

## **Quantifying carbon dioxide flux from dormant volcanoes with low-temperature fumarolic activity: demonstration from measurements at la soufriere, guadeloupe, and campi flegrei, italy**

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Quantifying the flux of magma derived CO<sub>2</sub> dissipated by fumarolic fields at dormant volcanoes is fundamental to assess their current state of hydrothermal activity and, therefore, the likelihood of a future phreatic/magmatic eruption. There is, in fact, documented evidence that gas fluxes, and CO<sub>2</sub> flux in particular, can increase substantially during volcanic unrests and prior to eruption, due to either degassing of new ascending magma or changes in the hydrothermal system physical regime. Quantifying CO<sub>2</sub> emissions is relatively straightforward at open-conduit volcanoes with high-temperature gas venting, which release high enough quantities of SO<sub>2</sub> remotely measurable with UV spectroscopy and where CO<sub>2</sub> flux can then be obtained by scaling to fumarole CO<sub>2</sub>/SO<sub>2</sub> ratios. Less actively degassing systems (T less than 300 C), in contrast, release sulphur predominantly as H<sub>2</sub>S, that is far more difficult to detect optically than SO<sub>2</sub>, and other procedures are therefore necessary. The main point is that many dormant volcanic systems worldwide fall in this second category.

Here we present the first CO<sub>2</sub> budget estimates for two dormant volcanoes displaying intense fumarolic activities: La Soufriere of Guadeloupe (Lesser Antilles arc) and Campi Flegrei (Naples, Italy). Both volcanic systems have produced voluminous pyroclastic eruptions in the past and have shown signs of unrest during the last decades (phreatic eruptions at La Soufriere; occasional seismic swarms, extensive degassing and ground uplift at Campi Flegrei). While the CO<sub>2</sub> output by diffuse soil degassing is relatively well known at both systems, no accurate CO<sub>2</sub> budget for their fumarolic exhalations has been obtained so far. For quantifying this budget we used a portable MultiGAS sensor system that allowed us to determine the horizontal/vertical concentration profiles of CO<sub>2</sub> (plus H<sub>2</sub>O, SO<sub>2</sub>, H<sub>2</sub>S and H<sub>2</sub>) along cross-sections of the gas plumes generated by the fumarolic activity, perpendicular to the plume transport direction. Gas sampling data at 0.5 Hz and simultaneous GPS positioning provided us with the required temporal/spatial resolution to map the chemical heterogeneity of plumes. By integrating over the entire plume section, and scaling to measured plume transport speed (for thermal/UV/video imaging), the CO<sub>2</sub> fluxes were calculated. The validity of this methodology was also tested at Campi Flegrei by comparison with independent CO<sub>2</sub> flux estimates which we obtained with a Tuneable IR Laser (good match between the two independent flux estimates). Our results show that fumarolic CO<sub>2</sub> emissions are significant at both volcanoes (e.g. 1000 t/day at Campi Flegrei, about as much as from diffuse soil degassing, and 30 to 60 t/day at La Soufriere). We propose that the experimental procedure described above can be applied to many other comparable volcanic targets worldwide in order to improve our knowledge of the budget for global volcanic CO<sub>2</sub> emissions.

## Detecting volcanic CO<sub>2</sub> signals from space

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We investigate the feasibility of space-borne detection of volcanic carbon dioxide (CO<sub>2</sub>) anomalies. Our goals are: (a) better spatial and temporal coverage of volcano monitoring techniques; (b) improvement of the currently highly uncertain global CO<sub>2</sub> emission inventory for volcanoes, irrespective of mode of emission, and (c) use of volcanic CO<sub>2</sub> emissions for high altitude, strong point source emission and dispersion studies in atmospheric science.

The Japanese GOSAT Fourier-Transform Spectrometer (TANSO-FTS) and Cloud & Aerosol Imager (CAI) aboard the IBUKI satellite have been producing data since January 2009, measuring CO<sub>2</sub> total column concentrations in polar orbit with a repeat cycle of 3 days and a field of view of 10km, at an altitude of 666km amsl. With this geometry, GOSAT has the potential to spatially integrate entire volcanic edifices with one point of measurement in target mode. At the expense of not providing a spatial scanning or mapping capability, it has strong spectral resolving power and can be pointed at targets of interest, in the lowest ppmv contrast of total column CO<sub>2</sub> and boundary layer XCO<sub>2</sub>.

However, the detection of strong volcanic point sources of CO<sub>2</sub> from space is hindered by several obstacles, including orographic clouds, unknown dispersion behavior, a high tropospheric CO<sub>2</sub> background, and sparse data coverage from existing satellite sensors. These obstacles can be overcome by a small field of view, enhanced spectral resolving power, by employing repeat target mode observation strategies, and by comparison to continuous ground based sensor network data.

Since summer 2010 we have conducted repeated target mode observations of now almost 30 persistently active global volcanoes including Etna (Italy), Erta Ale (Ethiopia), and Ambrym (Vanuatu), using L2 GOSAT FTS SWIR data. In this presentation we will summarize results from over two years of measurements. Ongoing and new collaboration with country specific observatories and agencies, as well as with the Deep Carbon Observatory and the Smithsonian Institution, enhances the potential for best ground truthing.

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## CO2 dynamics in volcanic lakes from hydroacoustic and complementary measurements

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Echosounding surveys have been performed on Indonesian (Rinjani, Kelud, Kawah Ijen, Galunggung) and Philippian (Taal) volcanic lakes with a SIMRAD ES60 single beam, dual frequency (50 and 200 kHz) echosounder, since 2007. CO<sub>2</sub> gas bubbles are strong scatterers of incident acoustic waves. Echosounding methods might therefore be helpful for mapping gas emissions at the floor of volcanic lakes and to quantify fluxes of CO<sub>2</sub> emitted through the lake. We focus on the analysis of echo sounding profiles recorded in Kelud volcanic lake where gas bubbles were found to mostly contain CO<sub>2</sub>.

In November 2007, the extrusion of a new lava dome evaporated Kelud volcanic lake in Java, Indonesia. 4 months before a detailed echo sounding survey of the volcanic lake coupled to floating accumulation chamber measurements detected abnormally high carbon dioxide emissions. It constituted the earliest sign of the volcanic unrest; well before any other monitored parameter. CO<sub>2</sub> flux is quantified using an empirical equation based on the volume of bubbles backscattered in the water column. Its comparison with the fluxes retrieved from the floating chamber method better constrain carbon dioxide dynamics in the volcanic lake. It reveals that 70 % of the carbon dioxide enters the lake in a dissolved form, while the remaining 30 % is supplied to the lake on a gaseous state. Almost three-quarter of the ascending bubbles dissolve in the water column leaving the majority of the 330 Tons/day of carbon dioxide diffusing at the air-water interface.

