, 2009). This region corresponds to the main regional morphological unit, the Banat Plain of Romania. The geology of this zone is very complex: two major NW-SE grabens prevail in the region: Sânnicolau Mare (called in this paper SMG) and Lugoj-Caransebeş (LCG). Between these structures, an uplifted tectonic block extends from Buziaş to Hungary, called Battonya-Buziaş horst (BBH). In the South, the Caraş graben (CSG) is developed (Oros, 1991, 2007) (Fig. 1). This central block has been divided through active fault systems in many smaller blocks, which had a differentiated evolution in time. Banat Plain, which is

part of the studied area, is composed of Neogene-Quaternary sedimentary rocks whose thickness surpasses 5000 m (Oros, 1991). Sedimentary rocks have a basement composed of pre-Mesozoic metamorphic rocks and Mesozoic ophiolites.

According to Borugă & Airinei (1981), this unit is pierced by eruptions such as Pliocene basalts (Lucareț) of subcrustal origin (Hertz et al., 1973, cited by Visarion & Săndulescu, 1979).

Plutonic banatitics rocks (Bocşa, Surduc, Găvojdia, Ivanda, Timişoara, Utvin, Bazoş, Biled, and Calacea) pierce the basement (Visarion & Săndulescu, 1979) (Fig. 1). Their occurrence is controlled by the existence of some deep faults, which generate epicenters of normal earthquakes along the same direction (Oros, 1991) (Fig. 1). The historical earthquake epicenters distribution reveals a very good spatial with the principal faults from the region (Oros, 2007; Grecea, 2009).

The epicenters are scattered all over in the Banat Plain of Romania, but they cluster into smaller source zones forming several groups of active areas: Arad-Sânnicolau Mare, Vinga-Variaş, Maşloc-Buziaş, Jimbolia-Sânmihai, Şag-Parţa, Moşniţa-Recaş, Banloc-Voiteg and small areas along the border of Romania with Serbia and Hungary (Grecea, 2009). The southwestern territory of Romania is the most important region of the country regarding the seismic hazard induced by crustal earthquakes sources (Oros et al., 2008). The profound and active character of these faults (e.g. F1 fault and F2 fault) separating tectonic blocks is confirmed by earthquakes, by thermal and mineral water sources (Biled, Calacea, Timişoara, Buziaş, Pişchia, Lipova, Ivanda), by the mofettes of Buziaş, and by He, Rn, Ar occurrence and of other gases with crustal origin (Oros, 1991) (Fig.1). For a long time has been admitted the connection between the location of mud volcanoes and faults (e.g. Manga et al., 2009).
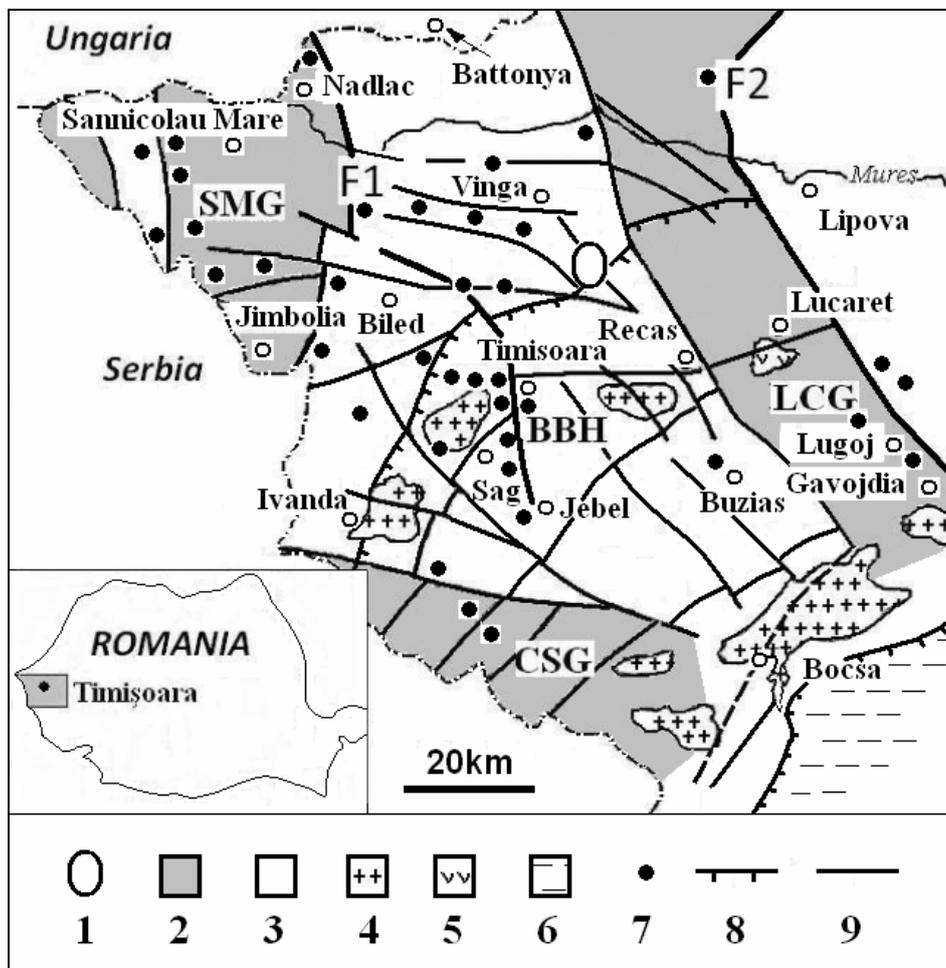

