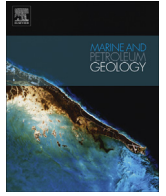




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## Research paper

## Radon and carbon gas anomalies along the Watukosek Fault System and Lusi mud eruption, Indonesia

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## ABSTRACT

An extensive survey was carried out in the Sidoarjo district (East Java, Indonesia) to investigate the gas leaking properties along fractured zones coinciding with a strike-slip system, the Watukosek Fault System (WFS) in NE Java. This structure has been the focus of attention since the beginning of the spectacular Lusi mud eruption on the 29<sup>th</sup> May 2006. The sinistral strike-slip WFS originates from the Arjuno-Welirang volcanic complex, intersects the active Lusi eruption site displaying a system of anti-thetic faults, and extends towards the NE of Java where mud volcanic structures reside.

In the Lusi region we completed a geochemical survey along three profiles combining measurements of a) <sup>220</sup>Rn and <sup>222</sup>Rn activity, b) CO<sub>2</sub> and CH<sub>4</sub> soil gas content, c) CO<sub>2</sub> and CH<sub>4</sub> fluxes, and d) gas analyses. The profiles are up to ~1.2 km long and intersect perpendicularly areas with intense fracturing and surface deformation along the WFS. The purpose was to investigate the presence and origin of soil degassing activity in potentially active fault zones.

Results show that the peripheral sectors of the profiles have high <sup>220</sup>Rn activity and reduced CO<sub>2</sub> and CH<sub>4</sub> fluxes and concentrations. This suggests low fluids migration that could be affected by shallow circulation. In contrast, the segments of profiles intersecting the fractured zones have the highest <sup>222</sup>Rn activity, CO<sub>2</sub> and CH<sub>4</sub> flux and gas concentration values.

The relationship existing among the measured parameters suggests that the WFS acts as a preferential pathway for active rise of deep fluids. The presence of such advective processes is suggested by the relatively high rate of migration needed to obtain anomalies of short-lived <sup>222</sup>Rn in the soil pores. Gas molecular and isotopic composition reveals that all sampled localities have a mixed hydrocarbon origin implying the presence of shallow microbial and deeper thermogenic hydrocarbons. CO<sub>2</sub> isotopic values ( $\delta^{13}\text{C}_{\text{CO}_2}$  ranges between –9.48‰ and 4.12‰ V-PDB) indicate the presence of mantle derived CO<sub>2</sub> and thermo-metamorphic CO<sub>2</sub> suggesting that elevated temperatures have a key role in this active system. The samples collected from fractured and faulted zones reveal to have gas composition similar to that obtained from Lusi crater, indicating deep origin fluids.

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## 1. Introduction

Several studies have highlighted the presence of anomalous gas concentrations over active faults defining these gases as fault

indicators (Baubron et al., 2002; Ciotoli et al., 1999, 2005, 2016; Fu et al., 2005; Klusman, 1993; Lombardi et al., 1996; Quattrocchi et al., 2012; Sciarra et al., 2015; Zhang and Sanderson, 1996; Zhiguan, 1991). Irwin and Barnes (1980) showed a close relationship between tectonically active areas and anomalous crustal emissions of CO<sub>2</sub>. Due to their high crustal permeability, faults act as preferential pathways for the upward migration and eventual release of deep fluids into the aquifers (Claesson et al., 2004; Lupi et al., 2010, 2011;

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Skelton et al., 2014; Yuce et al., 2014) or directly to the atmosphere. Toutain and Baubron (1999) emphasized that fault gases display a very wide range of geochemical signatures and that it is possible to relate these peculiarities to the contrasting characters and sources of the respective leaking gases. Thermal, radiogenic and geodynamic processes are involved in Earth degassing at active faults, therefore inducing complex patterns of degassing from the crust. In particular, the study of the spatial distribution of soil gases in faulted areas appears to be a suitable tool for identifying active tectonic structures. In a wide range of geological settings, radon ( $^{222}\text{Rn}$ ) measurements, often together with other gas (carriers) like carbon dioxide ( $\text{CO}_2$ ) and methane ( $\text{CH}_4$ ), is successfully used as pathfinder component to study fault and fractured fields (Annunziatellis et al., 2003; Ciotoli et al., 1999, 2005, 2016; Sciarra et al., 2015).

It is commonly accepted that gas migration at a large scale (e.g. ten to hundreds of meters) is supported by advection (pressure changes: movement of matter due to the action of a force), prevailing over diffusion (concentration changes: gas molecules move from points of high concentration towards points of low concentration) (Etiopie and Martinelli, 2002). Damage zones in faulted regions often exhibit a high permeability compared to the surrounding rocks and may facilitate the fluids advective transport. In hydrothermal areas or zones interested by endogenous gas rising, the pressure decrease allows the gases to escape from the fluids into soil-gas and eventually into the atmosphere as their migration towards the surface proceeds (Italiano et al., 2014).

The Watukoserk fault system (WFS) in NE Java, Indonesia, represents one of the most intriguing structures that have been recently discussed to understand the birth and evolution of the large Lusi eruption site (NE Java, Indonesia; Van Noorden, 2006). The WFS strikes SW-NE direction and elongates from the Arjuno-Welirang volcanic complex, crops out creating the 3 km long and up to 160 m high Watukosek escarpment, changes the course of the Porong river, and progresses towards the NE of the Java Island crossing the Lusi eruption site (Mazzini et al., 2007; Moscarillo et al., 2017; Obermann et al., 2017). Mazzini et al. (2009) documented evidences supporting the reactivation of the WFS after the M6.3 earthquake of 27<sup>th</sup> May 2006 (U.S. Geological Survey, <http://earthquake.usgs.gov/eqcenter/>) to which followed a series of fault-aligned eruption sites in the Sidoarjo regency (NE Java). Within weeks a prominent eruption site formed a distinct crater and covered with boiling mud a surface of nearly 1.5 km<sup>2</sup>. The new eruption site was called Lusi. After more than 10 years Lusi is still active.