Some recent results from Taal volcano (2011-2012) will also be discussed.

As echo sounding lakes/caldera is fast and non invasive, this method could thus provide useful information on degassing processes especially when coupled with complementary methods.

## **CO2 emissions from soils as indicators of tectonic-volcanic activity at Monogenetic Volcanic Fields**

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Gas emission at volcanoes is a routine technique for understanding the behavior of their internal systems and magmatic evolution. Gas emission measurements can be made at central volcanoes during eruptive and repose stages. At monogenetic volcanic fields however, there are no particular vents where volcanic gases can be conducted. Moreover, there are no volcanic gases present until a magmatic event starts but the time between the generation and startup of an eruption could be so fast until a new volcano is born that no monitoring is possible so far, in order to anticipate a volcano birth event. This is why Delgado Granados and Villalpando Cortes (2008) proposed a new method carrying out studies of CO<sub>2</sub> emissions from soil in order to identify areas of structural weakness that may serve as conduits for future eruptions. The CO<sub>2</sub> measured at volcanic fields is not magmatic but indicative of the tectonic activity in the field. So, these studies reflect the state of stresses in the field and the presumption is that the most tectonically active zones are prone to conduct magmas in case of the generation of melt that may ascend from depth to the surface. The areas with higher CO<sub>2</sub> emissions are then, related to the areas with larger possibility to host a new volcano. The results are thought to be useful for hazards assessment. Here, results of measurements made at several Mexican and New Zealand monogenetic fields (at Chichinautzin, Xalapa, Michoacan-Guanajuato, and Auckland) are presented. Also, a comparison with measurements made at the San Andreas Fault system is shown in order to compare the emissions in tectonic and volcanic regions.

## Reevaluating the global CO<sub>2</sub> emission from subaerial volcanism: an additional insight

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During the last two decades, scientists have paid attention to CO<sub>2</sub> volcanic emissions and its contribution to the global C budget. Excluding MORBs as a net source of CO<sub>2</sub> to the atmosphere, the global CO<sub>2</sub> discharge from subaerial volcanism has been estimated by Morner and Etiope, 2002, in about 300 Mtyr and this rate accounts for both visible, plume and fumaroles, and non-visible, diffuse, volcanic gas emanations. However, CO<sub>2</sub> emissions from volcanic lakes were not considered to estimate the global CO<sub>2</sub> discharge from subaerial volcanoes. A recent study by Perez et al., 2011, on global CO<sub>2</sub> emission from volcanic lakes had been estimated about 117 Mtyr, being 94 Mtyr reported as deep seated CO<sub>2</sub>. In order to improve the information on the global diffuse CO<sub>2</sub> emission estimated by Morner and Etiope, 2002, about 50 Mtyr, an extensive research on diffuse CO<sub>2</sub> emissions from subaerial volcanoes worldwide has been performed after evaluating the results of 287 diffuse CO<sub>2</sub> emission surveys from 83 volcanic systems situated at 23 different countries and volcanic regions. The estimated diffuse CO<sub>2</sub> emission at each survey has been normalized by the study area. Statistical-graphical analysis of the data showed three overlapping geochemical populations. The background mean is 2.6 tkm<sup>2</sup>d and represents 28.4 per cent of the total data. Peak population showed a mean of 901.4 tkm<sup>2</sup>d and represented 38.0 per cent of the data, and an intermediate group showed a mean of 74.4 tkm<sup>2</sup>d and represented 33.6 per cent of the data. Taking into account a the geometric mean of the normalized CO<sub>2</sub> emission rates for each population, b the average of the study area for each population, c the fraction of the three overlapping geochemical populations, and d the number of active subaerial volcanoes in the world, about 1400; the global diffuse CO<sub>2</sub> emission from subaerial volcanism could be estimated about 829 Mtyr of which 436 Mtyr could be reported as deep seated CO<sub>2</sub>. This study highlights the importance of a deeper revision of the actual global CO<sub>2</sub> discharge from subaerial volcanism since a new estimate of 780 Mtyr, 250 Mtyr from visible emissions plus 94 Mtyr from volcanic lakes plus 436 Mtyr from diffuse emissions, could be considered. Morner and Etiope, G. Global Plan. Chang, 33, 185 203, 2002. Perez et al., Geology, 39, 235 238, 2011.

## Mantle carbon and sulfur fluxes in subduction zone

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Carbon and sulfur dioxide have been discharging for a long time from the Earth's mantle to the atmosphere through volcanic and hydrothermal activity. In contrast to noble gases, they do not accumulate in the atmosphere but are chemically trapped as compounds in oceanic sediment and terrestrial deposits. Some of them may be recycled again into the mantle. Subduction-zone volcanism is a key area to study these volatile transfers to the mantle and/or the recycling. The mantle C flux at mid-ocean ridge (MOR) was assessed from the spreading rate of oceanic plates and their C content (1) and from the MOR-<sup>3</sup>He flux and CO<sub>2</sub>/<sup>3</sup>He ratio in basalt glasses (2). On the other hand, the degassing rate at convergent plate margins was estimated by the flux observation in arc volcanoes (3) and the ARC-<sup>3</sup>He flux and CO<sub>2</sub>/<sup>3</sup>He ratio in volcanic gases (4). Recently MOR-<sup>3</sup>He flux has been revised to 530 mol/y by an ocean general circulation model (5), which is about half of the previous value. Based on the new value and argument of global <sup>3</sup>He flux (6), ARC-<sup>3</sup>He flux could be corrected to 110 mol/y. From the literature, we have selected 26 arc volcanic gas and steam well data whose temperatures are higher than 200 °C. Their C is well explained by the mixing of three components, MORB, Sediment and Limestone (7). Since the average CO<sub>2</sub>/<sup>3</sup>He ratio of these data is 1.9±1.0×10<sup>10</sup>, ARC-C flux would become 2.1±1.1×10<sup>12</sup> mol/y, which is consistent with 1.9×10<sup>12</sup> mol/y by the most recent estimate (8). The mantle S flux of 0.1-2.6×10<sup>11</sup> mol/y at MOR has been reported by a new experimental crushing and extraction method of MORB (9), which is significantly smaller than the old value of 2.7×10<sup>12</sup> mol/y (10). On the other hand, the ARC-S flux of 3.15×10<sup>11</sup> mol/y was estimated by the SO<sub>2</sub> flux from volcanoes (8). We discuss here the ARC-S flux based on the ARC-<sup>3</sup>He flux and SO<sub>2</sub>/<sup>3</sup>He ratio in high temperature volcanic gases.

