



# A new hydrothermal scenario for the 2006 Lusi eruption, Indonesia. Insights from gas geochemistry

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## ABSTRACT

The 29th of May 2006 gas and mud eruptions suddenly appeared along the Watukosek fault in the north east of Java, Indonesia. Within a few weeks several villages were submerged by boiling mud. The most prominent eruption site was named Lusi. To date (November 2011) Lusi is still active and a ~7 km<sup>2</sup> area is covered by the burst mud breccia.

The mechanisms responsible for this devastating eruption remain elusive. While there is consensus about the origin of the erupted mud, the source of water is uncertain, the origin of the gas is unknown and the trigger of the eruption is still debated. In order to shed light on these unknowns, we acquired a wide set of data of molecular and isotopic composition of gas sampled in several Lusi vents, in the surrounding mud volcanoes, in the closest natural gas field (Wunut), and in the hydrothermal vents at the neighbouring volcanic complex in the period 2006–2011.

The boiling fluids erupted in the crater zone are apparently CO<sub>2</sub>-dominated, while colder CH<sub>4</sub>-dominated and C<sub>2</sub>–C<sub>3</sub> bearing fluids are identified at several sites around the crater zone. Gas genetic diagrams, maturity plots and gas generation modelling suggest that the hydrocarbons are thermogenic ( $\delta^{13}\text{C}_1$  up to  $-35\%$ ;  $\delta^{13}\text{C}_2$  up to  $-20\%$ ), deriving from marine kerogen with maturity of at least 1.5%Ro, for instance in the ~4400 m deep Ngimbang source rocks. CO<sub>2</sub> released from the crater and surrounding seeps is also thermogenic ( $\delta^{13}\text{C}$  from  $-15$  to  $-24\%$ ) related to kerogen decarboxylation or thermal CH<sub>4</sub> oxidation in deep rocks, although three vents just outside the crater showed an apparent inorganic signature ( $-7.5\% < \delta^{13}\text{C} = -0.5\%$ ) associated to mantle helium (R/Ra up to 6.5). High CO<sub>2</sub>–CH<sub>4</sub> equilibrium temperatures (200–400 °C) are typical of thermally altered hydrocarbons or organic matter. The data suggest mainly thermally altered organic sources for the erupted gases, deeper sourced than the mud and water (Upper Kalibeng shales). These results are consistent with a scenario of deep seated (>4000 m) magmatic intrusions and hydrothermal fluids responsible for the enhanced heat that altered source rocks and/or gas reservoirs. The neighbouring magmatic Arjuno complex and its fluid–pressure system combined with high seismic activity could have played a key role in the Lusi genesis and evolution. Within this new model framework, Lusi is better understood as a sediment-hosted hydrothermal system rather than a mud volcano.

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## 1. Introduction

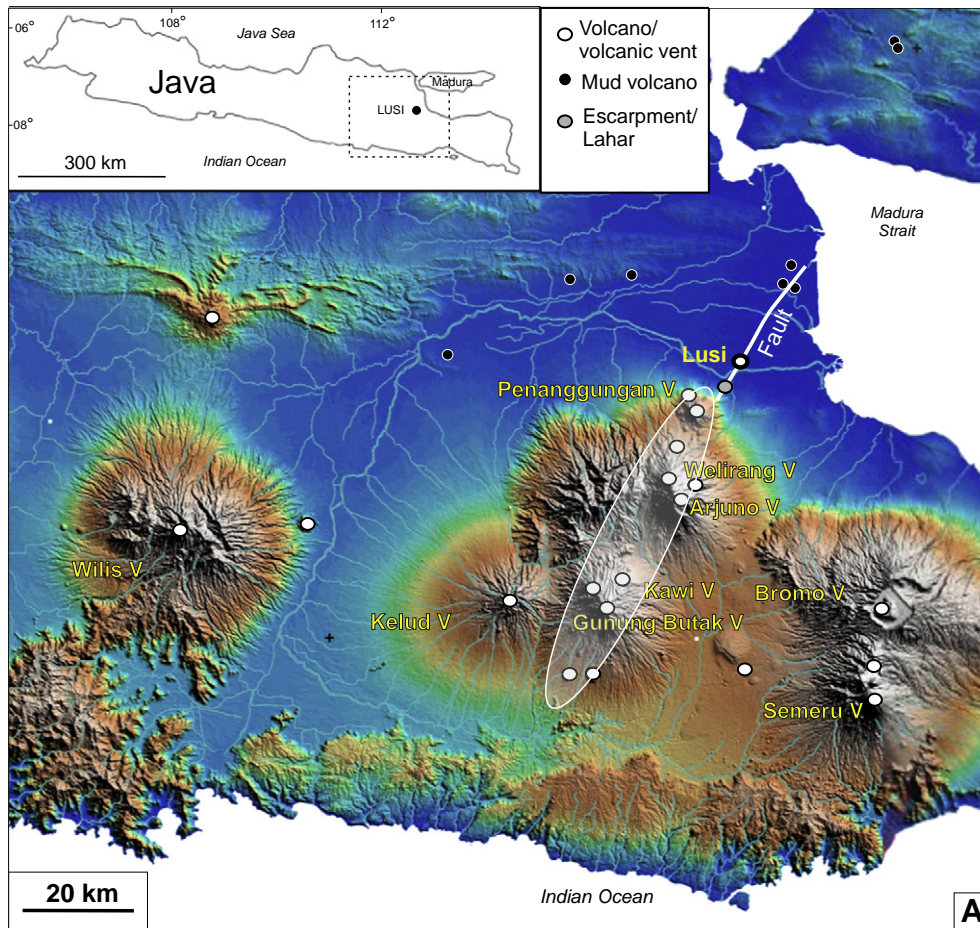
The 29th of May 2006 the Sidoarjo district in East Java, Indonesia witnessed the sudden birth of numerous gas and mud vents on the ground. These eruption sites extended over a distance of more than a kilometre forming a NE–SW alignment. Within a few days a prominent crater formed and the initial vents were quickly covered by the large volumes of erupted boiling mud. The flow-rates from the main crater reached 180,000 m<sup>3</sup>/day (Mazzini et al., 2007). The largest crater was named Lusi [acronym from LUmpur (mud) and Sidoarjo (the district name)]. To date (November 2011) Lusi is still active and has since 2006 gained the constant attention of the

media due to the spectacular nature of the eruption that devastated and submerged villages, displaced more than 50,000 people and covered a region of more than 7 km<sup>2</sup> by hot mud (Fig. 1).

The trigger of the Lusi eruption remains debated. One hypothesis links Lusi to a blowout from a neighbouring well (Davies et al., 2007; Tingay et al., 2008), whereas others have confuted the “man-made” scenario (e.g. Sawolo et al., 2009, 2010). An alternative hypothesis links the eruption events to the reactivation of a fault following a 6.3 M earthquake that struck the Java Island the 27th of May 2006 (Mazzini et al., 2007, 2009; Tanikawa et al., 2010). More than five years after the birth of Lusi, we stress that the geological knowledge of the Lusi region and the neighbouring areas shows that: 1) The presence of a NE–SW oriented fault (Watukosek fault, Fig. 1) that originates from the neighbouring Arjuno–Welirang volcanic complex, crosses Lusi and extends towards the NE of Java. Field observations showed that lateral shearing

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**Fig. 1.** (A) Elevation map of eastern Java Island. Highlighted the position of some known mud volcanoes, and main volcanoes and volcanic vents. Note: the orientation of the main vents of the Arjuno–Welirang volcanic complex has the same NE–SW direction of the Watukosek fault that also hosts a large escarpment as well as other mud volcanoes; (B) satellite image of Lusi the 14 of June 2011, courtesy of Crisp, NUS 2011. Note the brownish areas on the outskirts of the craters represent dry zones where it is possible to access. The central part closer to the crater remains muddy. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

was reactivated after the 27th of May 2006 6.3 M earthquake. 2) The fault escarpment is composed of volcanic rocks and is located only 4.6 km from Lusi. 3) The nearest volcano is the Holocene Penanggungan (~10 km from Lusi) and the active Arjuno–Welirang volcano is 25 km further to the SW. 4) Volcanic vents and mud volcanoes (Fig. 1A) in this part of Java are aligned towards Lusi with the same strike of the Watukosek fault (Carn, 2000; Mazzini et al., 2007).

Regardless what the trigger for the Lusi eruption is (man-made or natural), it remains crucial to further investigate the origin of the erupted fluids, the eruption mechanisms, and the possible interactions between volcanism, tectonics, and seismicity. New data is needed in order to answer the following key questions: Is Lusi a mud volcano? What is the origin of the gas, and is its source shallow or deep? Do mud and gas have the same origin? How long will the eruption last? And what are the triggers that feed the eruption and the episodic pulsations? Are more eruptions like Lusi likely to occur in other parts of Java? And finally, is there a connection between Lusi and the neighbouring volcanic complex?

In order to address some of the above questions and to investigate the possible involvement of deep volcanic fluids in Lusi, we present a new set of gas geochemical data collected from 2006 (i.e. during the initial phase of the eruption) to 2011. The gas samples were taken from the Lusi crater, from other mud volcanoes located nearby and from the closest exploited natural gas field, Wunut. We claim that understanding the gas origin (microbial, thermogenic or abiotic) and source (shallow or deep hydrocarbon reservoirs or geothermal fluids) are essential to move ahead towards a better understanding of Lusi.

## 2. Geological setting

The island of Java is a prospective province for hydrocarbon exploration and is an exceptional area where volcanism, piercement structure formation and tectonism interact. A detailed and broad overview of the Indonesian basins, their geology and petroleum systems, are provided by Doust and Noble (2008) and references therein. The Lusi area is particularly interesting as sedimentary and volcanic phenomena are intimately connected and can be studied in detail. Moreover the Lusi eruption provides a unique opportunity to study an erupting system from its birth.

Lusi is located in a Tertiary-aged back-arc basin of NE Java, in the East Java Basin (Kusumastuti et al., 2000), where the sedimentary sequences are characterised by high sedimentation rates of organic-rich deposits (e.g. rates of about 2.5 km/Ma since the Pleistocene) often capped by sealing units of tuffaceous or volcanoclastic sands. The presence of numerous mud volcanoes in the NE Java highlights that sedimentary volcanism is a wide-spread phenomena in this part of the island (Mazzini et al., 2007; Satyana and Purwaningsih, 2003). The formation of mud volcanoes is likely to be triggered by frequent seismic activity, leading to the release of overpressured fluids that formed by hydrocarbon generation and illitization of clays at depth.