Figure 1. General seismo-tectonic map of Banat Plain showing the position of the Mud volcanoes field (modified after Visarion & Săndulescu, 1979 and Oros, 1991): 1-Mud volcanoes field; 2 - Graben; 3 - Horst; 4 - Plutonic banatitics rocks; 5 - Pliocene basalts; 6 – Getic Nappe; 7 – Epicentres; 8 – Overthrust nappe; 9 – Faults; SMG – Sânnicolau Mare Graben; LCG – Lugoj-Caransebeş Graben; CSG – Caraş Graben; BBH – Battonya-Buziaş Horst; F1 – Nădlac-Timişoara-Jebel fault; F2 – Zarand-Lipova-Lugoj fault.

According to Lynch & Hudnut (2008), the location of mud volcanoes is sometimes used to infer the presence of active fault systems. The majority of mud volcanoes are related to the regional deep fault systems (Yusifov & Rabinowitz, 2004).

2.2. Description of mud volcano field

In the geological literature, the term "mud volcano" is used for geological structures formed by the emissions of natural gas controlled by geological settings, the majority of them being localized in areas of recent tectonic activity. Thus, Bonini (2009b) studied the relationships between seismicity of a carbon dioxide - driven mud volcano, located in the Northern Apennines of Italy (having 94.73% CO_2), whereas Etiope & Martinelli (2009) assert that the term "mud volcano" should be used for a region, where the gas is predominantly composed of methane (CH_4) and only, in few special cases, this gas can be dominated by CO_2 or N_2.
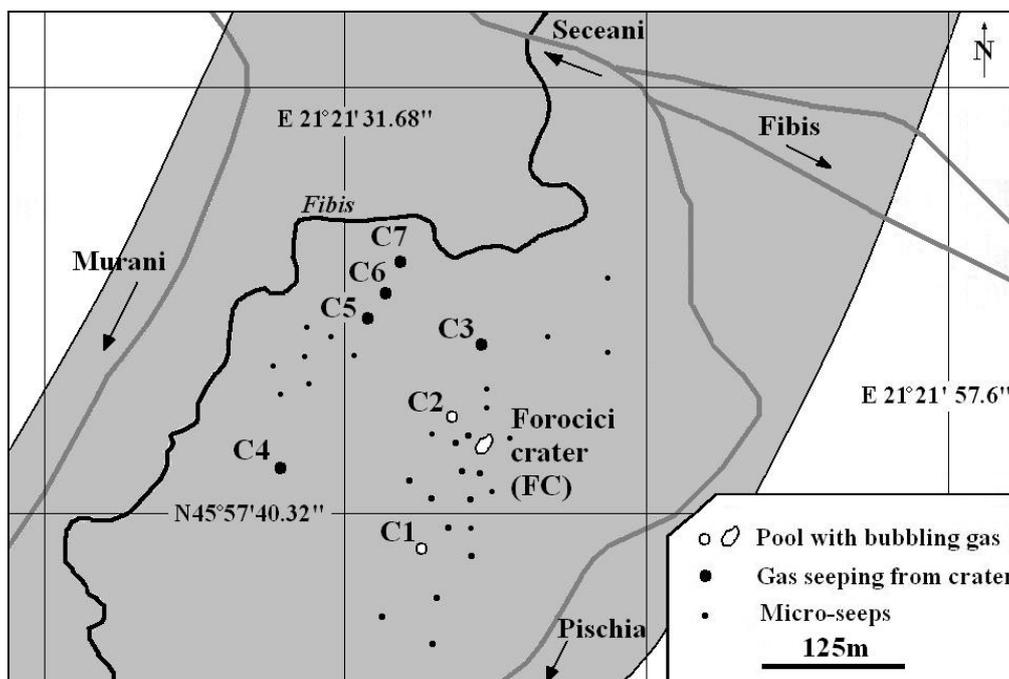

Figure 2. Mud volcanoes distribution in Fibiş Valley

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In the absence of specific analyses, we are going to include the natural manifestations of fluids (gas, water) and solid phases (fine-grained sediments) from deep sediments of Banat region to the mud volcanoes (MVs) category, as Bizerea called them in 1965.

In Fibiş Valley from Vinga Plain (part of Banat Plain) at the feet of the Luda-Bara Hill (187.7m altitude), occurs a mud volcano field which is less investigated (Fig. 2). These mud volcanoes have variable geometry and size, and they are formed as a result of the emissions of argillaceous material and fluids (water and gas) (Kopf, 2002). The different morphology (size and shape) of the mud volcanoes of the Fibiş Valley field may be related to different stages in their development. The activity of the mud volcanoes generally consists of a continuous gas bubbling and water overflow from salses (Baciu et al., 2007) and of small eruptions of gas from vents.