Reference (1) Javoy et al., 1982. *Nature* 300, 171-173 (2) Marty and Jambon, 1987. *EPSL* 83, 16-26 (3) Hilton et al., 2002. *RiMG* 47, 319-370 (4) Sano and Williams, 1996. *GRL* 23, 2749-2752 (5) Bianchi et al., 2010. *EPSL* 297, 379-386 (6) Torgersen, 1989. *Chem Geol* 79, 1-14 (7) Sano and Marty, 1995. *Chem Geol* 119, 265-274 (8) Fischer, 2008. *Geochem J* 42, 21-38 (9) Kagoshima et al., 2012. *Geochem. J.* 46, e21-e26 (10) Alt et al., 1985. *Init. Rep. DSDP* 83, 283-287

## **Volcanic gas CO<sub>2</sub> flux from Japanese volcanoes**

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Volcanic gas is the major source of the Earth's degassing. The CO<sub>2</sub> flux by the volcanic gas emission has been estimated with various methods, among them the fluxes obtained by multiplying the measured SO<sub>2</sub> flux and the CO<sub>2</sub>/SO<sub>2</sub> ratio of volcanic gases are likely more reliable, as the SO<sub>2</sub> flux is the most commonly measured fluxes of the volcanic volatiles. This method, however, includes an important inconsistency that the CO<sub>2</sub>/SO<sub>2</sub> ratios are estimated based on the compositions of the fumarolic gases, which are accessible but whose SO<sub>2</sub> fluxes are not large. The major SO<sub>2</sub> degassing sources are open-vent degassing volcanoes and the CO<sub>2</sub>/SO<sub>2</sub> ratios should be estimated based on the composition of gases discharged from such volcanoes. By the application of the Multi-GAS techniques, the examples of the composition measurements of the major degassing volcanoes are increasing and I summarize the examples of Japanese volcanoes to estimate the volcanic gas CO<sub>2</sub> flux from Japan.

The CO<sub>2</sub> flux is estimated based on the time-integrated SO<sub>2</sub> flux (Mori et al., submitted) and the CO<sub>2</sub>/SO<sub>2</sub> ratios of volcanic plumes from the major degassing volcanoes in Japan. The time-integrated compilation of the SO<sub>2</sub> flux revealed that several degassing volcanoes, including Sakurajima, Miyakejima, Asama, Aso, Satsuma-Iwojima and Suwanosejima, contribute more than 90

## CO<sub>2</sub> rich vents in Costa Rica: mantle volatiles behind the volcanic front

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Here we report the first analyses of gas discharges from CO<sub>2</sub>-rich wells and springs located behind the volcanic front in Costa Rica. We collected gas samples from hydrothermal springs, dry CO<sub>2</sub> vents (30C) and CO<sub>2</sub> wells. Samples contain >87mol% CO<sub>2</sub>, with the majority of samples >95 mol% CO<sub>2</sub> and well samples at >98% CO<sub>2</sub>. Other gases are CH<sub>4</sub> (to 0.3 %), N<sub>2</sub> (0.5 to 11 %), O<sub>2</sub> (to 0.7 %), H<sub>2</sub> (0.004 to 0.26 %), Ar (0.003 to 0.25 %), He (to 0.015 %). All but one sample have significant <sup>4</sup>He enrichment relative to <sup>36</sup>Ar and <sup>22</sup>Ne [F(<sup>4</sup>He) 7 to 1120] and have air corrected <sup>3</sup>He/<sup>4</sup>He ratios from 5.3 R<sub>A</sub> to 7.7 R<sub>A</sub>. δ <sup>13</sup>C values of CO<sub>2</sub> range from -0.29 to -4.3 ‰, indicating mixing between mantle and carbonates. CO<sub>2</sub>/<sup>3</sup>He ratios range from from 3.3 x 10<sup>9</sup> to 1.6 x 10<sup>10</sup>. CO<sub>2</sub>-N<sub>2</sub>-He abundances of well samples place them in the mantle field (CO<sub>2</sub>/N<sub>2</sub> = 80 to 200) whereas spring samples have lower CO<sub>2</sub>/N<sub>2</sub> ratios that imply crustal contamination or water-gas fractionation. N<sub>2</sub>-He-Ar abundances place most samples in the mantle field. Spring samples have heavier noble gas (Kr, Xe) abundances similar to air-saturated water and suggest fractionation during degassing. Low <sup>4</sup>He/<sup>40</sup>Ar\* for average crustal (and mantle) U,Th/K suggest He-Ar fractionation. Based on CO<sub>2</sub>-He systematics the median carbonate (L), sediment (S) and mantle (M) C contents are 68%, 6%, and 26%, respectively with a L/S of 12. The L and S components of these samples are lower than the Costa Rica average (L= 82%, S=9%) and the M component is significantly higher than the volcanic front average (M=10±3%) (Shaw et al., 2003, de Leeuw et al., 2007). Therefore, these samples represent volatiles with the highest proportion of mantle CO<sub>2</sub> sampled to date in Central America. The L/S ratio is higher than that of Central America (9.7) and the world (6) average (Shaw et al., 2003). We interpret samples to represent volatiles from a region in the mantle wedge that has experienced the least contribution of subducted CO<sub>2</sub> and N<sub>2</sub> in Central America, likely the result of sediment off-scraping and slab devolatilization. We did not determine the CO<sub>2</sub> flux from this region, but the commercial plant alone produces about 3 x 10<sup>9</sup> mol CO<sub>2</sub>/yr, about 0.1% of the MORB flux. Future work on global volatile emissions should include these sources of mantle derived CO<sub>2</sub>.

de Leeuw GAM et al. (2007) EPSL 158:132-146

Shaw AM et al.(2003) EPSL 214:499-5



## Carbon dioxide manifestations and related hazard along the Hellenic territory

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Like other geodynamically active areas also the Hellenic territory is affected by a large number of geogenic gas manifestations. These occur either in form of point sources (fumaroles, mofettes, bubbling gases) or as diffuse soil gas emanations.

Geogenic sources release huge amounts of gases, which, apart from having important influences on the global climate, could have strong impact on human health. Gases have both acute and chronic effects. Carbon Dioxide and Hydrogen Sulphide are the main gases responsible for acute mortality due to their asphyxiating and/or toxic properties.

Gas hazard is often disregarded because in fatal episodes connected to geogenic gases the death cause is often not correctly attributed. Due to the fact that geodynamic active areas can release geogenic gases for million years over wide areas, it is important not to underestimate potential risks.

The present work produced a first catalogue of the geogenic gas manifestations of the whole Hellenic territory also considering literature data. Carbon dioxide dominated manifestations are the majority (61 out of 81). Most of them are found along the South Aegean Active Volcanic Arc. Many sites are also found in northern Greece and along the Sperchios basin - north Evia graben (central Greece) which are characterised by extensional tectonic activity. A preliminary estimation of the gas hazard has been made for the time period of the last 20 years considering the whole population of Greece. In this period at least two fatal episodes with a total of three victims could be certainly attributed to geogenic gases (specifically carbon dioxide). This would give a risk of  $1.3 \times 10^{-8}$  fatalities from geogenic gas manifestations per annum. Of course this risk is unevenly distributed along the whole Hellenic territory and it will depend on many factors. The most important factor will be the geographical distribution of the natural gas manifestations while also the strength of the source, the chemical composition of the gases, the meteorological conditions and the topography of the area will contribute to the determination of the local risk. The assessment of the geographical distribution of the risk levels is a difficult task, but the present catalogue of the gas manifestations of the natural gas manifestations of Greece will be a contribution to its determination.