The setting of the Lusi eruption is peculiar when compared with the other mud volcanoes in Java as it is situated at the southernmost tip of the back-arc region neighbouring the large Arjuno–Welirang volcanic complex. Arjuno–Welirang is part of the volcanic arc contouring the southern part of Java (Figs. 1, 2). The complex comprises two older volcanoes named Mount Ringgit (to the east), and Mount Linting (to the south). Arjuno–Welirang is a stratovolcano and volcanic cones and craters are located in a 6 km line between Arjuno and Welirang. The northernmost volcano of this complex is Penanggungan which crater is located 10 km to the SW of Lusi.

## 3. Lusi phenomena: open questions

What is Lusi? Is it really a mud volcano (i.e. sedimentary volcanism driven by gravitational sediment imbalance and fluid overpressures (e.g. Kopf, 2002)) as commonly wisdomed?

Geochemistry of the fluids and petrography of the solids erupted by Lusi were first reported by Mazzini et al. (2007). The early samplings showed the presence of mud breccia and significant amounts of methane, typical of mud volcanoes worldwide. Preliminary analyses of the erupted material indicated that the main source of clay and water is the overpressured units located at ~1500–1800 m that were rapidly buried and under-compacted. This type of deposits is typical at many mud volcano settings. Moreover, water compositional analyses indicated a strong signature of illitized clay minerals; this is common for many mud volcanoes, however a possible deeper water component could not be excluded. The first gas analyses from Lusi, showed the presence of methane of mixed microbial–thermogenic origin, but the actual source/reservoir rocks (shallow or deep shales and limestones) remained elusive.

Despite the research carried out so far, many aspects of this phenomenon remain without explanation.

The longevity of the Lusi eruption (still active after more than 5 years) is atypical when compared to other mud volcanoes that usually extinguish the largest amount of overpressure after few hours or days and become dormant (Aliyev et al., 2002; Deville and Guerlais, 2009; Shnyukov et al., 1986). Is this surprising activity related to the Lusi infancy, or are mud volcanoes not relevant analogues for understanding Lusi? Several lines of evidence suggest that Lusi is similar to a hydrothermal system rather than a mud volcano. For instance, Lusi is characterised by high temperature and pulsations with sudden increase in flow rates that, so far, have not been clearly explained (Mazzini et al., 2009). Is the frequent seismicity in Indonesia altering Lusi plumbing system? Or the fluctuating flow rate is merely related to volumetric contractions of the conduit? The generally high geothermal gradient at Lusi locality (42 °C/km) is likely due to the proximity to the adjacent volcano (Mazzini et al., 2007), which may explain the clay mineral and geochemical transformations occurring at relatively shallow depths. For example the illitization of clays is occurring at 1100 m (and possibly shallower) while in most of the other basins it takes place at greater depths (e.g. Kholodov, 2002 and references therein). Finally, unlike the commonly CH<sub>4</sub>-dominated gases erupted from mud volcanoes worldwide, the main gas erupted at Lusi is CO<sub>2</sub>. This questions the applicability of standard mud volcano models (e.g. Kopf, 2002) to Lusi.

## 4. Methodology

### 4.1. Field work sampling and measurements

Five fieldwork campaigns (2006, 2007, 2008, 2010 and 2011) included sampling of the seeping fluids at Lusi site, from the neighbouring Wunut gas field, from other nearby mud volcanoes, and from some of the hydrothermal seeps at the Arjuno–Welirang volcanic system. A total of 43 gas samples were collected and grouped according to their geological setting; Group 1: Fluids emitted from the main vent that today coincides with the Lusi crater; Group 2: Gas sampled from the satellite cold seeps surrounding the crater; Group 3: Gas from the Wunut gas field; Group 4: Gas from seven mud volcanoes in the NE of Java region (Gunung Anyar, Kalang Anyar, Pulungan, Gunung Sening, Gunung Bulag, Pangangson, Bledug Kuwu), Group 5: Gas from four hydrothermal seeps at or close to the Arjuno volcano. All gas samples were stored into brine-filled glass bottles that were subsequently hermetically sealed (see Mazzini et al., 2011 for details).

The sampling from the erupting Lusi crater (Group1) was conducted with the help of an excavator that was driven to the circular dike framing the crater to collect a scoop of boiling mud. Additional samples were collected on the framing dike standing against the wind facing the crater. When violent burst of mud and gas were emitted, gas samples were vacuumed from the thick clouds and stored. A pioneering technique was used in 2010 (at that time all



**Fig. 2.** (A) Helicopter view of Lusi (19 May 2011), on the background the Watukosek escarpment and the Penanggungan volcano. Note: the broken containment dam that was destroyed 5 h after the 26-04-2011 earthquake is aligned along the Watukosek fault system connecting Lusi and the volcanic complex; (B) View of the close volcanic complex from Lusi site. From left to right: Arjuno, Welirang (erupting white smoke), and Penanggungan; (C) View of erupting Lusi crater approached walking on dry erupted mud; (D) One hour after the 26-04-2011 earthquake two distinct craters formed at Lusi site clearly indicating a correlation between the two events; (E) Helicopter view with detail of the damaged dam after the earthquake suggesting reactivation of the strike-slip fault. A, B, D, E courtesy of S. Hadi.

the dikes framing the crater were already submerged by the boiling mud) when an excavator on a float drifting to the crater zone allowed sampling. Thus it was impossible to collect gas samples directly from the central part of the crater zone. Gas samples from the other localities (Groups 2–4) were collected with more traditional techniques, as funnelling the gas and storing it into brine-filled glass bottles.

Group 2 includes satellite seeps that are either aligned along the direction of the Watukosek fault or distributed around the crater where the main caldera collapse occur (i.e. Istadi et al., 2009). Samples were collected during the campaigns in 2006, 2007 and 2011.

Group 3 refers to the Wunut gas field in shallow sandstones (350–700 m). The main wells are located about 4 km to the west of Lusi and the reservoir extends laterally towards Lusi. Samples were collected in 2010 directly from two production pipes accessing two

reservoir zones respectively between 341–379 and 670–680 m depth. Samples for helium analyses were collected from a pipeline merging the production units mentioned above.

Seven additional mud volcanoes (Group 4 samples) are located in the NE Java Basin and Madura Island, and one of them further to the west of central Java (see Fig. 1 in Mazzini et al., 2009 for location). These gas samples were collected during the campaigns in 2006 and 2008.

Gases from Group 5 were collected in 2011 at natural hydrothermal seeps around the Arjuno–Welirang volcanoes (Songgoriti, Cangar, Pacet localities) and from a hydrothermal seep connected to a deep water well close to Penanggungan volcano (Kepulungan locality).

At all seepage sites, the exit temperature was measured with a TFX 392 SK-5 thermometer with a precision of 0.1 °C.

## 4.2. Laboratory analyses

The 2006, 2007, 2008 gases ( $N_2$ ,  $O_2$ ,  $CO_2$ ,  $C_1$ – $C_2$ + hydrocarbons) were analysed at the Institute for Energy Technology (IFE), Norway, using a Hewlett Packard 5890 Series II GC equipped with Porabond Q column, a flame ionisation detector (FID), a thermal conductivity detector (TCD) and a methylization unit. The accuracy of the gas composition is better than 3% (1 sigma) for all compounds. The stable carbon isotopic composition of methane and carbon dioxide was determined by a GC-C-IRMS system at IFE. Repeated analyses of standards indicate that the reproducibility of  $\delta^{13}C$  values is better than 1‰ VPDB (2 sigma).

The gas samples collected in 2010 were analysed at the Isotech Labs Inc. (Illinois, USA) for  $C_1$ – $C_6$  hydrocarbons, He,  $H_2$ , Ar,  $O_2$ ,  $CO_2$ ,  $N_2$ , (Carle AGC 100–400 TCD-FID GC); precision 2% (1 $\sigma$ ), and isotopic compositions  $\delta^{13}C_1$ ,  $\delta D_1$ ,  $\delta^{13}C_{CO_2}$  (Finnigan Delta Plus XL mass spectrometer, precision  $\pm 0.1\%$  (1 $\sigma$ ) for  $^{13}C$  and  $\pm 4\%$  (1 $\sigma$ ) for  $^2H$ ). Results are reported in  $\delta$  notation, as per mil (‰) deviation relative to the VPDB (for carbon) and VSMOW (for hydrogen) standards.

As a result of the difficult sampling logistics, several of the crater samples have high air contamination. We applied air-free correction to verify the order of magnitude of non-atmospheric  $CH_4$  and  $CO_2$  concentration and compare with less contaminated samples outside the crater. The corrected compositional values must be considered as “apparent”, since they could be affected by the relative analytical error at low concentrations.

At the sites with vigorous gas seepage, samples for He analyses were collected in annealed copper tubes sealed in the field using a cold welding clamp. At localities where gas was seeping as small bubbles, water samples were collected for further gas extraction. The samples were analysed at the Istituto Nazionale di Geofisica e Vulcanologia (Italy). The isotopic analyses were performed on a purified helium fraction (Sano and Wakita, 1988) by a static vacuum mass spectrometer (GVI5400TFT) that allows simultaneous detection of  $^3He$  and  $^4He$ -ion beams, thereby keeping the  $^3He/^4He$  error of measurement to very low values. Typical uncertainties in the range of low- $^3He$  samples are within  $\pm 5\%$ . The results are given in the R/Ra notation, where R is the sample  $^3He/^4He$  ratio and Ra is the atmospheric  $^3He/^4He$  ratio,  $1.39 \times 10^{-6}$ .

During the 2011 fieldwork seven samples were collected for helium analyses. Four of these samples revealed significant air contamination (i.e.  $0.5 < He/Ne < 0.8$ ). Although these contaminated values cannot be considered quantitatively, we have included them in Tables 1A, 1B (see details in the caption).