The most important mud volcano is Forocici, assigned by Bizerea (1965) to the gryphon type, ["small mud volcanoes on land (1–3m tall) are usually called mud cones or gryphons" (Lynch & Hudnut, 2008)]. This volcano has a sub-circular crater (diameter of 9/11 m and depth of 1.5 - 1.8 m), with a cone height lower than 0.6 m (Fig. 3, 4A, 4C and 5). The cone has mud layers, and its slope is variable, from almost horizontal to a maximum of 30-40°, allowing water with minor amounts of fine-grained sediments to seep outside.
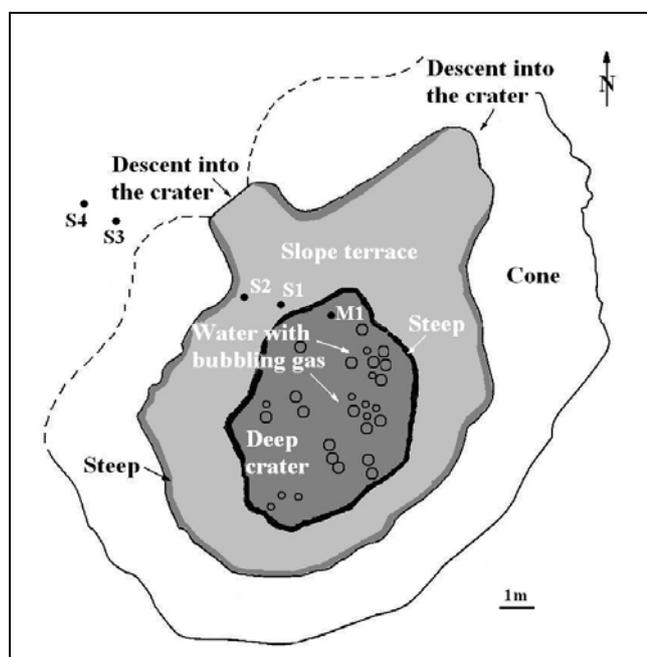

Figure 3. Schematic representation of Forocici Crater (FC). The sketch was modified after Bizerea (1965). M1 - sample mud, S1, S2- soil samples on slope terrace, S3, S4- soil sample on Fibiş Valley

This activity takes place during spring or autumn, when the aquiferous level is high and the crater brims up with water. When water level decreases with at least one meter, the internal structure of the crater can be observed (Fig. 4C).

Thus, the cone shows a slope that continues with a sloping terrace, which temporarily develops *Polygonum aviculare* L. An inferior, sub-circular crater follows, identified as a deep crater. The general shape of Forocici Crater (FC) results from long-term activity, probably more violent in the past. During the first stage of the Forocici mud volcano evolution can be identified a product of violent eruption manifesting itself by creating a large crater. We suggest that a huge quantity of mud and parts of rocks was deposited along the sides. During times of fluid migration, the crater could be filled with mud with different viscosities, when it could generate a gryphon-like crater. Because the gryphon crater was filled up with water - pools with gas seeps - we also adopted the terminology salse and bubble pool from Baciu et al. (2007), Bonini (2009 a) and Mazzini et al. (2009). The activity of the seething cauldron Forocici consists of gas eruptions, water and turbulent gas bubbling. Visible concentric wrinkles on the pool surface suggest that many conduits occur, able to penetrate through sedimentary layers (Fig. 4E).

Gas bubble flows are prominent above each vent, but it is very likely that gas diffuses also through the ground around the each pool (Deville & Guerlais, 2009) or in crater without water. The turbulent bubbling of the gas causes mud to be carried with water for deposition, around the edges of the gryphon. During dry periods, when water disappears completely, in Forocici Crater interior, activity continues through secondary decimetric craters with spherical mud caps (Uruioc et al., 2011).

Also seven secondary craters (diameters between 4/4 m and 1/1 m) occur, including two temporarily active, with several seepages (centimetric and millimetric in sizes) with gas and water eruptions (Fig. 2). The crater C1 (diameters 0.8/0.7m, 0.45m deep) was formed later and, irregular in shape. The sub-circular crater C2 (diameters 4.2/3.4 m, 0.45 deep) is salse type (gas-bubbling water pool) and it was formed in 1962 (Bizerea, 1965) (Figs. 2 and 4). The mud volcano shape is generally irregular developing at soil level (Crater C1, C3, C4, C5, C6, C7) with the exception of crater C2 (sub-circular shape) and FC which has a gryphon type cone (Bizerea 1965).

The shape mud volcanoes of the Fibiş Valley, differs of that of the mud volcanoes from Carpathian Foredeep [conical edifices up to 10 m high, with an apical vent (Baciu et al., 2007)], being more similar with the ones in Transylvanian Depression [bubbling pools (Baciu et al., 2007)].

Figure 4. Photos of gas and water seeps: Forocici Crater with high level (A); C2 crater (B); Forocici Crater with low level (C); new crater with irregular form (D); visible concentric wrinkles on the pool surface of Forocici Crater (E); water with bubble gas in first phase of one crater (F).