Since deaths due to natural gases are often wrongly attributed we cannot exclude that some fatal episode has not been recognized and thus that the risk is somewhat higher than that here assessed. Although very low this risk has not to be neglected, not only because possibly underestimated but also because simple countermeasures could be adopted. Dangerous area can be easily identified and delimited by geochemical prospections and their hazard properly evidenced.

## Isotopic and mass balance constraints on the origin(s) of carbon dioxide emissions from Merapi volcano, central Java, Indonesia

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Carbon dioxide produced by arc volcanism can have a complex origin, involving different sources: the upper mantle wedge, the subducting slab, and the crust in case of continental arcs. Discriminating between these sources is thus important to properly constrain the deep carbon cycle at regional and global scales. Merapi volcano, in central Java, is famous worldwide for its sustained dome forming eruptive activity but also for the presence of metamorphosed carbonate xenoliths and calcium rich xenocrysts in its basaltic andesite products, providing evidence of direct magma interaction with carbonate host rocks in its crustal basement. This has raised the idea that crustal derived CO<sub>2</sub> could contribute significantly to its volatile budget and even play a key role in its explosive phases. Based on data for Merapi volcanic gases over the past 35 years, here I outline a prevalent slab derivation and minor bearing of crustal interactions for CO<sub>2</sub> emitted during the prevalent dome forming activity. Carbon dioxide is released at an average rate of 300 tons per day by hot (900-600 degree C) gas venting from the extruding lava dome and summit fumarolic fields, plus 200 tons per day from summit soil degassing. Its uniform  $\delta^{13}\text{C}$  of -4.0 per mil at all degassing sites, and the remarkable constancy of this parameter over 35 years of volcanic activity (except for one single value of -2.4 measured just after a nearby tectonic earthquake in 2006), demonstrate its bulk derivation from a steady magmatic source. In the same time, its high CO<sub>2</sub> to helium-3 ratio (6 times the average MORB ratio) implies a large fraction (0.8) of non-mantle carbon with maximum possible  $\delta^{13}\text{C}$  of -3.2 per mil. Such a component cannot be produced by metamorphism or melting of local carbonate sediments (-2.2 to 1.4 per mil) which, moreover, should lead to higher isotopic variability in emitted gases. Instead, it fits with the expected C isotopic signature for Indian Ocean sediments subducted beneath Java. Therefore, these observations, as well as typical arc isotopic signatures for water, sulfur and nitrogen in the volcanic gases, support a prevalent slab carbon contribution, in agreement with O, Sr, Nd, Pb isotopic data for bulk Merapi lavas. The CO<sub>2</sub> to HCl ratio of the volcanic gases, combined with melt inclusion data for chlorine, imply a pre-eruptive CO<sub>2</sub> content of between 0.5 and 1 wt per cent in Merapi feeding magma.

## **Quantifying a soil CO<sub>2</sub> flux baseline in an urban monogenetic volcanic field: the Auckland Volcanic Field, New Zealand**

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The Auckland Volcanic Field (AVF) is a potentially active monogenetic basaltic field underlying Auckland, New Zealand's major metropolis and economic centre. An eruption could occur with very little warning in anywhere within the city or its harbours. Through the DEtermining VOlcanic Risk in Auckland (DEVORA) project, scientists are attempting to understand the driving forces of monogenetic volcanism, in part to improve Auckland's monitoring networks and refine eruption forecasting models. In polygenetic volcanic settings, soil gas CO<sub>2</sub> flux measurements are routinely used to monitor the volcano's state of unrest. To date, there have been very few published studies determining baseline soil CO<sub>2</sub> flux or soil gas CO<sub>2</sub> concentrations in monogenetic fields, and none within a city centre atop a dormant monogenetic volcanic field, such as in Auckland. From 2010 to 2012, soil gas CO<sub>2</sub> fluxes and soil gas CO<sub>2</sub> concentrations were measured to establish a baseline soil gas CO<sub>2</sub> flux range and to determine the major sources of and controlling influences on Auckland's soil gas CO<sub>2</sub> flux for comparison during potential unrest. Flux measurements varied from 0 to 203 g m<sup>-2</sup>d<sup>-1</sup>, with an average of 27.1 g m<sup>-2</sup>d<sup>-1</sup>. Using a graphical statistical approach, two populations of CO<sub>2</sub> fluxes were identified. Both populations may represent the biological CO<sub>2</sub> production background flux in the AVF, with soil permeability, soil temperature and soil moisture acting as the main controls. Higher fluxes were attributed to varying land use properties (e.g., landfill). Isotope analyses of  $\delta^{13}\text{C}_{\text{CO}_2}$  confirm that the source of CO<sub>2</sub> in the AVF is biogenic with no volcanic component. This data may be used to assist with eruption forecasting in the event of precursory activity in the AVF and highlights the importance of knowing land use history when assessing soil gas CO<sub>2</sub> flux surveys, particularly in urban environments.

## He, CO<sub>2</sub> and heat fluxes in Aira caldera, Kyushu, Japan: Evidence for distinct degassing activity at Wakamiko and Sakurajima volcanoes

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Subduction zone volcanism is a key area to study volatile transfers from the upper mantle to the atmosphere and volatile recycling in the mantle. Aira caldera is a typical submarine caldera in subduction setting, composed of a subaerial volcano, the well known Sakurajima volcano, and a submarine crater, so-called Wakamiko. Sakurajima volcano presents many hot springs and bubbling gas spots in its flanks. Wakamiko crater shows large fumarolic bubbling gas emanation on its seafloor. To better understand the degassing activity related to the hydrothermal activity in Aira caldera, we calculate the volatile fluxes using two different methods. First, we determine for the first time, the helium and heat fluxes and also CO<sub>2</sub> flux based on the correlation between <sup>3</sup>He/<sup>4</sup>He, temperature and water depth in Wakamiko crater, along with CO<sub>2</sub>/<sup>3</sup>He ratios. Second, we calculate the helium flux at Sakurajima volcano from its estimated CO<sub>2</sub> flux and the global CO<sub>2</sub> flux.