## 5. Results

### 5.1. Molecular and isotopic composition of gas

Tables 1A, 1B summarise the molecular and isotopic composition of the sampled gas. The interpretation of gas origin is exclusively based on isotopic data and the relative composition of methane, alkane and propane. Five samples (Lusi crater JV10-07; west of Lusi crater JV11-12, JV11-13; Songgoriti JV11-07; and Wunut WU-1) provided reliable helium isotopic data without significant air contamination ( $He/Ne > 1$ ).

Gas from Group 1 (Lusi crater with boiling mud) is apparently  $CO_2$ -dominated (up to 89%). The  $\delta^{13}C_{CO_2}$  measured during 2006–2010 varies between  $-13.1$  and  $-23.5\%$ . A smaller portion of the emitted gas is represented by hydrocarbon gases from  $C_1$  to  $C_4$ . The  $\delta^{13}C_{CH_4}$  varies from  $-51.8$  to  $-35.7\%$ .

Group 2 gas (satellite seeps) is methane-dominated (up to 97%) and the isotopic values are in the same range of those measured from the crater zone (Group 1). Ethane, propane and heavier alkanes in Groups 1 and 2 occur in concentrations typical of thermogenic gas, with  $C_1/C_2 + C_3$  ratios from 3 to 349. Only two exceptions show

significantly higher values (i.e. JV11-03, JV11-04) where molecular fractionation is likely to be present. The helium from the closest seep to the Lusi crater (92 ppmv) has a R/Ra isotopic ratio = 5.53. Two more seeps (JV11-12 and JV11-13) located to the west of the Lusi crater revealed consistent high R/Ra isotopic ratio (5.01 and 6.47 respectively). Station JV11-03 (~300 m NW from the crater) showed significant air contamination (i.e.  $He/Ne = 0.86$ ) but still gave a high R/Ra = 2.79. Group 3 gas (Wunut gas field) is methane-dominated (up to 98%). The deepermost units of the field reveal a larger amount of  $C_2$ + hydrocarbons (5.4%) compared with the shallower units (1.5%). The  $\delta^{13}C_{CH_4}$  ranges from  $-38.9$  to  $-38.3\%$ , while  $\delta^{13}C_{CO_2}$  shows positive values ( $+13.2\%$ ). Helium (53 ppmv) shows a slight mantle component with R/Ra ~1.4.

Group 4 (NE Java Basin mud volcanoes) samples are all  $CH_4$ -dominated ( $79\% < CH_4 < 99\%$ ) with  $\delta^{13}C_{CH_4}$  signatures from  $-33.5$  to  $-55.7\%$  and  $\delta^{13}C_{CO_2}$  ranging from  $-0.3$  to  $-18.8\%$ . A remarkable exception is Bledug Kuwu that has a similar morphology and eruption mode to Lusi, and, like Lusi, releases  $CO_2$  dominated gas ( $CO_2 = 84\%$ ).

Group 5 gas (hydrothermal vents for the neighbouring volcanic arc) is represented by the Songgoriti locality that has a  $CO_2$ -dominated gas (99%) with high  $\delta^{13}C_{CO_2}$  (10.7‰) and relatively high R/Ra isotopic ratio (2.75). Similarly to station JV11-03, the helium sampling for the other hydrothermal localities (Kepulauan, Cangar, Pacet) was affected by air contamination. Still relatively high values were measured with R/Ra up to 3.69.

### 5.2. Temperature data

Since 29-05-2006, Lusi has been constantly erupting mud (including clasts), gas and water. A constant plume of aqueous vapour has been observed rising from the centre indicating that the temperature of at least 100 °C has been present since the beginning. This is consistent with the reported measurements (Tables 1A, 1B). So far it has not been demonstrated if there is a direct correlation between temperature rise and the sudden bursts with increased flow rates. The recorded temperature data (Tables 1A, 1B) from the crater zone (Group 1) have temperatures close to 100 °C. This contrasts the much lower (~30 °C) temperatures at satellite seeps (Group 2). It is interesting to notice that in all the samples but one, the  $CO_2$ -dominated samples have consistently higher temperatures, while the colder satellite seeps (Group 2) are  $CH_4$  dominated.

Measurements from a seep of Bledug Kuwu gave a temperature of 31.1 °C. It was impossible to collect temperature measurements from the crater. However, judging from the significant clouds of steam released after each burst, temperatures close to 100 °C are expected.

All the other mud volcanoes showed relatively low temperatures with values ranging between 27.7 °C and 34.6 °C. These values are consistent with those typically found in other mud volcanoes during dormant periods (e.g. Etiope et al., 2002; Nakada et al., 2011) and are likely to be affected by air temperature.

## 6. Discussion

### 6.1. Methane and heavier alkanes

Classic gas genetic zonation diagrams (Schoell and Bernard plots, Fig. 3A–B) suggest that the Lusi samples collected in 2006 contain thermogenic hydrocarbons mixed with a minor microbial  $CH_4$  component, as preliminary reported by Mazzini et al. (2007). However the samples collected after 2006 show that the gas composition evolved, the microbial component disappeared (Tables 1A, 1B). The emitted gas shows a complete thermogenic signature, similar to the gas sampled from the Wunut gas field, Gunung Senig and Gunung Bulag MVs. In the Bernard diagram (Fig. 3B), the gas of these MVs appear to be molecularly fractionated, with higher  $C_1/(C_2 + C_3)$  ratios. Molecular fractionation during gas migration, probably due to

**Table 1A**  
Molecular composition of sampled gas in vol.%. N<sub>2</sub> for samples JV06-07 and JV06-10 is respectively 1.703 and 1.736.

Sample ID	Locality	Date	Comments	Group	T int. (°C)	C <sub>1</sub>	C <sub>2</sub>	C <sub>3</sub>	iC <sub>4</sub>	nC <sub>4</sub>	iC <sub>5</sub>	nC <sub>5</sub>	C <sub>6+</sub>	He	H <sub>2</sub>	Ar	CO <sub>2</sub>	C <sub>1</sub> /C <sub>2+</sub>
JV06-18	Lusi crater	09-2006	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	74.465	0.170	0.043	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	25.321	348.83
JV06-18	Lusi crater	09-2006	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	35.227	1.328	0.629	0.106	0.158	bdl	bdl	bdl	nd	nd	nd	62.552	15.86
JV06-18	Lusi crater	09-2006	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	26.148	1.174	0.560	0.085	0.139	bdl	bdl	bdl	nd	nd	nd	71.894	13.35
JV07-03	Lusi crater	02-2007	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	20.015	0.988	0.398	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	78.598	14.43
JV07-03	Lusi crater	02-2007	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	38.934	2.116	1.053	0.158	0.249	bdl	bdl	bdl	nd	nd	nd	57.489	10.89
JV07-03	Lusi crater	02-2007	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	39.801	2.133	1.044	0.166	0.251	bdl	bdl	bdl	nd	nd	nd	56.604	11.073
JV07-05	Lusi crater	02-2007	> 100 m S of crater. Boiling mud taken with excavator scoop	1	95.0	10.364	0.381	0.118	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	89.137	20.76
JV08-13	Lusi crater	02-2008	Sample taken from excavator scoop along the crater	1	93.0	33.400	0.141	0.030	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	66.428	194.44
JV08-13	Lusi crater	02-2008	Sample taken from excavator scoop along the crater	1	93.0	12.657	0.827	0.336	0.060	0.088	0.027	0.012	bdl	nd	nd	nd	85.994	9.39
JV08-14	Lusi crater	02-2008	Dense clouds of erupted fumes vacuumed <sup>a</sup>	1	95.0	11.654	0.752	0.308	0.091	0.058	bdl	bdl	bdl	nd	nd	nd	87.136	9.64
JV10-04	Lusi crater	02-2010	Sample taken from excavator scoop along the crater	1	87.5	11.718	1.234	1.850	0.617	0.925	0.308	0.308	0.925	nd	nd	bdl	82.114	1.90
JV10-05	Lusi crater	02-2010	Sample taken from excavator scoop along the crater	1	87.5	8.949	0.942	1.884	0.471	0.942	0.471	0.471	0.942	nd	nd	5.592	79.337	1.46
JV10-06	Lusi crater	02-2010	Sample taken from excavator scoop along the crater	1	87.5	8.554	0.684	1.026	0.342	0.342	0.000	0.000	0.684	nd	nd	bdl	88.367	2.78
JV040706	Lusi	07-2006	Close to crater 04.07.2006	2	nd	85.47	2.286	0.978	0.206	0.317	0.124	0.124	0.551	nd	nd	nd	9.94	18.63
JV130706	Lusi	07-2006	Close to crater 13.07.2006	2	nd	83.010	3.0181	1.283	0.261	0.404	0.154	0.143	0.380	nd	nd	nd	11.35	14.71
JV06-07	Lusi	09-2006	Small seep outside main dam with microbial colonies around	2	30.8	79.097	0.521	0.208	0.031	0.031	0.007	0.005	bdl	nd	nd	nd	18.395	98.34
JV07-06	Lusi	02-2007	1.5 km south of main crater, very strong bubbling. Was documented in December 2006 and became stronger and stable	2	29.0	92.359	4.134	1.648	0.248	0.287	0.083	0.069	bdl	nd	nd	nd	1.172	14.28
JV07-07	Lusi	02-2007	1.5 km south of main crater, very strong bubbling diffused in a large ditch	2	30.0	93.080	4.102	1.700	0.272	0.320	0.095	0.077	bdl	nd	nd	nd	0.355	14.18
JV10-07	Lusi	02-2010	Isolated bubbling point close to crater. Colder site	2	35.4	94.469	1.078	0.106	0.022	0.007	0.004	0.001	0.004	nd	nd	0.124	4.184	77.28
JV11-03	Lusi	05-2011	300 m to the NW of Lusi crater, isolated seep, moderate activity	2	40.5	97.523	0.029	0.003	bdl	bdl	bdl	bdl	bdl	0.038	0.783	nd	1.625	3343.84
JV11-04	Lusi	05-2011	350 m to the SW of Lusi crater, small pool with microbial colony	2	35.0	97.587	0.030	0.004	bdl	bdl	bdl	bdl	bdl	0.046	0.000	0.106	2.228	3282.46
JV11-12	Lusi	05-2011	900 m west of Lusi crater, in the village, old well now collapsed forming a 10 × 10m water pool with bubbling gas	2	28.5	95.686	1.906	0.358	0.053	0.007	bdl	bdl	bdl	0.063	0.011	0.024	1.892	41.19
JV11-13	Lusi	05-2011	1 km south west of Lusi crater, in the village, old well with bubbling gas, water. Not hot seepage	2		85.561	1.686	0.358	0.066	0.036	0.020	0.001	0.014	0.030	0.230	0.005	11.991	39.20
JV11-22	Lusi	05-2011	900 m west of Lusi crater, in the village, gas seepage from tube on the side of the road where old water well used to be	2		57.590	2.446	0.856	0.141	0.114	0.024	0.009	0.007	0.013	0.717	0.044	34.250	16.01
JV10-08	Wunut W-11S Zone D	02-2010	Sampling from pipe at Wunut gas field. Zone D: D10: 1120 ft MD – 1140 ft MD	3	nd	98.316	1.146	0.296	0.062	0.026	0.008	0.002	0.004	0.003	0.026	0.003	0.107	63.67
JV10-09	Wunut W-18LS Zone F-11	02-2010	Sampling from pipe at Wunut gas field. Zone F → 2200 ft MD – 2230 ft MD	3	nd	92.593	3.557	1.359	0.246	0.194	0.042	0.011	0.012	0.002	0.053	0.005	1.928	17.08
WU-1	Wunut	02-2010	From Wunut extraction plant. This pipeline merges production levels from JV10-08 to JV10-09	3	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
JV06-10	Gunung Anyar	09-2006	Seepages on flat area at the foot of MV	4	33.1	79.083	0.521	0.208	0.031	0.031	0.007	0.005	bdl	nd	nd	nd	18.377	98.33
JV08-11	Gunung Anyar	02-2008	Seepage inside the crater, oily fluids and mud seeping with gas	4	30.3	84.873	0.110	0.003	0.001	0.002	bdl	bdl	bdl	nd	nd	nd	15.011	731.82
JV06-12	Kalang Anyar	09-2006	Small seep outside main dam with microbial colonies around	4	30.6	95.542	0.106	0.001	0.006	bdl	bdl	bdl	bdl	nd	nd	nd	4.344	843.36
JV06-13	Kalang Anyar	09-2006	Small seep	4	31.2	96.793	0.115	0.001	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	3.091	835.03
JV06-16	Kalang Anyar	09-2006	Small seep	4	32.9	90.538	0.114	0.010	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	9.338	733.15
JV08-02	Kalang Anyar	02-2008	Large field of several pools where mainly water and gas seep out	4	31.0	96.034	0.111	0.009	0.001	0.001	bdl	bdl	bdl	nd	nd	nd	3.845	789.29
JV06-17	Pulungan	09-2006	Seepage with salt crusts and microbial colonies at crater site	4	29.3	95.643	0.102	0.001	0.000	bdl	bdl	bdl	bdl	nd	nd	nd	4.254	923.81
JV08-05	Gunung Sening	02-2008	Tall cone ~10 m high, dense mud erupted	4	27.7	99.571	0.050	0.000	0.000	bdl	bdl	bdl	bdl	nd	nd	nd	0.379	1975.72
JV08-07	Gunung Bulag	02-2008	8 m lake inside the crater where seepage occurs at 2 distinct points	4	31.1	97.411	0.123	0.001	0.000	bdl	bdl	bdl	bdl	nd	nd	nd	2.465	786.04
JV08-09	Pangangson	02-2008	One of the numerous pools inside the crater. 30 cm wide pool, mainly water and gas seeping	4	33.5	93.322	0.235	0.032	0.004	0.001	bdl	bdl	bdl	nd	nd	nd	6.404	341.24
JV08-10	Pangangson	02-2008	System of terraced pools on the east flank of the crater	4	34.6	92.390	0.262	0.035	0.006	0.003	0.001	0.001	bdl	nd	nd	nd	7.303	300.22
JV08-11	Bledug Kuwu	02-2008	1 m sized seepage inside the crater, bursts every ~1 min	4	31.1	16.243	0.041	0.008	bdl	bdl	bdl	bdl	bdl	nd	nd	nd	83.708	333.33
JV11-06	Kepulauan	05-2011	Source of hydrothermal water from ~90 m deep well	5	39.8	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
JV11-07	Songgoriti	05-2011	Source of hydrothermal water; vigorous seepage of gas, water	5	43.0	0.638	0.003	bdl	bdl	bdl	bdl	bdl	bdl	0.009	0.102	nd	99.248	224.55
JV11-09	Cangar	05-2011	Source of hydrothermal water	5	48.5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
JV11-10	Pacet	05-2011	Source of hydrothermal water mixed with river water	5	47.2	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd

bdl: below detection limit, nd: not determined.

<sup>a</sup> Approximate minimum temperature of steam clouds.

**Table 1B**

Isotopic composition of sampled gas. Isotopic data:  $\delta^{13}\text{C}$ : ‰, VPDB;  $\delta\text{D}$ : ‰, VSMOW; atm. R/Ra = (3He/4He)<sub>sample</sub>/(3He/4He)<sub>atmosphere</sub>; Ra =  $1.39 \times 10^{-6}$ ; nd: not determined.  $\diamond$  helium gas extracted from water bottles collected at seepage sites.  $\dagger$  indicates samples with He/Ne < 1.

Sample ID	Locality	Group	$\delta^{13}\text{C}_1$	$\delta\text{D}_1$	$\delta^{13}\text{C}_2$	$\delta^{13}\text{C}_3$	$\delta^{13}\text{C}_{i4}$	$\delta^{13}\text{C}_{n4}$	$\delta^{13}\text{C}_{\text{CO}_2}$	$^3\text{He}/^4\text{He}$ (R/Ra)
JV06-18	Lusi crater	1	−51.8	−207	nd	nd	nd	nd	−19.1	nd
JV06-18	Lusi crater	1	nd	nd	nd	nd	nd	nd	−21.0	nd
JV06-18	Lusi crater	1	nd	nd	nd	nd	nd	nd	−20.3	nd
JV07-03	Lusi crater	1	nd	nd	nd	nd	nd	nd	−19.8	nd
JV07-03	Lusi crater	1	−40.5	nd	nd	nd	nd	nd	−13.1	nd
JV07-03	Lusi crater	1	−40.4	nd	nd	nd	nd	nd	−19.6	nd
JV07-05	Lusi crater	1	nd	nd	nd	nd	nd	nd	−18.8	nd
JV08-13	Lusi crater	1	nd	nd	nd	nd	nd	nd	−18.7	nd
JV08-13	Lusi crater	1	nd	nd	nd	nd	nd	nd	−20.5	nd
JV08-14	Lusi crater	1	nd	nd	nd	nd	nd	nd	−23.5	nd
JV10-04	Lusi crater	1	nd	nd	nd	nd	nd	nd	nd	nd
JV10-05	Lusi crater	1	nd	nd	nd	nd	nd	nd	nd	nd
JV10-06	Lusi crater	1	nd	nd	nd	nd	nd	nd	nd	nd
JV040706	Lusi	2	nd	nd	nd	nd	nd	nd	nd	nd
JV130706	Lusi	2	nd	nd	nd	nd	nd	nd	nd	nd
JV06-07	Lusi	2	−48.6	−293	−26.8	−25.4	nd	nd	−15.0	nd
JV07-06	Lusi	2	−40.1	nd	−26.8	−24.7	−25.2	−23.6	−24.3	nd
JV07-07	Lusi	2	−40.0	nd	−26.6	−24.7	−25.1	−23.6	−20.0	nd
JV10-07	Lusi	2	−35.74	−171.8	−20.2	nd	nd	nd	−6.0	5.53
JV11-03	Lusi	2	nd	nd	nd	nd	nd	nd	−7.4	2.79 $\dagger$
JV11-04	Lusi	2	nd	nd	nd	nd	nd	nd	−13.9	nd
JV11-12	Lusi	2	nd	nd	nd	nd	nd	nd	−7.4	5.01 $\diamond$
JV11-13	Lusi	2	nd	nd	nd	nd	nd	nd	−0.5	6.47
JV11-22	Lusi	2	nd	nd	nd	nd	nd	nd	−2.3	nd
JV10-08	Wunut W-1LS Zone D	3	−38.98	−195.6	−20.9	−18.0	nd	nd	nd	nd
JV10-09	Wunut W-18LS Zone F-11	3	−38.34	−186.8	−25.2	−23.5	−25.6	−22.7	13.3	nd
WU-1	Wunut	3	nd	nd	nd	nd	nd	nd	nd	1.37
JV06-10	Gunung Anyar	4	−56.0	−191	nd	nd	nd	nd	−3.8	nd
JV08-01	Gunung Anyar	4	−54.7	−201	nd	nd	nd	nd	−6.3	nd
JV06-12	Kalang Anyar	4	−55.0	−214	nd	nd	nd	nd	−9.9	nd
JV06-13	Kalang Anyar	4	−55.7	−214	nd	nd	nd	nd	−6.1	nd
JV06-16	Kalang Anyar	4	−55.0	−216	nd	nd	nd	nd	−4.0	nd
JV08-02	Kalang Anyar	4	−48.4	−219	nd	nd	nd	nd	−16.5	nd
JV06-17	Pulungan	4	−54.1	−207	nd	nd	nd	nd	−5.1	nd
JV08-05	Gunung Sening	4	−39.5	−172	nd	nd	nd	nd	nd	nd
JV08-07	Gunung Bulag	4	−33.5	−159	nd	nd	nd	nd	−8.8	nd
JV08-09	Pangangson	4	−52.6	−233	nd	nd	nd	nd	−0.3	nd
JV08-10	Pangangson	4	−52.1	−236	nd	nd	nd	nd	−4.1	nd
JV08-11	Bledug Kuuwu	4	−54.2	−211	nd	nd	nd	nd	−18.8	nd
JV11-06	Kepulauan	5	nd	nd	nd	nd	nd	nd	nd	3.69 $\diamond$ $\dagger$
JV11-07	Songgoriti	5	nd	nd	nd	nd	nd	nd	10.7	2.75
JV11-09	Cangar	5	nd	nd	nd	nd	nd	nd	nd	3.00 $\diamond$ $\dagger$
JV11-10	Pacet	5	nd	nd	nd	nd	nd	nd	nd	2.95 $\diamond$ $\dagger$

differential molecular adsorption on the solid grains of the mud and differential solubility, is a common phenomenon observed in many seeps worldwide (Etiope et al., 2009a). This is typical for gas seeping at vents with relatively low flux and/or high mud content (Etiope et al., 2011b), as those of Gunung Sening and Gunung Bulag. We exclude that the higher  $C_1/(C_2 + C_3)$  ratio may be due to secondary methanogenesis (Milkov, 2011) as  $\text{CO}_2$  from Gunung Bulag is not particularly  $^{13}\text{C}$  enriched (as it happens for residual  $\text{CO}_2$  during methanogenesis; e.g. Etiope et al., 2009b), and it is similar to the other samples. All other mud volcanoes analysed in this study released a mixed gas.