The mud volcano activity can be best observed during rainfall intervals, when the gas under pressure, along with the ground and meteoric water, gets to the surface through clefts, generating new craters in the proximity of the craters already formed (Fig. 4, D and F). As well, dry micro-seeps are relatively abundant in Fibiş Valley. Small pools filled with water and marshy areas with a weak gas bubbling activity were observed (Fig. 4 F) (Baciu et al., 2007). The mud volcano fields are covered with specific vegetation, characteristic for river meadows. All craters occur along the valley, having a NNE-SSW direction, at an altitude of 121.00 meters.

The mud volcanoes occur along an active fault system, acting as conduits for migrating fluids (Loncke et al., 2004). The total surface occupied by

recent or ancient activity of mud volcanic products along the Fibiş Valley is about 0.9 km^2. The term mud volcano is used to indicate larger edifices or fields with similar features (Bonini, 2009 a).

3. MATERIALS AND METHODS

This study is based on data obtained during the fieldwork undertaken in April 2010, including sampling of water, gas, mud, soil and plants of the main area with visible seeps along the Fibiş Valley, named as the Forocici Crater (FC).

The gases (CH_4, CO and H_2S) from FC were collected by immersing a glass funnel in the pool connected to a Crowcon Detection Instruments LTD Custodian portable gas detector. Carbon dioxide was determined through the following method: the air sample (50 ml) containing CO_2 is drawn through a solution of titrated barium hydroxide. After harvesting, the excess barium hydroxide was titrating with solution of oxalic acid, with known normality, using phenolphthalein as indicator. The difference between the amount of barium hydroxide added at the beginning of the analysis and titrated with oxalic acid was consumed by CO_2 of air sample analyzed.

The mud sample was collected from the deep crater (M1 sample). The soil samples were collected from the terrace of Forocici Crater (S1, S2) and Fibiş Valley beside FC (S3, S4). Soil and mud samples preparations were undertaken in accordance with ISO 11464/98, whereas the determination of chromium, copper, iron, nickel, manganese, lead and zinc, followed the ISO 11047/99 method. Soil and mud samples were dried at room temperature prior to the analysis. Dried soil and mud samples were macerated with *aqua regia* (Berrow & Stein, 1983).

Analyses of the metal content in the aerial parts of the plants (leaves and aments) were undertaken. The samples of *Salix cinerea* L. were collected from the terrace of Forocici Crater. The dried plant tissues were macerated with concentrated hydrochloric acid. The plant sampling was undertaken in accordance to the methodology described in STAS 9597/1-74. The sample analysis was undertaken in accordance to the STAS 9597/17-86. Plant, soil and mud extracts analysis was undertaken using a Varian Spectra AAS atomic absorption spectrophotometer, at the National Institute of Research and Development for Industrial Ecology Timişoara laboratories. The content of metals from water samples collected from FC were analysed with same spectrophotometer.

Diatom samples were collected from the clay substratum in the Forocici Crater, with a syringe.

The collected samples were preserved in 4% formalin and labeled. Further processing involved the removal of organic matter in order to obtain siliceous frustules and permanent microscopic slides preparation. In the first stage, the samples were oxidized with concentrated nitric acid, and then rinsed with distilled water several times, with the help of a vacuum pump, until the pH 7 was reached. During the second stage, 1-2 drops of the obtained sample were placed on a cover slip together with a small surfactant quantity (Tween 80, 2%); after the material dried, the cover slips were heated with a gas lamp for 6 hours. During the last stage, the cover slips with the heated material were placed on microscopic slides in a small quantity of rosin, preheated to 95°C. The resulted preparations were eventually cooled, finished and labeled, and the species identification was done using an Olympus BX51 microscope with an immersion objective (100x), photo camera (Olympus E330) and image acquiring software (Quick Photo Micro 2.3). To identify the species, diatom guides were used (Krammer & Lange-Bertalot, 1986, 1988; Krammer, 2000; Lange-Bertalot, 2001).

4. RESULTS AND DISCUSSIONS

4.1. Gas composition and origin

The free gases composition of Forocici mud volcano is completely different from other Romanian CH_4 - dominant seeps [Pâclele Mari - 82.7% , Pâclele Mici - 94.9%, Andreiaşu, - 95.8%, Bazna - 97.9% and Sărmăşel - 98.7% (Baciu et al., 2007)]. The concentration of CH_4 in Forocici Crater is about 12-13%. The gas emission from FC is represented by CO_2, CH_4, CO and H_2S. It was estimated that carbon dioxide emissions could surpass 15% in FC (Uruioc et al., 2011). However, this content is high compared to other areas of Romania, where it does not exceed 7% [Transylvanian Depression (CO_2 = 0.7 - 6.7%) and Carpathian Foredeep (CO_2 = 2.0 - 2.5%) (Baciu et al., 2007)]. Carbon dioxide is responsible for MVs activity from Fibiş Valley. Its origin can be crustal (Bizerea, 1965; Oros, 1991), crustal rocks being the main Ar and He source in the underground waters. In addition, the underground water contains 93.9% CO_2, 0.04% Ar and 0.001% He which emanates from diffusion along faults such as Pişchia-Seceani fault (Table 1) (Mihăilă & Giurgea, 1987). The carbon dioxide flow may be highly variable with time, due to climate variations and to tectonic activity of the area (Etiope et al., 2004a). Emissions of CO_2 and special CH_4 show that these MVs

represent an inevitable source of greenhouse gases for the atmosphere (e.g. Etiope et al., 2004a; Baciu et al., 2007; Etiope, 2009; Mazzini et al., 2009).