The helium isotopic composition of Sakurajima hot springs ranges between 6.6 to 7.7Ra; it is 7.2Ra for Wakamiko seawater. This similarity of helium signature is strong evidence for a common magmatic reservoir for both Wakamiko and Sakurajima. He, CO<sub>2</sub> and heat fluxes calculated for Wakamiko are 900±211 mol/y (<sup>4</sup>He), 0.01±0.002 mol/y (<sup>3</sup>He), 184±43 t/d, and 195±22 MW, respectively. All these Wakamiko fluxes are at least one order of magnitude lower than those observed for Sakurajima (<sup>3</sup>He: 0.71 mol/y, CO<sub>2</sub>: 1800 t/d, heat: 2100 MW; Kagiya, 1981), implying that degassing at Sakurajima volcano is much stronger than that at Wakamiko crater. The variation of Sakurajima CO<sub>2</sub> flux (calculated from SO<sub>2</sub> flux (Kazahaya et al., 2012) and CO<sub>2</sub>/SO<sub>2</sub> ratio) with time, source (Minamidake or Showa crater) and eruptive activity, appears not to significantly affect the CO<sub>2</sub> flux at Wakamiko crater, much more stable (132-307 t/d; Horibe et al., 1980; Dissanayake et al., 2012) during the last 30 years. This indicates that there is no link between Sakurajima and Wakamiko degassing activity, despite having the same magmatic source.

## Monitoring CO<sub>2</sub> flux, gas geochemistry and heat output from El Chichon volcano, Mexico.

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El Chichon volcano, the youngest of the Chiapas volcanic arc, is located in the northwestern portion of the state of Chiapas, Mexico, between the Trans-Mexican Volcanic Belt and the Central American volcanic arc. The volcano crater, which is 1 km in diameter and 160 m deep, contains a warm (30-32°C) crater lake, boiling springs and steam-heated pools, and fumaroles with temperatures close to 100°C which are the surface manifestation of an active volcano-hydrothermal system (Taran et al., 1998).

After the last strong and destructive eruption occurred in 1982 the geochemical monitoring of El Chichon volcano has been developed (e.g., Peiffer et al., 2011 and references therein). The first CO<sub>2</sub> flux measurements at the lake surface was performed in 2007 using the floating accumulation chamber (Mazot and Taran, 2009).

The first data yielded values of 1190 g m<sup>-2</sup> d<sup>-1</sup> (March 2007), 730 g m<sup>-2</sup> d<sup>-1</sup> (December 2007) and 1134 g m<sup>-2</sup> d<sup>-1</sup> (April 2008) with the total output of 164 ± 9.5 t/d (March 2007), 59 ± 2.5 t/d (December 2007) and 109 ± 6.6 t/d (April 2008). The estimated average flux from the crater floor using data only from the lake surface was estimated as 1.102 g m<sup>-2</sup>d<sup>-1</sup> in April 2008 with the total emission of 144 ± 5.9 t/d (Mazot et al. 2011). The main permeable structure discovered in these works identified as the Cathedral fault crosses the lake in NW-SE direction and is characterized by a high CO<sub>2</sub> emission. In this work we report new data on the total output of CO<sub>2</sub> from the crater area (lake plus soil) and present a generalized map of the crater floor with the structure, CO<sub>2</sub> flux, soil temperature and carbon isotopic composition. Furthermore, the total convective heat output will be estimated using CO<sub>2</sub>/H<sub>2</sub>O ratios obtained from gas chemistry of the fumaroles. The study of these geochemical data will provide insights into El Chichon volcano-hydrothermal system.

## Spatial and temporal variations of soil CO<sub>2</sub> flux in geothermal areas of the Tatun Volcano Group, Northern Taiwan

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Soil CO<sub>2</sub> flux variation in volcanic area has been considered as a useful tool to investigate the volcanic activity in a safe distance. In this work, we have measured the soil CO<sub>2</sub> flux by closed-chamber method in representative geothermal areas of Tatun Volcano Group (TVG) and further to discuss its spatial and temporal variations.

Soil CO<sub>2</sub> flux can be obtained ca. 537 g m<sup>-2</sup> day<sup>-1</sup> at Geng-tze-ping (GTP), ca. 122 g m<sup>-2</sup> day<sup>-1</sup> at Da-you-keng (DYK), ca. 425 g m<sup>-2</sup> day<sup>-1</sup> at She-haung-ping (SHP) and ca. 24.6 g m<sup>-2</sup> day<sup>-1</sup> at Tatun Natural Park, respectively. The results show that the soil CO<sub>2</sub> flux at DYK is much lower than the values of GTP and SHP, although the DYK fumaroles exhibit highest emission flux with highest <sup>3</sup>He/<sup>4</sup>He ratios. It could be explained that most CO<sub>2</sub> gas can be released to the surface through the highly permeable conduit/pathway (fumaroles) at DYK and hence, less soil CO<sub>2</sub> flux can be observed. Furthermore, we can estimate the total amount of 113 t day<sup>-1</sup> of soil CO<sub>2</sub> in the geothermal area of TVG. It is close to the values from other active geothermal areas in the world.

The Hsiao-You-Keng (SYK) area was chosen for continuous monitoring of soil CO<sub>2</sub> flux. The station, located about 50 meters away from the major fumaroles, is equipped with two CO<sub>2</sub> sensors covered and protected with plastic box, and then can be used for long term monitoring under corrosive environments. The system has continuously worked from 2008 until recent; during the monitoring period, the soil CO<sub>2</sub> flux is from ca. 0.012 to 3350 g m<sup>-2</sup> day<sup>-1</sup>. The results of soil CO<sub>2</sub> flux showed significant variations and also closely related to local rainfall. The flux apparently reduced after heavy rainfall. This could be explained by the decrease of the soil permeability, and consequently blocked the degassing pathways, due to the heavy rain. Meanwhile, no clear relationship between the flux variation and local earthquake activities can be observed in this study.

## The first record of a high time resolution carbon dioxide flux for the North-East Crater of Mount Etna

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A brand new methodology has been developed, combining Ultra-Violet (UV) camera spectroscopy with a field portable gas analyser to record CO<sub>2</sub>/SO<sub>2</sub> ratios for the production of the very first high time resolution (~1 Hz) CO<sub>2</sub> flux. The technique was tested successfully using the North-East crater of Mount Etna over the period of ~1 hour. The resultant dataset demonstrated that CO<sub>2</sub> emissions are highly variable over timescales of seconds to hours. This has significant implications for the estimation of global volcanic emissions which are currently poorly constrained, whilst the unprecedented resolution of the data opens up new possibilities for volcano monitoring and eruption forecasting. Results fit well with previous estimates of gas emissions from Mt Etna during a quiescent period with contributions of ~11.66 kt d<sup>-1</sup> and ~2.68 kt d<sup>-1</sup> assessed for CO<sub>2</sub> and SO<sub>2</sub> respectively. The CO<sub>2</sub> and SO<sub>2</sub> flux record presented non-stationary periodicities between ~40-500 s, with a shared period of ~89 s. A ~85 s period was also present in the CO<sub>2</sub>/SO<sub>2</sub> ratio, which cannot be a feature of atmospheric transport, thus upholding the volcanogenic nature of gas periodicities present. Further research needs to be completed focusing on acquiring longer datasets.