The hydrocarbon composition of the Lusi gas is essentially the same either sampled at the crater or at satellite seeps. The alkanes gas plot ( $\delta^{13}\text{C}$  vs inverse carbon number; based on the model by Chung et al., 1988; Fig. 4) also highlights that the Lusi gas is very similar to that of Wunut, with a major microbial component in 2006 only. The ethane vs propane maturity plot (Berner and Faber, 1996; Fig. 5A) was applied using kerogen precursors with  $\delta^{13}\text{C}$  of  $-26\%$  for terrestrial organic matter (Type III) and from  $-30$  to  $-26\%$  for marine organic matter (Type I–II). These values were selected considering that East Java Basin source rocks have terrestrial and marine kerogen with an average  $\delta^{13}\text{C}$  value around  $-26\%$  and  $-28\%$ , respectively (Satyana and Purwaningsih, 2003). Fig. 5A suggests that both Lusi and Wunut alkanes actually derive from marine

kerogen, with maturity around 1.5–2%Ro (if kerogen has  $\delta^{13}\text{C}$ :  $-28\%$ ) or 2–2.5%Ro (if kerogen has  $\delta^{13}\text{C}$ :  $-30\%$ ). In any case, these source rocks and maturities are different from those (terrestrial, Type III at about 0.6%Ro) attributed to the mud released at Lusi (Mazzini et al., 2007; Plumlee et al., 2008). Moreover, small amounts of fresh and nonbiodegraded oil were found in the erupted mud (Plumlee et al., 2008), and this further supports the hypothesis that marine kerogen, which is more oil-prone than terrestrial, is the source for Lusi hydrocarbons. This shows that the source of the Lusi gas is different (and deeper) than that of the erupted mud. Thus after 2006, the Lusi gas had a limited or no contribution from the Upper Kalibeng shales.

The hypothesis of a marine source rock with maturity  $> 1.5\%$ Ro, suggested by the maturity plot in Fig. 5A, is supported by an independent interpretative tool (Fig. 5B) based on thermogenic gas generation modelling (GOR–Isotopes modelling, Etiope et al., 2011a; Tang et al., 2000). GOR (Gas–Oil–Ratio)–Isotopes is a PC-based simulator to predict gas and oil generation, including the primary carbon isotopic composition of methane, based on the mathematical model developed by Tang et al. (2000). The combination of  $\delta^{13}\text{C}_2$  and  $\delta^{13}\text{C}_3$  produced by marine kerogen, suggests that Lusi gas was generated at temperature of 210–220 °C, corresponding to a maturity of 2%Ro. This is similar to that derived in Fig. 5A for a precursor kerogen with  $\delta^{13}\text{C} = -30\%$ .

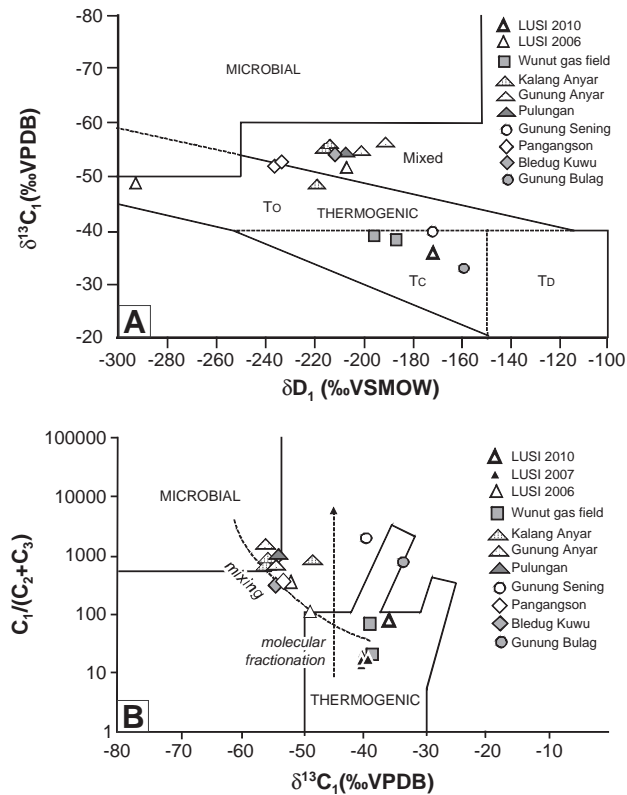


Fig. 3. Genetic zonation diagrams of methane (A: Schoell plot; Schoell, 1983) (B: Bernard plot; Bernard et al., 1978). To: thermogenic with oil; Tc: thermogenic with condensate; Td: dry thermogenic.

Regardless the value of the  $\delta^{13}C$  of the kerogen (most likely from  $-28$  to  $-30$ ‰), marine source rocks are present in the oil-prone Eocene Ngimbang shales (e.g. Satyana and Purwaningsih, 2003; Wiloso et al., 2009). Considering the geothermal gradient of  $42$  °C/km (Mazzini et al., 2007), the gas generation temperature derived by GOR Isotopes modelling would suggest a source rock located at about  $4400$  m, which is consistent with the location of the Ngimbang Formation (see Fig. 6). The modelling also suggests that for  $\delta^{13}C_2$  of about  $-25$ ‰ (average value of Lusi and Wunut), the original  $\delta^{13}C_1$  of thermogenic methane would be around  $-35$ ‰ (Fig. 5C). This is consistent with the highest value recorded at Lusi (sample JV10-07, a cold seep outside the crater).

The shallow reservoir of the Wunut gas field ( $\sim 350$ – $700$  m deep) should accordingly be considered accumulations along a seepage

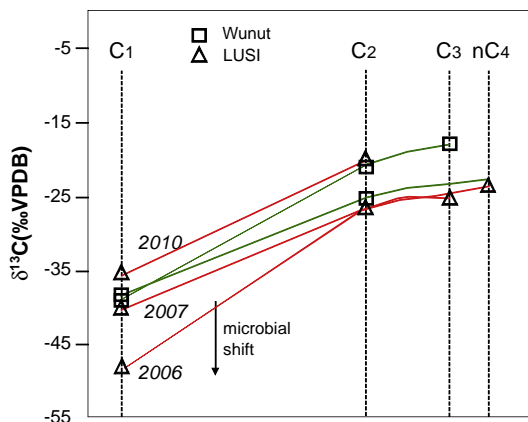


Fig. 4. Natural gas plot, with the  $\delta^{13}C$  sequence of  $C_1$ – $C_4$  alkanes of Lusi and Wunut gas field data.

system, linked to a deeper main reservoir. This is in agreement with Kusumastuti et al. (2000) who suggested that the Wunut field consists of thermogenic gas migrating from the deep Porong Reef reservoir. This is also consistent with the positive value of  $\delta^{13}C$  of  $CO_2$  ( $+13.2$ ‰) that suggests  $CO_2$  reduction in secondary methanogenesis (see Tables 1A, 1B) and hydrocarbon biodegradation (heavier  $\delta^{13}C_3$ ), typical of shallow pools (Etiopie et al., 2009b; Milkov, 2011).

Considering the Ngimbang shales as the source rocks of Lusi and Wunut, the main gas reservoirs are likely to be part of the Kujung sequence including Prupuh or most likely Tuban limestones (Ardhana,