Table 1. The content of gas in underground water from Pişchia-Fibiş area (Mihăilă & Giurgea, 1987)

Gas sample	CO_2	O_2	N_2	Ar	He	CH_4
%	93.90	1.00	3.90	0.04	0.001	1.06

The concentration of CH_4 (12-13%) can derive from organic material dissolution in anaerobic conditions or from another source of hydrocarbons (Uruioc et al., 2011). Concentration in H_2S and CO is between 0.39 - 0.87 ppm. Gas emissions from vents were not measured, as well as diffuse soil degassing around the mud volcano. The total gas output was estimated as being at least 35 million m^3 per year (Bizerea, 1965). The global quantity of gas escaping from the Fibiş Valley site is thus difficult to estimate (e.g. Deville & Guerlais, 2009). Further measurements are necessary to estimate the total seepage throughout the Fibiş Valley. In addition, further investigations including gas analyses may provide better knowledge of the origin of the gas, its relation with reservoirs at greater depths, regional magmatism and deeper faults (Baciu et al., 2007).

4.2. Metal composition of water

The mud volcanoes of the Fibiş Valley are located at lower altitude (121.00 meters); hence, meteoric fluids may flush this site and affect the chemistry (Mazzini et al., 2009). The seeping water from the Forocici Crater (FC) shows a relatively low proportion of heavy metals. Fe dominates the seven water samples collected at FC pool, with most samples having Fe concentrations between 2.900 mg/l (in November 2006) and 1.940 mg/l (in March 2007) (Table 2). Low Fe contents (0.060mg/l in May 2007 and 0.059mg/l in April 2010) were observed

when the water dominates the Forocici Crater pool. The Fe occurrence as hematite has been identified at the ground surface in springs, brought together with the mixture of gas (CO_2, CH_4, CO and H_2S), water and mud (Uruioc, 2002). The Fe origin can be biogenic, as a product of ferrobacteria (Bizerea, 1965), or paleoclimatic, the Fe resulting from paleo-alteration of silts deposited during the Pleistocene (Florea, 1964; Rogobete & Ţărău, 1997).

Red siltic clay, rich in Fe, washed by mud and fluids, can derive from other warmer or cooler climate areas (Ianoş et al., 1997).

A low concentration of Cr (0.020-0.050 mg/l) and Ni (0.014-0.050 mg/l) was observed. Low Cr and Zn concentrations suggest than the fluid can possibly derive from shallow reservoir levels, or from meteoric water. Zn (0.740mg/l) alone was identified in one sample (May 2007). We consider that the concentration in heavy metals is contrelled by the mineralized fluids brought from deep layers of the terrestrial crust (Planke et al., 2003). The geochemistry of the expelled water at Forocici Crater may thus represent a mixture of deeper water and shallow meteoric water (e.g. Planke et al., 2003; Mazzini et al., 2009; Uruioc et al., 2011). Furthermore, surface processes diluted by rain during spring-autumn or concentrated by evaporation in summer may also modify it. Seasonal variations in the composition of rain and snow can periodically alter the water composition in the seeps. Hence, it is important to monitor the seep activity. It is important to use satellite images combined with mapping allowing interpretations on the flow chronology and on the identified faults controlling the seep distribution (Mazzini et al., 2009).

Data presented in table 2 show a low variation in time of metal content, thus demonstrating relatively constant water compositions during the six-year time span, excepting Fe. This can be explained by different content of the fluid flow and by annual or seasonal variations of water level in the pool.

Table 2. Heavy metals concentration from water's Forocici Crater (FC) in period 2005 - 2010

Time	Metal mg/l						References
	Cr	Cu	Fe	Ni	Pb	Zn	
November 2005	0.027	bdl*	2.400	0.036	bdl*	bdl*	Uruioc et al. (2011)
November 2006	0.040	bdl*	2.900	0.022	bdl*	bdl*	,,
March 2007	0.020	bdl*	1.940	0.014	bdl*	bdl*	,,
May 2007	0.050	bdl*	0.060	0.050	bdl*	0,740	,,
May 2008	0.030	bdl*	1.085	0.030	bdl*	bdl*	,,
September 2009	0.028	bdl*	1.990	0.020	bdl*	bdl*	,,
April 2010	0.020	bdl*	0.059	0.019	bdl*	bdl*	This paper

bdl* - below detection limit

4.3. Fluids origin from Fibiş Valley seeps

According to Suceavă (1967) cited by Mihailă & Giurgea, (1987), Fibiş Valley seeps are locate along faults zones, Pişchia-Seceani being the main fault. The active character of these faults is confirmed by the occurrence of thermal and mineral water from Pişchia, Lipova, from mofettes of Buziaş, by He, Rd, Ar occurrence and of other gases occurrence with crustal origin (Oros, 1991). The gases origin of the Fibiş valley is the same. Borugă & Airinei (1981) remark the occurrence of carbon dioxide (CO_2) in Banat in different permeable layers. The carbon dioxide emissions under pressure, in their migration pathways, encounter the deeper water that carries it along on faults (Bizerea, 1965). Water moistens limestone and shale layers forming a mixture of gas, water and mud, which reaches the surface through more concentrated seepage. The fluid migration in MVs is controlled by overpressure in depth (Deville & Guerlais, 2009).