## Recent (2006-2013) unrest at Mammoth Mountain, California and the evolving role of magmatic CO<sub>2</sub>

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Mammoth Mountain is a dacitic dome complex located on the southwestern rim of Long Valley caldera in California. In 1989, an 11-month-long seismic swarm occurred beneath Mammoth Mountain and was followed by diffuse emissions of magmatic CO<sub>2</sub> and development of associated areas of tree kill, along with increases in the ratio of <sup>3</sup>He/<sup>4</sup>He in fumarolic gas and frequency of mid-crustal long-period earthquakes. These events are thought to be related to magma intrusion and upward migration of CO<sub>2</sub>-rich hydrous fluid. Over the subsequent 20+ years, an estimated 3.5 Mt of CO<sub>2</sub> have been emitted from Mammoth Mountain. Long-term geophysical and geochemical monitoring carried out by the U.S. Geological Survey was recently augmented by a multidisciplinary geophysical, geochemical, hydrologic, and biologic research project aimed at improved understanding of the origin, transport, and environmental impacts of CO<sub>2</sub> at Mammoth Mountain. Components of this project include: (1) studies that encompass CO<sub>2</sub> solubility experiments with hydrous basaltic melt, deployment of a temporary broadband seismic network, spatio-temporal analysis of fluid-driven earthquakes, and modeling fluid flow with dynamic permeability; (2) hydrologic/atmospheric studies of fumarole and spring fluid geochemistry and soil CO<sub>2</sub> flux; and (3) biologic studies of radiocarbon in tree growth rings, soil geochemistry, microbiology, and disruption ecology. Key observations of and inferences from this work to date have included the recording of multiple lower-crustal (13-31 km depth) seismic swarms beneath Mammoth Mountain between 2006 and 2013. Based on spatio-temporal analysis, the 2009 swarm was interpreted to be driven by ascending CO<sub>2</sub>-rich fluids derived from underlying magma. Solubility experiments support substantial loss of CO<sub>2</sub> from hydrous basaltic melt at these lower-crustal conditions, while hydraulic diffusivities calculated from time-depth earthquake hypocenter progression suggest transient states of high permeability suitable for enhanced fluid transport. The 2009 swarm was followed in 2009-2010 by an increase in shallow (<10 km depth) seismicity and in 2010-2011 by an increase in diffuse CO<sub>2</sub> emissions and magmatic carbon content of tree rings at the largest (Horseshoe Lake) tree kill area, as well as a rise in fumarolic <sup>3</sup>He/<sup>4</sup>He ratios on Mammoth Mountain. Preliminary analysis of the 2011-2013 seismicity suggests northeastward migration of deep seismic swarms to directly beneath Mammoth Mountain. Further detailed analysis of earthquakes coupled with continued geochemical monitoring should elucidate the relationship between magma injection, seismicity, and CO<sub>2</sub> emission in this latest phase of activity at Mammoth Mountain.



## Hydrogeochemical fluctuations related to Popocatepetl activity during the last decade: relevance of spring water monitoring to hazard assessment

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Most of the geochemical monitoring of active volcanoes is based on the measurement of gaseous emissions from the crater vent or fumaroles. However, at Popocatepetl volcano, Mexico, where sampling crater fumaroles is unviable because of the unpredictable dome destruction explosions, regular analyses of spring waters provides a valuable alternative to the sampling of the plume gas. Important variations of magma-derived gases dissolved in water have been observed preceding or concomitant to increasing activity. Such results have been informed to civil protection authorities and used for hazard assessment since the beginning of the current volcanic eruption in 1994. Integration of the information provided by diverse observation methods to achieve an accurate evaluation of volcanic hazard is particularly relevant since Popocatepetl poses a risk to a large population. About half a million may be exposed to primary volcanic manifestations, and nearly 20 million may be affected by ashfall. Hydrogeochemical analyses in a laboratory located in Mexico City have included main ions, boron, sulfide and fluoride, in addition to basic parameters such as pH and temperature, measured in the field. Dissolved CO<sub>2</sub> was calculated with the PHREEQC geochemical program. Results of the last ten years of monitoring showed that boron, sulfate, chloride and dissolved CO<sub>2</sub> are the main chemical species that may be considered as precursors to volcanic activity. However, each spring had a particular behavior, i.e. while SO<sub>4</sub>/Cl ratio steadily increased years before the appearance of a new lava dome in one site; it maintained constant values in another spring. Concentration of CO<sub>2</sub> had an important increase in two spring water samples during 2010 and 2011 previous to the appearance of a new dome in October 2011, which was also preceded by seismic unrest. Such signals prompted an aerial reconnaissance that confirmed its presence. After almost a year of boron-depleted samples, it reappeared in one of the springs before the explosion of January 25, 2012, that destroyed a dome emplaced in December, 2011. These results confirmed the need of frequent sampling and analyses of spring waters. A regular hydrogeochemical monitoring in close communication with decision makers must be maintained to provide additional factors for decision-making directed to protect endangered population.

## Monitoring of CO<sub>2</sub> soil diffuse degassing at Izu-Oshima volcano, Japan

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Izu-Oshima is a 15x9 km active volcanic island located around 100 km SSW of Tokyo. The centre of the island is occupied by a caldera complex with a diameter of 3 km. A large post-caldera cone known as Mt. Mihara is located at the south-western quadrant of the caldera. Izu-Oshima has erupted 74 times, consisting mainly in fissure eruptions, both inside and outside of the caldera. The last eruption of Izu-Oshima occurred in 1986. Since 2007, four soil gas surveys have been carried out to investigate the spatial and temporal evolution of diffuse CO<sub>2</sub> emission from this volcanic system and to identify those structures controlling the degassing process: (i) March 2007, when a soil gas survey was performed at both the entire island (91 Km<sup>2</sup>) and the central caldera, and (ii) August 2010, July 2011 and July 2012, when soil gas surveys were performed only at the summit area of Mt Mihara. Diffuse CO<sub>2</sub> emission surveys were carried out following the accumulation chamber method. The location of the CO<sub>2</sub> anomalies showed a close relationship with the structural characteristics of the volcano, with most of the gas discharged from the rim of the summit crater and the fissures of the 1986 eruption out of the caldera. As part of the volcanic surveillance program of the island and to improve the knowledge of CO<sub>2</sub> and H<sub>2</sub>S diffuse emission dynamics in Izu-Oshima, an automatic geochemical station was installed in March 25, 2008 inside summit crater of Mt. Mihara. Soil CO<sub>2</sub> and H<sub>2</sub>S efflux and several meteorological and soil physical variables have been measured in an hourly basis until present. Soil CO<sub>2</sub> efflux ranged from non detectable values up to 94.5 g m<sup>-2</sup> d<sup>-1</sup>, whereas soil H<sub>2</sub>S has been not always detected reaching efflux values up to 14 mg m<sup>-2</sup> d<sup>-1</sup>. Soil CO<sub>2</sub> efflux time series has shown a stable behaviour with variations due to external variables (environmental parameters) during monitoring period. In the other hand, temporal evolution of diffuse CO<sub>2</sub> emission rate from Mt. Mihara has shown a drastic decrease during the last six years, from 27 t d<sup>-1</sup> in March 2007 to 2.2 t d<sup>-1</sup> in July 2012. The observed behaviour on the CO<sub>2</sub> emission rate for the 4 surveys could be related to short-term meteorological variations and/or changes on the gas pressure at depth. Future work at Izu-Oshima volcano will include soil helium measurements since helium can play an important role in the prediction of volcanic events. Therefore to perform regularly soil CO<sub>2</sub> and He efflux surveys seems to be an effective geochemical surveillance tool for Izu-Oshima volcano in order to detect a change in the tendency of the gas emission rates in case of future episodes of volcanic unrest.