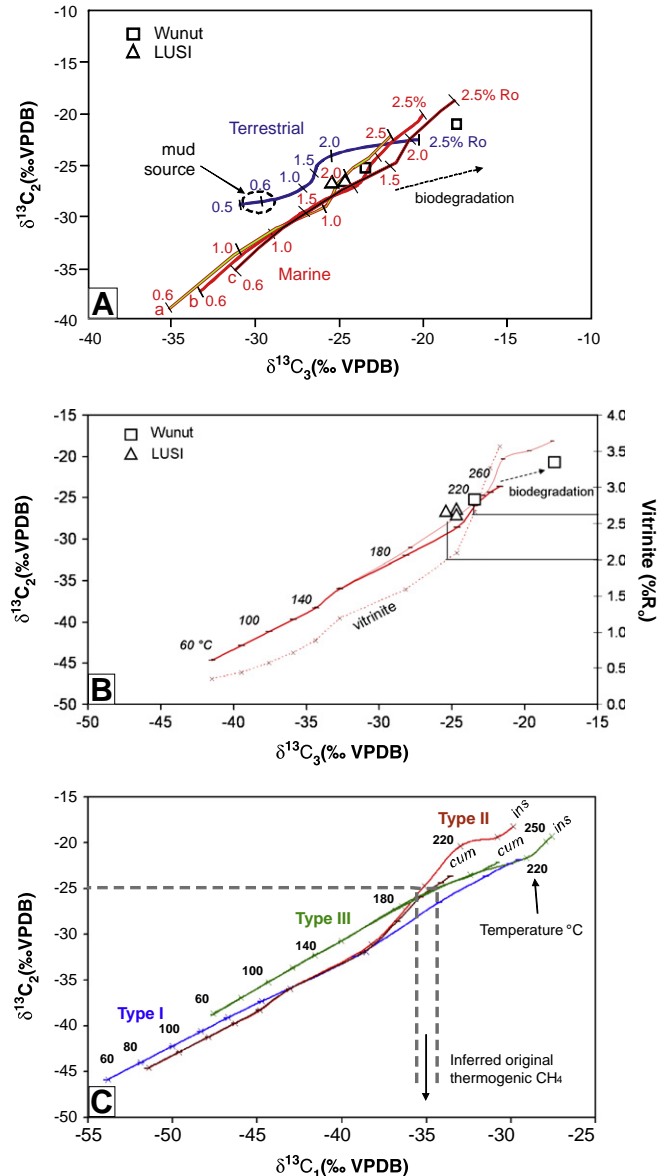
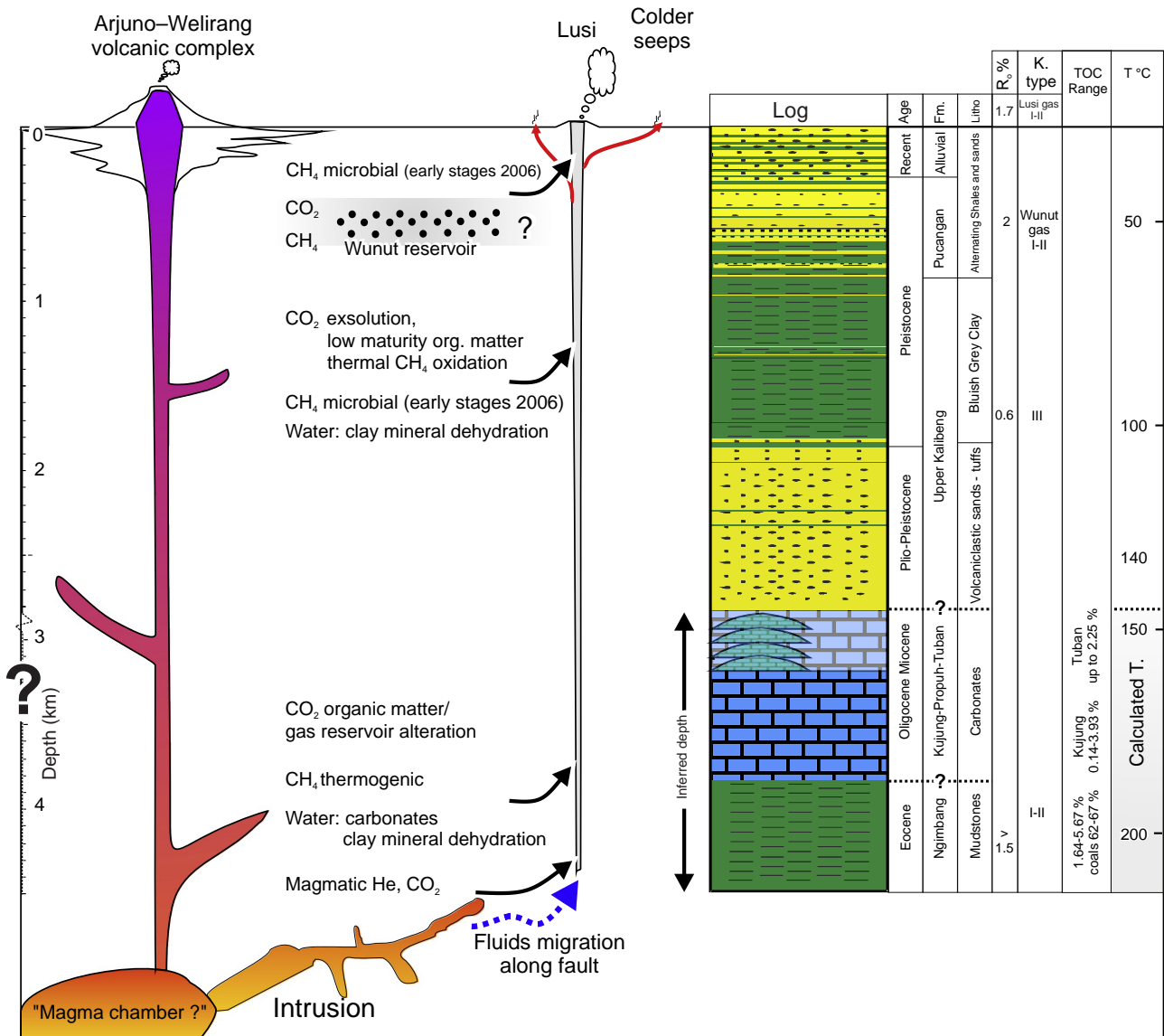


Fig. 5. (A) Ethane vs. propane maturity plot (vitrinite reflectance, % Ro; model by Berner and Faber, 1996). Initial carbon isotope value of marine (Type I–II) kerogen is taken as (a)  $-30$ ‰; (b)  $-28$ ‰ (that is the mean value for East Java Basin source rocks) and (c)  $-26$ ‰. The terrestrial (Type III) kerogen precursor is taken as  $-26$ ‰ (mean for terrestrial East Java Basin kerogen). "Mud source" indicates the conditions of the Lusi mud (Upper Kalibeng shales; Mazzini et al., 2007). (B) Thermogenic gas formation modelling for ethane vs. propane, from default Type II kerogen (calculated using Geolsochem Corp. GOR-Isotopes software 1.94; heating rate of  $5$  °C per million year; Tang et al., 2000). Bold and thin lines refer to cumulative and instantaneous gas generation, respectively. (C) Same modelling for ethane vs. methane from default Type I–III kerogen (calculated as in B). Maturity values estimated independently in (A) and (B) are similar, and the measured isotopic composition of ethane ( $\delta^{13}C_2$ ) would correspond to methane with  $\delta^{13}C_1$  around  $-35$ ‰, which is consistent with that measured in the Lusi crater (see Tables 1A, B).





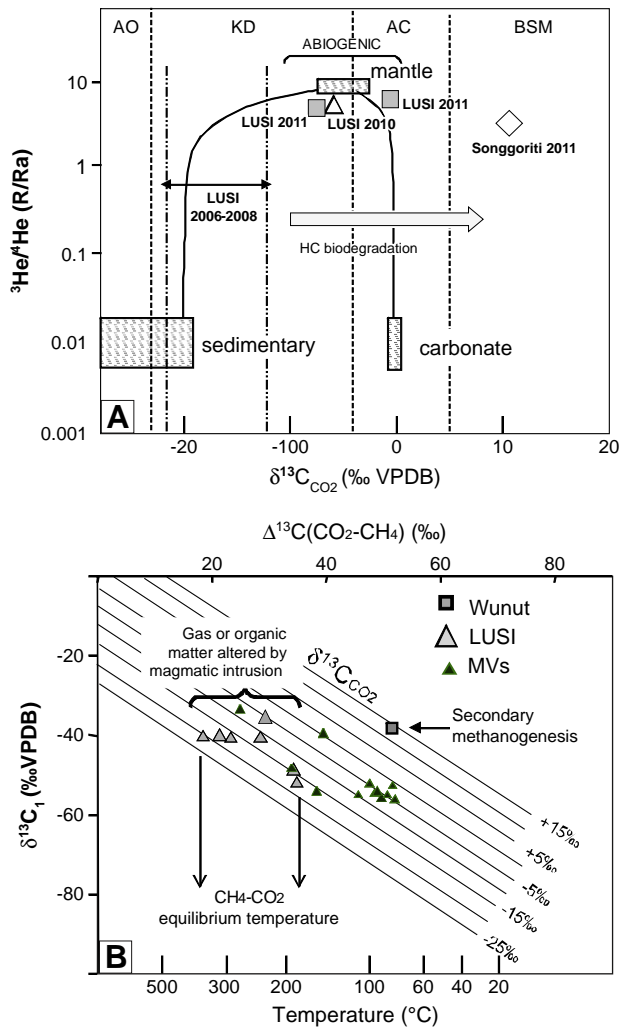
**Fig. 6.** Schematic cartoon summarising the known stratigraphy at Lusi site and depicting the Lusi plumbing system with the fluids contributions from different depths. Geochemistry shows the evidence of a deeper sited magmatic intrusions and/or hydrothermal fluids that feeds the Lusi system. This link may also explain why Lusi activity still increases after earthquakes even when occurring at significant distances. We suggest that these earthquakes likely affect the plumbing system of the magmatic chamber at depth resulting in overpressure buildup that periodically enhances Lusi eruption. TOC values from the East Java Basin are extracted from *Satyana and Purwaningsih (2003)*.

1993) (see Fig. 6), as they all represent a main regional reservoir of the petroleum systems in North East Java Basin (Doust and Noble, 2008; Kusumastuti et al., 2000, 2002). Lusi would then be part of the seepage system belonging to the so-called Ngimbang–Kujung petroleum system (Doust and Noble, 2008; Essam Sharaf et al., 2005).

The disappearance over time of the microbial components of the Lusi gas (after 2006) can then be explained by the ascending gas from the deeper units initially mixed with biogenic gas present at shallower depths (Upper Kalibeng shales and/or alluvium clays; Fig.6); then the microbial component in the seepage channel was exhausted and the thermogenic gas flow dominated. To summarise, the initial mixing-microbial component should be considered as a “contamination” during early gas migration. In comparison with the other mud volcanoes of NE Java Basin, Lusi seems to have hydrocarbons of deeper origin, or in any case a seepage system less influenced by shallower microbial components. The lack of isotopic data for C<sub>2</sub>+alkanes for the surrounding mud volcanoes prevents an evaluation of the type and maturity of their source rocks.