Overpressured fluids rise along the Forocici mud volcano conduits and the excess gas is partially expelled along small fractures connected with the Pişchia-Seceani fault. This allows the fluids to mix with shallow meteoric water, finally reaching the surface as pool where only water and gas are seeping (Mazzini et al., 2009). Depending on the oversaturation, these gases can form gas bubbles, which potentially emanate into the water column (Mastalerz, et al., 2007).

According to Etiope et al., (2009 a) the terrestrial mud volcanoes (about 20%) release gas with features between thermogenic and microbial methane. "Gas can be already mixed in the reservoir or mixing can happen during the ascent of gas through the sedimentary horizons of the mud volcano system, where microbial gas pools may exist at shallower depths" (Etiope et al., 2009b).

We propose a schematic view describing the gas and fluid migration pathways of the Forocici mud volcano (Fig. 5). Gases arriving from a deep reservoir migrate through the sedimentary layers. During the migration, the gases mix with deep water and a minor amount of argillaceous material and they partially accumulate within an appropriate lithological reservoir (Mastalerz, et al., 2007). Water, mud and gas migrate through the mud volcano conduits, and rising fluids are in many cases stored in intermediate chambers. Water from the mud volcano may represent deeper fluids slowly percolating to the surface. Deeper water mix with the meteoric waters, but there is considerable variation in the degree of mixing between the two (Planke et al., 2003). Within Forocici Crater, both deep fluids and shallow meteoric fluids converge and mix.

The Fibiş Valley mud volcanoes confirm the presence of regional active fault systems. This aspect is frequently cited in literature (Yusifov & Rabinowitz, 2004; Lynch & Hudnut, 2008; Manga et al., 2009). MVs are especially developed along faults where fluids migrate through hydraulic fracture systems (Deville & Guerlais, 2009).
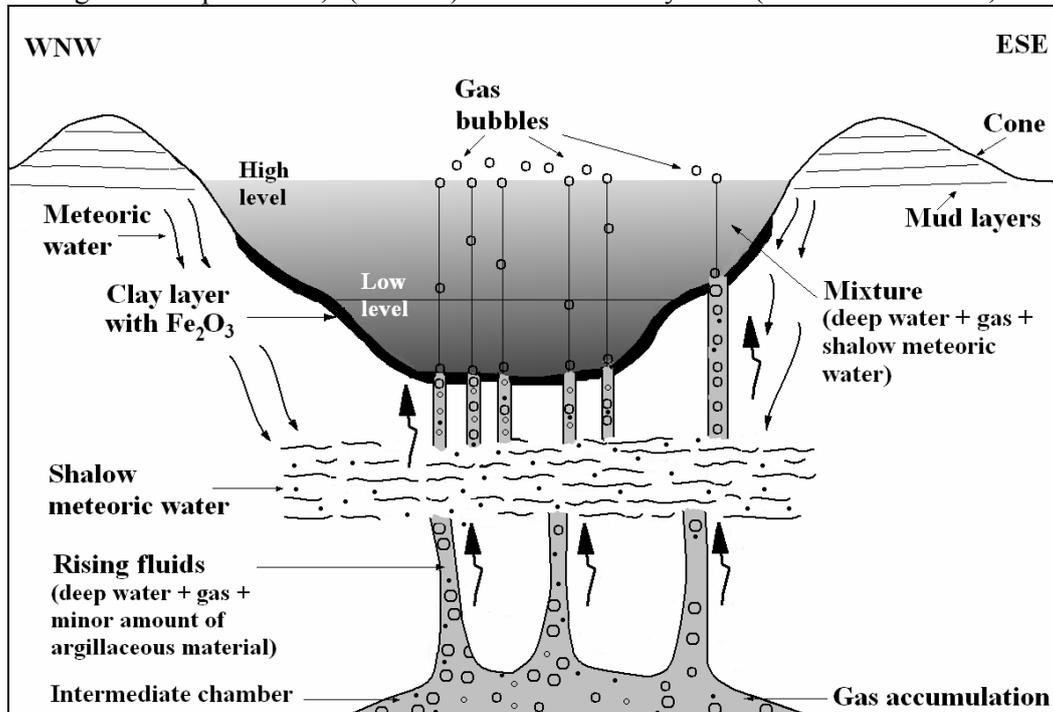

Figure 5. Conceptual drawing (not to scale) the migration pathways of fluids into the Forocici Crater (modified after Bizerea, 1965)