## **Diffuse CO<sub>2</sub> measurements in Taal Volcano Main Crater from 2008-2012 and record of possible magmatic intrusion event in 2010-2011**

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Presented here are diffuse CO<sub>2</sub> emissions data for Taal Volcano Main Crater Lake (MCL) and fumarolic areas within the Main Crater from 2008 to 2012. Baseline total diffuse CO<sub>2</sub> emissions of less than 1000T/D were established for the MCL from 3 campaign-type surveys between April, 2008 to March, 2010. Areas with high diffuse CO<sub>2</sub> emissions within the geothermal field in the Main crater were also identified. Seismicity during this period was within background and no abnormal activity was shown by the volcano. In April, 2010, anomalous seismic activity from the volcano started and the total CO<sub>2</sub> emission from the MCL increased to 2716±54T/D as measured in August, 2010. CO<sub>2</sub> emissions from the ground within the geothermal area in the Main Crater also increased from 10±1 T/D in March 2010 to 36±4 T/D in August 2010. The CO<sub>2</sub> emission from the lake was highest last March, 2011 at 4670±159T/D when the volcano was still showing signs of unrest. This large increase occurred before the highest count of high frequency earthquakes was recorded in May 2011 for this period. Total CO<sub>2</sub> emissions from the MCL then decreased to twice the baseline values from May to June 2011. The increase in CO<sub>2</sub> emissions from the MCL, geothermal area and also emissions from fumarole vents coinciding with increased seismicity during this period may be interpreted as magmatic activity. These were probably the effect of magma intrusions moving from a deep reservoir to a shallower reservoir. This activity however, did not result into an eruption and CO<sub>2</sub> emissions have gone down to below background levels since, as measured in October 2011. There has been other intrusion episodes in Taal identified by previous studies since its last eruption period in 1965-1977.

## Volcanic flank soil degassing of carbon dioxide at Gede volcano (West Java, Indonesia)

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Gede Volcano, West Java is part of an andesitic stratovolcano complex consisting of Pangrango in the north-west and Gede in the south-east. The last recorded eruptive activity was a phreatic subvolcanian ash eruption in 1957. Current activity is characterized by episodic swarms at 2-4 km depth, and low-temperature (~160 °C) crater degassing in two distinct summit crater fumarolic areas. Hot springs occur in the saddle between the Gede and Pangrango edifice, as well as on the NE flank base. The most recent eruptive events produced pyroclastic material, their flow deposits concentrate toward the NE.

A collaborative effort between the Center for Volcanology and Geological Hazard Mitigation (CVGHM) and the Earth Observatory of Singapore (EOS) is since 2010 aimed at upgrading the geophysical and geochemical monitoring network at Gede Volcano. To support the monitoring instrumentation upgrades under way, surveys of soil CO<sub>2</sub> degassing have been performed on the flanks of Gede, in circular and radial traverses. The goal was to establish a spatial distribution of flank CO<sub>2</sub> fluxes, and to allow smart siting for continuous gas monitoring stations. Crater fluxes were not surveyed, as its low-temperature hydrothermal system is likely prone to large hydraulic changes in this tropical environment, resulting in variable permeability effects that might mask signals from deeper reservoir or conduit degassing.

The high precipitation intensity in the mountains of tropical Java pose challenges to this method, since soil gas permeability is largely controlled by soil moisture content. Simultaneous soil moisture measurements were undertaken. The soil CO<sub>2</sub> surveys were carried out using a LI-8100A campaign flux chamber instrument (LICOR Biosciences, Lincoln, Nebraska). This instrument has a very precise and highly stable sensor and an atmospheric pressure equilibrator, making it highly sensitive to low fluxes. It is the far superior choice for higher precision low-flux flank surveys in tropical environments.

The mean flank fluxes measured were 19.8 g/m<sup>2</sup>/day in 2011 and 11.7 g/m<sup>2</sup>/day in early 2012. The mean flank flux for all the surveys is 17.9 g/m<sup>2</sup>/day. Statistical analysis of the data set reveals at least three distinct flux populations. Results from 2011 and 2012 indicate that flank fluxes were as high as 112.5 g/m<sup>2</sup>/day, suggesting recent intrusive activity. The spatial distribution of fluxes indicates a strong focus on the NE sector. This finding appears concurrent with an area previously documented as continuously subsiding and filled with recent pyroclastic deposits. The surveys also permit selection and validation of sites for continuous CO<sub>2</sub> monitoring stations, representing medium and low flank flux populations.