The aim of this work is to a) to condensate and summarize the main concepts and approaches to study gas transport in sediments and faulted zones, and b) to investigate with a multidisciplinary approach and quantitative data the fluid migration in the region around the Lusi eruption site where clear fault planes or diffused fracturing zones have been mapped as part of the WFS (Istadi et al., 2009; Mauri et al., 2017).

## 2. Radon and migration mechanisms

Radon is a radioactive gas that is naturally present in the earth's crust in varying concentrations. Thirty-nine radon isotopes have been known from  $^{193}\text{Rn}$  to  $^{231}\text{Rn}$ , but just  $^{222}\text{Rn}$ ,  $^{220}\text{Rn}$  and  $^{219}\text{Rn}$  have a significant abundance (Nazaroff and Nero, 1988).  $^{222}\text{Rn}$  ( $^{238}\text{U}$  series) is the most abundant in confined environments because of its larger half-life (3.823 days);  $^{220}\text{Rn}$  ( $^{232}\text{Th}$  series), commonly referred to as thoron, is characterized by a shorter half-life (55.6 s) that makes its concentration in the soil gas generally higher than  $^{222}\text{Rn}$ , since  $^{232}\text{Th}$  in the crust is about 3.5 times more abundant than  $^{238}\text{U}$ .  $^{219}\text{Rn}$  is derived from the most stable isotope of actinium

( $^{227}\text{Ac}$ ), named actinon, and it has a half-life of 3.96 s and also emits alpha radiation. Due to the different half-lives of individual isotopes, the distances that can be travelled by the respective isotopes before decay differ significantly.  $^{220}\text{Rn}$  can migrate over much shorter distances than  $^{222}\text{Rn}$ . As a result,  $^{220}\text{Rn}$  concentrations in soil gas reflect the very local conditions of the soil and are not significantly influenced by advective soil gas movements and mixing. Due to its geochemical properties  $^{222}\text{Rn}$  cannot be transported over long-distances by diffusion migration mechanism. It accumulates in the pore space before being transported to the rock or ground surface (Tanner, 1964).

The transport of radon over long-distances occurs advectively and needs the existence of a naturally occurring flux of a carrier gas (Yuce et al., 2010, 2017; Etiopie and Martinelli, 2002; Kristiansson and Malmqvist, 1982). The soil gas concentrations of  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$  are influenced by processes on different length scales (Huxol et al., 2013). During advective transport,  $^{222}\text{Rn}$  is moved passively in flowing water or soil gas (like  $\text{CO}_2$  or  $\text{CH}_4$ ). It therefore highlights not only fractures which undergo fluid circulation, but also those in which U-bearing minerals are commonly associated with clays. These characteristics allow radon to be used as a tool for the mapping of active faults in seismotectonic environments (Birchard and Libby, 1978; Ciotoli et al., 1999; Etiopie and Martinelli, 2002; King et al., 1996; Hauksson and Goddard, 1981; Nishimura and Katsura, 1990; Toutain and Baubron, 1999; Walia et al., 2005; Wang et al., 1991; Yang et al., 2005; Yuce et al., 2010, 2017).

## 3. $\text{CO}_2$ and $\text{CH}_4$ as carrier gases

$\text{CO}_2$  and  $\text{CH}_4$  are considered as carrier gases since they may play a dominant role for non-diffusive transport and redistribution of trace gases, like radon, toward the Earth's surface (e.g. Ciotoli et al., 1999; Etiopie and Lombardi, 1995; Hermansson et al., 1991; Malmqvist and Kristiansson, 1984; Sugisaki et al., 1983; Toutain and Baubron, 1999).

Besides aqueous vapour, carbon dioxide is the most abundant gas present in hydrothermal and volcanic environments.  $\text{CO}_2$  is often used for fault mapping (Annunziatellis et al., 2003; Beaubien et al., 2003; Sugisaki et al., 1980) as well as for monitoring both seismic and volcanic areas (Baubron et al., 1991; Shapiro et al., 1982). According to Toutain and Baubron (1999), high  $\text{CO}_2$  fluxes appear correlated with both high heat flux areas (associated with active and ancient volcanism) and limited areas with deep fracturing (emitting carbon originated from the mantle and from decarbonation processes, with possible mixing of these two sources). Irwin and Barnes (1980) suggested that discharges of  $\text{CO}_2$  might indicate areas with high pore pressure at depth, and therefore may serve to identify potential tectonically active areas. The presence of carbon dioxide in fault zones may be the result of mixing of some of the following potential sources: mantle degassing, carbonate metamorphism, carbonate dissolution with subsequent  $\text{CO}_2$  release, organic material oxidation and surface biological activity, as microorganism or plant respiration (Sugisaki, 1983). Isotopes of carbon generally allow discriminating the different sources: the  $\delta^{13}\text{C}$  values range between  $-2.0\text{‰}$  and  $+2.0\text{‰}$  vs. VPDB for sedimentary carbon (Craig, 1963), from  $-7.0\text{‰}$  to  $-3.0\text{‰}$  vs. VPDB for mantle degassing (Javoy et al., 1982) and from  $-13.0\text{‰}$  to  $-30.0\text{‰}$  vs. VPDB for organic carbon (Cerling et al., 1991).

Methane is a reduced form of carbon, which is thought to derive from a variety of sources. Major processes involved in  $\text{CH}_4$  genesis are microbial and thermogenic, both operating on biologically formed organic matter (Klusman, 1993). In active tectonic environments, methane is released close to major crustal discontinuities that show heat anomalies, and therefore is often associated

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