Table 3. Heavy metals content in deep crater, from soil on the slope terrace of Forocici Crater (FC) and Fibiş Valley

Site	Sample ID	Metal mg/kg d.s.*					
		Cu	Fe	Mn	Ni	Pb	Zn
Deep crater	M1 mud	25.5	2266	52.6	8.9	23.6	58.0
Slope terrace	S1 soil	20.0	2144	61.3	4.1	25.8	38.3
Slope terrace	S2 soil	16.3	2190	38.5	2.3	20.6	25.4
Fibiş Valley	S3 soil	17.0	2424	43.1	7.4	27.5	39.6
Fibiş Valey	S4 soil	18.1	2585	43.4	11.5	38.0	74.0

*Dried substance

Table 4. Heavy metals content in *Salix cinerea* L.

Salix cinerea L.	Plant sample	Metal mg/kg d.s.*					
		Cu	Fe	Mn	Ni	Pb	Zn
Leaf	PL	4.50	55.50	udl**	0.15	udl**	42.20
Flower	PF	2.74	80.00	udl**	udl**	1.35	udl**

*Dried substance; **under detection limit

4.4. Heavy metals from mud and soil

The fluids migrating upwards control concentrations of heavy metals in soil on the terrace of the Forocici Crater and of the Fibiş Valley, besides FC and in mud from deep crater. The fluids migrate through the sedimentary layers and carry in solution heavy metals reaching the surface through more seepages affecting the soil composition.

Soil samples of the first 10 cm, were chemically analyzed for the content of heavy metals: Cu, Fe, Mn, Ni, Pb and Zn. Chemical analyses of heavy metal concentrations were compared with reference values of the Ord. MAPM 756/1997. Analyses of heavy metal concentrations of soil show a wide variation (Table 3). All concentrations of metal are low and below maximum allowable, limit.

However, some samples show high concentrations of metal as those that contain Fe, Pb and Cu. Retention and mobility of heavy metals in soil is due to organic and inorganic soil components and of physical, chemical and biological processes (Damian et al., 2010).

All examined samples show higher content of Fe. Content in Fe included between 2144 mg/kg d. s. and 2585 mg/kg d. s. The samples from outside Forocici Crater (S3 and S4) are enriched in Fe compared to those of the inner side (M1, S1 and S2) (Table 3). According to Order no. 756/1997, all samples have Cu content under maximum allowable limit. Exception is M1 sample inside of deep crater, which has a Cu content higher (25.5 mg/kg) over maximum allowable limit (20 mg/kg for soil). This situation is explained by the permanent occurrence of fluids flow that arrives from depth with heavy metals. Zn concentrations do not exceed the allowable maximum values. Ni is the metal that presents low and moderate concentrations in mud and soil. The Mn content has low values between 38.5 mg/kg and 43.4mg/kg.

Generally, heavy metal concentrations in soil do not exceed the allowable maximum values, except Pb, which is a toxic metal for animals grazing in Fibiş Valley.

4.5. Metal concentration in plant

Salix cinerea L. (Salicaceae) grows on the soil at the boundary between the crater and FC cone. It contains Cu, Fe Ni, Pb, and Zn (Table 4). The analysis of flowers grouped in aments, belonging to *Salix cinerea* L. (PF), showed that the flower does not accumulate Ni and Zn, but has high concentrations of Fe (80 mg/kg) and less Cu (2.27 mg/kg) and Pb (1.35mg/kg).

Due to competition in retention of metallic ions, the iron excess in the soil solution decrease the uptake by plant roots of other metals such as Cu, Pb, Mn, or Ni. The results show that the willow is adaptable to high Fe concentrations in the soil (2144 mg/kg d. s. and 2585 mg/kg d. s). Due its the growth performance, heavy metal accumulation and mobilization, *Salix cinerea* L. can be considered a valuable plant for phytoextraction of metals in polluted soils (Vysloužilová et al., 2006). Every year, *Salix cinerea* L. also drops aments later incorporated in soil through bioaccumulation processes, leading to a concentration of heavy metals in the topsoil surrounding the plant. It is possible that deeper soil horizons are reached (Uruioc et al., 2011). The data in table 4 show that significant differences were found for leaf and flower concentrations.

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The leaves have high content of Cu (4.50 mg/kg) and Zn while flowers have low content in Cu (2.74 mg/kg). Other plants growing on the FC slope terrace (*Polygonum aviculare* L.) compared to the same species outside the perimeter, assimilate a high quantity of heavy metal (e.g. Fe between 961.3 mg/kg and 639.2 mg/kg) (Uruioc et al., 2011).

These data suggest that fluids arriving from a deep reservoir migrate through the sedimentary layers and become enriched in heavy metals. Fluid composition and concentration in heavy metal are controlled by lithology, by diagenetic processes and by water mixtures (Planke et al., 2003).