6.2. Carbon dioxide and helium

The origin of the Lusi CO<sub>2</sub> is clearly organic, with δ<sup>13</sup>C<sub>CO2</sub> ranging from −13‰ to −23‰; although three samples showed a values ranging from −7.4 to −0.5‰, associated with a high helium isotopic ratio (R/R<sub>a</sub>: up to 6.47; Fig. 7A) hence, apparently, a magmatic gas. However, <sup>13</sup>C-enrichment could also result from reduction of organic CO<sub>2</sub>, as typically occurs during secondary methanogenesis (Etiope et al., 2009b; Milkov, 2011). Organic CO<sub>2</sub> in petroleum systems typically have concentrations below 10 vol.% (Jenden et al., 1993) and higher concentrations were always associated with inorganic <sup>13</sup>C-enriched CO<sub>2</sub>, as also occurs in some CO<sub>2</sub>-rich gas fields in Indonesia (Cooper et al., 2006). Therefore, the apparent very high organic CO<sub>2</sub> concentration of Lusi (up to 89 vol.%) is unusual. The strong discrepancy observed in the CO<sub>2</sub> content at localities of Groups 1 (crater zone) and 2 (satellite seeps) can be interpreted as the result of much lower temperatures recorded at the sites away from the crater zone, where CH<sub>4</sub> is clearly dominating. Here, the rising gas diverges



**Fig. 7.** (A) He isotopic ratios vs.  $\delta^{13}\text{C}_{\text{CO}_2}$ . Dotted lines indicate minimum and maximum values of  $\delta^{13}\text{C}_{\text{CO}_2}$  (analysed in 2006, 2007 and 2008; R/Ra not available). Carbon dioxide isotope zonation after Etiope et al. (2011c). AO, aerobic hydrocarbon oxidation; KD, kerogen decarboxylation; AC, alteration of marine carbonates; BSM, biodegradation and secondary methanogenesis. (B)  $\delta^{13}\text{C}_{\text{CH}_4}$  versus difference in stable carbon isotope composition [ $\delta^{13}\text{C}(\text{CO}_2\text{-CH}_4)$ ] between carbon dioxide and methane of Lusi, Wunut and other MV gases. Relation between fractionation of stable carbon isotope and temperature from Bottinga (1969).

from the main conduit and has sufficient time to cool hence partly buffering the  $\text{CO}_2$  exsolution mechanism that is dominating in the crater zone (i.e. resulting in  $\text{CO}_2$  dominated gas). Mazzini et al. (2007) proposed that this exsolution mechanism operates where temperatures exceed  $100^\circ\text{C}$  (i.e. at 1700 m and below) along with the fast rise of fluids to the surface with a sudden pressure drop (Fig. 8).

However, the high  $\text{CO}_2/\text{CH}_4$  ratio, found in high temperature crater samples, can also be explained by an increased  $\text{CO}_2$  generation by thermal  $\text{CH}_4$  oxidation or alteration of organic matter (e.g. Seewald et al., 1994).  $\text{CH}_4$  oxidation typically would give  $\delta^{13}\text{C}_{\text{CO}_2}$  lower than  $-25\text{‰}$  (Clayton, 1995; Jenden et al., 1993) but thermal degradation of organic matter can produce  $^{13}\text{C}$ -enriched  $\text{CO}_2$  with  $\delta^{13}\text{C}_{\text{CO}_2}$  up to  $-8\text{‰}$  (Seewald et al., 1994). Assuming equilibrium conditions after  $\text{CH}_4$  oxidation to  $\text{CO}_2$  (as reported for thermal alteration of organic-rich sediments by Seewald et al., 1994), and that  $\text{CO}_2$  and  $\text{CH}_4$  are not mixed from different sources, the formation temperature of Lusi  $\text{CO}_2$  and  $\text{CH}_4$  would be between  $200$  and  $400^\circ\text{C}$  (Fig. 7B). This temperature is higher than that derivable for the other mud volcanoes of this study. These data, combined with the strong mantle

helium signature, point towards the presence of sub-volcanic intrusions or migrating hydrothermal fluids affecting the source and/or reservoir rocks below Lusi.

We cannot exclude that a component of the  $\text{CO}_2$  is derived from calcite thermometamorphism ( $\delta^{13}\text{C}$  around  $0\text{‰}$ ) (e.g. Sano and Marty, 1995). In this case, the Lusi  $\text{CO}_2$  could be explained by a mixing of these inorganic and organic ( $\text{CH}_4$  or shale alteration) components.

The fact that Wunut and Lusi can be considered as two separate systems, is supported by the different He isotopic values (Wunut: R/Ra  $\sim 1.4$  and He/Ne  $\sim 2$ , indicating crustal signature with slight mantle component vs Lusi: R/Ra up to 6.47 and He/Ne  $\sim 22$  indicating mantle signature).

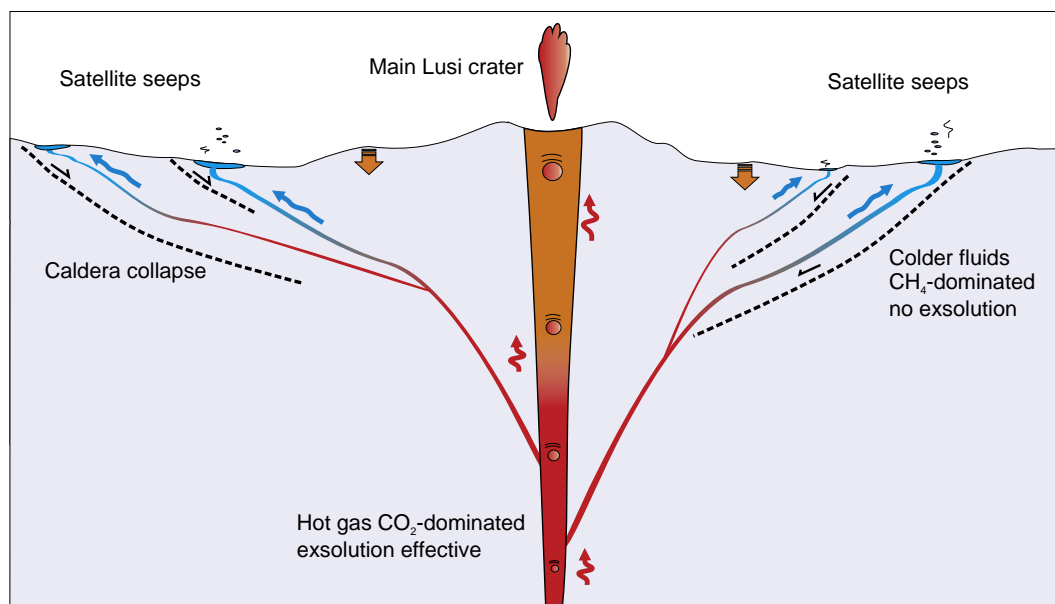
The other mud volcanoes of this study release a more  $^{13}\text{C}$ -enriched  $\text{CO}_2$ , with values from  $-0.3$  to  $-18.8\text{‰}$ . Some of these values are similar to those reported for geothermally-perturbed mud volcanoes of Alaska (Motyka et al., 1989) and sediment-hosted seepage near Salton Sea in California (Mazzini et al., 2011), where  $\text{CO}_2$  was interpreted as predominantly magmatic, mixed with a minor component from thermometamorphism of limestones. In this respect, the Lusi gas is different from that of the nearby mud volcanoes; it drains a different and more pressurised system (leading to higher fluid flows) where deep source rocks and gas are thermally altered. The Davis-Schrimp seep field in the Salton Sea (with magmatic  $\text{CO}_2$  and He, and thermogenic  $\text{CH}_4$ ; Mazzini et al., 2011) is probably the closest analogue to the Lusi system. The Salton Sea magmatic intrusions affect mainly the carbonate rocks therefore the resulting  $\text{CO}_2$  is much heavier and undistinguished from magmatic  $\text{CO}_2$ . In the Sidoarjo area the intrusions, or the migration of hot fluids, likely perturbed mainly shales (kerogen) and gas ( $\text{CH}_4$ ), leading to larger amounts of isotopically light  $\text{CO}_2$ , that likely masks eventual magmatic  $\text{CO}_2$ .

The link to the nearby volcanic complex is stressed by a) the apparent N-NE trend of the volcanic vents in this part of Java (Fig. 1), b) the overall chronology within this trend, and c) by the presence of lahars baked by hydrothermal fluids in the exposed Watukosek fault escarpment 4.6 km from Lusi. Our 2011 fieldwork identified the presence of altered lahars that follow the direction of the Watukosek fault indicating that it was acting as a preferential pathway for hydrothermal fluid migration towards the north east. If the arc volcanism will migrate further towards Lusi is an open question, but in any case the whole volcanic system has evolved chronologically from Mount Kawli (the oldest), to Arjuno Welirang and finally to Penanggungan (the youngest volcano) following the same direction as the Watukosek fault (Fig. 1).

### 6.3. Towards a new model of Lusi seepage system

The presented data converge towards a scenario characteristic of sediment-hosted hydrothermal-magmatic system, where igneous intrusions and hydrothermal fluids alter organic matter in deep sediments (e.g. Simoneit, 1985), as in the case of "shale subduction" (e.g. Furi et al., 2010). This scenario may also explain the geochemical characteristics of the hot crater water and the signature of clay mineral transformation of the Kalibeng shales (Mazzini et al., 2007). Our new data clearly show the presence of a much deeper plumbing system reaching the deep limestones and the source rocks. Fig. 6 summarises the new seepage model for Lusi.

Rather than representing a traditional mud volcano, we suggest that Lusi is a surface manifestation of a deep-seated sediment-hosted hydrothermal system where the vertical migration of fluids and mud is analogue to so-called hydrothermal vent complexes (Svensen et al., 2004, 2006). Hydrothermal vent complexes represent piercement structures formed as a consequence of thermally induced fluid flow and pressure build-up and are associated with igneous intrusions in sedimentary basins (Jamveit et al., 2004). The association of the deep gas with shallower mobilised shale (Kalibeng Fm.)



**Fig. 8.** Cartoon not to scale of the shallow plumbing system at Lusi site. The fluids rising through the main conduit have high temperature (at least 100 °C) and are CO<sub>2</sub> dominated. During the fast rise from units with high T and P, the fluids cannot cool significantly and the sudden pressure drop allows the exsolution mechanism to be very efficient. This implies that the CO<sub>2</sub> dissolved in the interstitial waters is liberated as gaseous phase once it reaches the surface. The satellite seeps, located along the Watukosek fault trend or on the rims of the caldera collapse, are CH<sub>4</sub> dominated with low temperatures. The fluids seeping at these locations branch off from the main conduit and have sufficient time to cool down hence interrupting the exsolution mechanism.

suggests that the first pressure driver of the seepage system is the gas itself, associated with hot water in the main feeder channel. This means that the Lusi system may be controlled by deep fluid pressure conditions, likely linked to a thermal effect from the intrusive magmatic bodies. In this respect the adjacent Arjuno–Welirang magmatic volcano would play an important role.