4.6. Benthic diatom communities from Forocici Crater

In the collected samples of the clay substratum of FC, were identified six species of benthic diatoms: *Gomphonema minutum* (Agardh) Agardh, *Pinnularia obscura* Krasske, *Geissleria acceptata* (Hustedt) Lange-Bertalot & Metzeltin, *Hippodonta linearis* (Østrup) Lange-Bertalot, Metzeltin & Witkowski, *Stauroneis anceps* Ehrenberg and *Nitzschia solita* Hustedt (Fig. 6). Of these, the first species was reported sporadically in the analyzed samples, whereas the others are rare. Compared to the reports on diatom species from this site, collected in April 2007 (Uruioc et al., 2007), no new species were identified and there were no alterations regarding the frequency and abundance of species, mainly due to the fact that samples were collected in the same period of the year.

A monthly monitoring, or at least seasonally, could provide new information. Diatom species identified in the bubbling pool of the Forocici Crater show a wide ecological tolerance, allowing them to vegetate under particular environmental conditions occurring here. In terms of diatom ecology, these waters can be characterized as mesotrophic – eutrophic, with a medium to high electrolytic content.

Different species of diatoms were reported before in such particular environments, as components (sometimes with significant occurrence, fact indicated by a yellow-brownish color) of photoautotroph communities found at the water/sediment interface, usually in the superior stratum, which is the most illuminated layer (Franks & Stolz, 2009). Sometimes, they occur even in aquatic bodies with high temperature (20 - 60°C); their abundance varying inversely with the temperature (Fernandez-Turiel et al., 2005). Due to their preferences, diatoms are considered among the most sensitive organisms which can be used as instruments to evidence effects of the volcanic or post-volcanic activities in aquatic ecosystems (Urrutia et al., 2007).

Diatoms, as a part of the protist community, excrete metabolic products in the environment, contributing to the concentrations of some metal ions, such as Cu or Cd (Gouvêa et al., 2005).

4.7. Mud volcanoes environment impact

The fluid that flows from mud volcanoes from Fibiş Valley has an important impact upon gas budget of atmosphere. The total gas output was estimated to 35 million m^3 per year (Bizerea, 1965). A long term monitoring is necessary in order to generate new data regarding modification of fluids composition in time. The plants that grow near the craters or on the Forocici Crater terrace assimilate larger quantity of heavy metal. Thus, *Salix cinerea* L. accumulates a quantity of Fe without showing a morphologic change, through the assimilation of metals.

Figure 6. Diatoms found on the clay substratum of Forocici Crater (FC)
A - *Hippodonta linearis;* B - *Stauroneis anceps*

On the other hand, *Polygonum aviculare* L. growing on the FC terrace, has red leaves at maturity, because of the assimilation of a high quantity of Fe (Bizerea, 1965). The same plant, out of the perimeter with seeps, has a low Fe content and has no red leaves (Uruioc et al., 2011).

Generally, heavy metal concentrations in soil do not exceed the allowable maximum values, except for Pb, a toxic metal for animals grazing in Fibiş Valley. However, diseases were not reported yet among the cattle stock of this area.

During summer, the water level in the Forocici Crater is very low, and the water may disappear almost entirely. Animals, such as birds standing incidentally in the interior of FC, due to the CO_2 accumulation, die. In the same time, on the horizontal surface of Fibiş Valley, the thin layer of CO_2 accumulated above the field allows the development of more abundant vegetation than that of Beregsău River, flowing almost parallel to Fibiş.

5. CONCLUSIONS

Although lacking specific analyses, we assign the active processes of fluid migration from the deep horizons of Banat area, to the mud volcanoes (MVs) category. The activity of the mud volcanoes from this area consists of a continuous gas bubbling and water overflow from salses and small eruptions of green-house gases from vents. Therefore, further water analyses and measurement of the gas flux, are necessary to estimate the total seepage throughout the Fibiş Valley. During the migration, the gases mix with deep water charged with minor amount of argillaceous material. FC deep fluids and shallow meteoric fluids converge and mix. Water samples collected at the Forocici Crater pool are dominated by Fe. Low concentration has been observed at Cr, Ni and Zn. Concentrations in heavy metals are controlled by the mineralized fluids from deep layers. Water, mud and gas migrate through mud volcano conduits, and fluids are stored in intermediate chambers.

Analyses of heavy metal concentrations of soil show a wide variation. All concentrations of metal are low and below maximum allowable limit, but some samples show high concentrations of metal, such as those containing Fe, Pb and Cu. Heavy metal concentrations in soil, generally do not exceed the allowable maximum values, except for Pb, which is a toxic metal for animals grazing in Fibiş Valley.

Salix cinerea L*.,* growing on the formed soil at the boundary between the crater and FC cone contains Fe, Cu, Ni, Pb and Zn. The results show that this willow species is adaptable to high Fe concentrations in the soil of this site.

From the substratum of the Forocici Crater, six species of diatoms were identified. They may be considered among the most sensitive organisms, which can be used as markers for the effects of post-volcanic activities in this area. In Fibiş Valley, the mud volcanoes confirm the occurrence of regional active fault systems. These faults may act as conduits for rising fluids.

These data setes represent an important step to understand the contributions of deep and shallow fluids of the mud volcano activities and to estimate the environmental impacts of MVs. Further studies will assess the origin of gases and their relationships with the local tectonics.

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