Therefore, an important consequence of the new findings is that either invoking an “earthquake–fault” or a “man-made” triggering hypothesis for the Lusi eruption, a third factor has to be considered: the role and plumbing system of the adjacent magmatic volcano and the high heat flow induced by an intrusion. This type of interactions between volcanoes and sedimentary basins is probably not unique for Lusi. A screening of seismic data acquired in the NE back arc basin of Java, shows numerous buried piercement structures. One of the most spectacular is the collapse structure that is visible through the Porong 1 well in the Lusi vicinity (Istadi et al., 2009). This structure likely represents an extinct mud volcano or a piercement structure like Lusi that, once it terminated its activity, gradually collapsed around a vertical feeder channel and got buried.

The mechanisms ongoing at Lusi and the described hydrothermal scenario could be used as a modern analogue to understand the mechanisms of the hydrothermal vent complexes that are linked to environmental crises in the geological record. Recent studies have highlighted the correlation between Large Igneous Provinces developing in the geological record and periods of global warming/environmental crises (e.g. Wignall, 2001). It has been proposed that these climatic crises were triggered by extensive hydrothermal activity and formation of piercement structures (Svensen et al., 2004; Svensen et al., 2006).

#### 6.4. Seismicity and the volcanic complex

There are several implications to the newly proposed link between Lusi and the Arjuno magmatic complex and in particular regarding how the frequent seismic activity may affect this setting in critical conditions.

In contrast to Manga et al. (2009) that measured the strength of mud erupted at Lusi site, Tanikawa et al. (2010) measured also the

permeability and specific storage of outcrops of the Upper Kalibeng Formation (the source of the Lusi mud) coupled with various parameters of well data logged during the drilling of the same formation. Tanikawa et al. (2010) demonstrate that the Upper Kalibeng Formation prior to the eruption was overpressured and in critical conditions, thus prone to lose strength resulting in liquefaction even by moderate perturbations and/or stress field fluctuations (i.e. like those induced by the Yogyakarta earthquake). In that respect, Mori and Kano (2009) described evidence showing that the 6.3 M 27-05-2006 earthquake may have altered the local fluid conditions in the Lusi region. These authors also highlighted the possible effect of cascading mechanisms that can amplify fluid pressure changes resulting in surface eruption triggered by an initial seismic activity. These scenarios seem to be applicable to the described system of Lusi–Watukosek fault–Arjuno Welirang volcanic complex.

The new scenario implies that the analysis of the seismic effects at Lusi locality (e.g. Manga, 2007) should be revised by considering the catalytic role of the volcano. Further, it has been demonstrated that the reactivated strike-slip Watukosek fault played a crucial role in facilitating the Lusi eruption via reducing the critical fluidization pressure (Mazzini et al., 2009). Our new findings suggest that an eruption in this region was even more likely to occur considering the potential effect of an intrusion and/or the overpressured fluids generated by it in an already critical plumbing system.

Recent works stressed the link between seismicity and volcanic and hydrothermal activity. For example Delle Donne et al. (2010) tried to quantify the response in the activity of magmatic chambers and the heat flux of volcanoes following to large earthquakes. They demonstrated that even distant earthquakes have a significant effect over magmatic systems. The large database collected by Delle Donne et al. (2010) also showed that the distance between the 6.3 M 27-05-2006 earthquake (when the venting started in the Sidoarjo area) and the Arjuno complex, is well within their sensitivity threshold. Indeed it has been documented that the same earthquake triggered responses in activity at Javanese volcanoes (e.g. Merapi and Semeru) located away from the epicentre, one at even greater distance than the Arjuno complex (Harris and Ripepe, 2007; Walter et al., 2007). Therefore it is clear that the frequent seismic activity

occurring in the region has a strong impact on unstable systems like the one described herein. *Delle Donne et al. (2010)* also pointed out that the observed responses of volcanic complexes occur generally within 21 days of the triggering earthquake, but the large majority of complexes (67%) responded within 5 days. Interestingly, the venting activity in Sidoarjo occurred 2 days after the 6.3 M earthquake therefore within the same time window indicated by *Delle Donne et al. (2010)*.

Since 2006, Lusi responded to seismic activity in numerous instances behaving with sudden bursts and increases in flow rate and even the Watukosek fault has been reactivated with spectacular sudden collapse and shearing (*Mazzini et al., 2007, 2009*). To date (November 2011) the most recent response of Lusi to seismic activity occurred after the 26-04-2011 5.4 M Javanese earthquake (Cilacap coast) resulting in two separate and distinct craters one hour after the event (*Fig. 2D*). Additionally, 5 h after the event one of the large containment dams underwent severe contraction (*Fig. 2E*) resulting in breaking (S. Hadi, pers. comm.). Interestingly the dam broke on the north-easternmost part following the trend of the Watukosek fault system. All these evidences are supporting a scenario where the seismic activity periodically alters the conditions of the deep magmatic fluids that, in turns, may trigger the more powerful eruptions at Lusi site and reactivates the Watukosek fault. The lateral shearing along the Watukosek fault also contributes to facilitate this overpressure release.

#### 6.5. A new triggering scenario

Our new findings open new implications and scenarios for understanding the Lusi plumbing system and its triggering.

- Deep sited magmatic intrusions or hot hydrothermal fluids originating from the conduit of the Arjuno volcanic complex extend towards the north-east migrating through organic-rich source rocks and reservoir sedimentary sequences. The generally high geothermal gradient of the region supports this scenario. The whole volcanic complex may extend towards the north east following the Watukosek fault system trend (*Fig. 1*).
- The presence of the Watukosek fault system (that originates from the volcanic complex) represents an ideal pathway for the propagation of deep magmatic fluids towards the backarc region of the island.
- Great overpressures are generated in the new hydrothermal system. The heat from the intrusion and its fluids produces CO<sub>2</sub> from the organic-rich sediments and/or thermogenic gas, pushing the fluids towards the already overpressured and unstable shallower shales (i.e. Upper Kalibeng Fm.).
- A subsurface dome-shaped feature grows underneath the location of the future Lusi crater (*Mazzini et al., 2009*).
- 27th of May 2006: 6.3 M earthquake and reactivation of the strike-slip fault. Simultaneously, the magmatic chamber underneath the volcanic Arjuno complex is affected and in turn the migration of magma and fluids alters the critical equilibrium of the already formed and overpressured growing piercement structure beneath Lusi site.
- Fluids from the sedimentary units are drained towards the shear zone and migrate towards the surface using the fault zone as preferential pathway. Numerous vents form on the surface forming a 1.2 km alignment that follows the NE–SW fault direction.
- The mud flow from a prominent crater (later called Lusi) covers the other eruption sites.
- In several instances Lusi behaviour seemed to respond with abrupt increase in flow rates after seismic events. The collapse in the region has an ellipsoidal shape that follows the direction of the Watukosek fault.

We suggest that deep fluid pressure build-up is the main driver and co-trigger of the Lusi eruption. We cannot exclude that these pressure variations may also be independent of earthquakes as they directly respond to the volcanic evolution and behaviour of the volcanic complex, as also observed in other hydrothermal systems. Moreover we cannot exclude a man-made contribution to trigger the Lusi phenomena, however, based on our findings, it would appear unlikely that a shallow borehole may perturb a >4 km deep thermogenic and magmatic system.

#### 6.6. Predictions of Lusi longevity

Our new scenario stresses that predictions of the Lusi longevity based on a mud volcano approach, especially if only mud is considered to drive the fluid flow, are doomed to fail. A predictive model should firstly understand the structure of Lusi plumbing system and then take into account the combined effect of seismicity that may alter the Arjuno magmatic chamber, periodically reactivates the Watukosek fault, and, in turn, affects the rheology of the sediments and the critical and overpressured plumbing system of the Lusi piercement. If our new model scenario is correct then the published models (*Davies et al., 2011; Rudolph et al., 2011*) attempting to predict the evolution of Lusi are incomplete and irrelevant. Even today (November 2011), field observations show flow rates and level of activity that are both much lower when compared with the predictions of the above models. Since several months Lusi has been erupting constant flow rates of ~5000–10,000 m<sup>3</sup>/d. Additionally a geyser-like activity with periods (up to 16 min) of no eruption and completed calm has been observed (i.e. since April 2011, S. Hadi pers. comm.). So far, the periodicity of these no activity periods has not yet been investigated. Besides these daily fluctuations, peaks of much higher flow rate discharge were observed. These lasted only few days and appeared in large part coinciding with recorded seismic activity.

Future studies should aim to monitor the variation of the behaviours of the volcanic complex and the responses in Lusi activity.

### 7. Conclusions

- Molecular and isotopic composition of hydrocarbon alkanes, carbon dioxide and helium, combined with maturity plot and thermogenic gas formation modelling, indicate that Lusi's system is deeper than what was previously assumed and fluids are thermogenically produced in source rocks more than 4 km deep (e.g. Ngimbang Fm.).
- Isotopically light CO<sub>2</sub> (<–14‰) and CH<sub>4</sub>–CO<sub>2</sub> equilibrium at temperatures above 200 °C are suggestive of thermal alteration of hydrocarbons or organic matter. The strong helium magmatic signature (R/Ra: 5.5) further supports the hypothesis of a deeper sited intrusions that bifurcates from the neighbouring Arjuno–Welirang magmatic complex affecting source and reservoir rocks.
- This scenario would explain the local high geothermal gradient and it would be in agreement with the presence of a growing piercement structure as observed in seismic profiles from the 80s (*Mazzini et al., 2009*).
- In the Lusi hydrothermal system a deep heat anomaly appears to be the main driving force, generating CO<sub>2</sub> from organic material (marine shales), creating deep over-pressures, and altering deep hydrocarbon reservoirs. In this respect, the term “mud volcano” for Lusi may be misleading, and Lusi cannot be a representative example of the birth of a traditional mud volcano. Indeed Lusi is likely part of a larger sediment-hosted hydrothermal system related to the adjacent volcanic complex on the south west.
- The deep fluids then migrated upward and mobilised the shallower shales. These shales were already in critical, over-pressured conditions, which is very common in the area, as shown by diffuse mud volcanism.

- Lusi's longevity may be linked to the evolution and fluid pressure build-up in the Arjuno–Welirang magmatic complex which in turn may be influenced by seismic activity and by the strike slip movement of the Watukosek fault. Any model aiming to predict the eruption longevity should implement the new results and model scenario.

The hydrothermal scenario would also open up new prospectives for geothermal energy production at this site, and thus transforming it into a resource for the community rather than a mere disaster.

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