## CO<sub>2</sub> and heat fluxes in central Apennine, Italy

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Central and southern Italy are affected by an active and intense process of CO<sub>2</sub> Earth degassing in both volcanic and non volcanic environments, as revealed by the presence of many CO<sub>2</sub> rich gas manifestations in the western sectors of the region, and by regional aquifers rich in deeply derived CO<sub>2</sub> in the eastern sector, i.e. in the Apennine belt. We mapped the process and estimated the total CO<sub>2</sub> involved on the base of the carbon mass balance of the Apennine aquifers. The deeply derived CO<sub>2</sub> involved in this large Earth degassing process resulted in 2-2.5x10<sup>11</sup> mol/a that represents ~10% of the estimated present-day total CO<sub>2</sub> discharge from the sub aerial volcanoes of the Earth. The groundwaters enriched in deeply derived CO<sub>2</sub> systematically display a slight temperature anomaly, which becomes significant when the differences between the water temperatures at the springs and the temperature of corresponding recharging meteoric waters are compared. These temperature difference, together with the hydrogeologic parameters of the different aquifers, have been used to compute the total amount of heat transported by these groundwaters, which results of ~2.2x10<sup>9</sup> J/s. Most of the heat (57%) is given by geothermal warming while the remaining 43% is due to gravitational potential energy dissipation. This geothermal warming implies very high heat flux, with values higher than 300 mW/m<sup>2</sup>, in a large sector of the Apennines. The estimated heat flux in the Apennine is higher than that affecting the famous geothermal provinces of Tuscany and Latium, and the total heat release is about half of the total heat discharged at Yellowstone. This finding is in some way surprising because so far the central Apennines is though to be a cold area. This high heat and CO<sub>2</sub> flux opens a new vision of the Apennines belt and requires the existence of a thermal and fluid source such as a large magmatic intrusion at depth. Recent tomographic images of the area confirm the presence of such intrusion visible as a broad negative velocity of seismic waves. From the deep zone of the magmatic intrusion the heat is transported toward the surface by hot and CO<sub>2</sub> rich fluids, which enter the aquifers and mix with the meteoric waters. This study on the Earth degassing process in Italy reveals how the investigations based on large groundwaters systems are important for a more reliable estimation of both deep CO<sub>2</sub> and heat fluxes. In particular this is true for the tectonically young and active areas of the Earth, where large amount of meteoric waters infiltrate and deeply circulate dissolving the gas and cooling the crust.

## Interactions between magmatic degassing and regional tectonic revealed through stable carbon isotopes, Long Valley Caldera, California

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Long Valley Caldera is a large volcanic complex located on the western margin of the Basin and Range province. Its rich history of volcanic activity and its location on the eastern escarpment of the Sierra Nevada make it an ideal place to study volcano-tectonic interactions. Recent seismic swarms associated with resurgence as well as an extensive hydrothermal network of fumaroles, hot springs and tree-kill zones suggest the magmatic system is active and may potentially erupt in the future. In this project we present a comprehensive characterization of soil gases aimed at improving our understanding of relationships between the magmatic system and regional tectonics. This study marks the first field-based deployment of a portable cavity ring-down spectrometer used to obtain  $\delta^{13}\text{C}$  values in near real-time fashion. The speed and precision of this method open doors for a new generation of stable isotopic studies. A total of 223 soil gas samples were collected as point measurements, transects and grids, and they fall into three sets based on their characteristic  $\text{CO}_2$  (%) and  $\delta^{13}\text{C}$  (permil) signatures: background soil flux (–15 to –20 permil), vegetated soil flux (–20 to –25 permil) and magmatic soil flux (–2 to –7 permil). The first group of magmatic sites clusters along the southern boundary of the resurgent dome where regional throughgoing faults intersect the domal structure (average: –5.7 permil). The second group extends to the Hot Creek geothermal area located 2.5 km to the southeast of the resurgent dome in a zone dominated by the WNW-ESE restraining band of regional faulting that propagates through the caldera (–5.26 permil). The third group is associated with Mammoth Mountain volcanism and its surrounding tree kill zones (–4.57 permil). The fourth group is related to volcanism north of the Mono-Inyo domes (–6.88 permil). Based on this preliminary grouping, degassing at Mammoth Mountain, the resurgent dome and Hot Creek, and Mono Lake appears to be controlled by three distinct sources of  $\text{CO}_2$ . The high water vapor content and presence of numerous hot springs within the caldera implies significant groundwater interaction with the hydrothermal system. Furthermore, the high temperature, heavy  $\delta^{13}\text{C}$  gas requires a magmatic heat source, or a very efficient network of fluid flow pathways that circulate water and gas between the heat source and the surface along faults. Such structures are likely provided by the caldera-related and resurgence-related faults as well as by regional structures. Field observations show that degassing sites occur in close proximity to major faults, and the dimensions of these sites commonly reflects the morphology of the faults. We propose that both caldera-related and tectonic fault structures play an important role in the occurrence of magmatic gas signatures at Long Valley Caldera.

## Periodic gas release from the LUSI mud volcano (East Java, Indonesia)

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The LUSI mud volcano has been erupting since May 2006 in a densely populated district of the Sidoarjo regency (East Java, Indonesia), forcing the evacuation of 40,000 people and destroying 10,000 homes. Peak mud extrusion rates of 180,000 m<sup>3</sup>/d were measured in the first few months of the eruption, which had decreased to <20,000 m<sup>3</sup>/d in 2012. Mud volcanoes often release fluids in a pulsating fashion, with periodic timescales ranging from minutes to days, and LUSI is no exception. These oscillations, common in natural systems of multi-phase fluid flow, are thought to result from complex feedback mechanisms between conduit and source geometry, fluid compressibility, viscosity and density, changes in reservoir pressure, fluid phases or vent conditions. Crisis management workers reported observations of pulsating eruptive cycles lasting a few hours during the first two years of the eruption, and possibly beyond. Since that time, activity has shifted to individual transient eruptions recurring at intervals of a few minutes. In May and October, 2011, we documented the periodic explosive release of fluids at LUSI using a combination of high-resolution time-lapse photography, open path FTIR, and thermal infrared imagery. The mud, consisting of approximately 70% water, is erupted at temperatures close to boiling. Gases are periodically released by the bursting of bubbles approximately 3 m in diameter, triggering mud fountains ~20 m in height. No appreciable gas seepage was detected in the quiescent intervals between bubble bursts. Infrared absorption spectrometry reveals that the gas released during explosions consists of 98.5% water vapor, 1% carbon dioxide, and 0.3% methane. On rare occasions, minor amounts of ammonia were also detected. Using simplified plume geometries based on observations, we estimate that LUSI releases approximately 2,300 t/yr of methane, equivalent to 0.5% of the yearly methane production from the 4.7 million heads of cattle in East Java. We observed explosion periods from 1 to 3 minutes with a mean period of 55 s and 114 s in May and October, respectively. Two conceptual models for the periodic behavior are assessed: 1) decompressional boiling of water as fluids ascend a pathway to the surface suggests that bubbles form 10s of meters below the surface and continue to expand as they rise; periodicity results from the time to reheat and/or replace the fluid in the vicinity of bubble formation; and 2) slug flow in which carbon dioxide bubbles are seeded at much greater depths and coalesce to form evenly spaced gas slugs which rise to the surface. Our estimates of gas and mass flux, along with well-constrained fluid and mud densities, are consistent with the development of slug flow, but high measured H<sub>2</sub>O/CO<sub>2</sub> ratios in the gas plume suggests that bubbles form by decompressional boiling of water, rather than by carbon dioxide bubbles exsolved at great depths.

## **A reconnaissance survey of the CO<sub>2</sub> soil fluxes in the Michoacan Guanajuato Monogenetic Volcanic Field**

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Measurement of gas emissions from central volcanoes has long been used as an efficient monitoring methodology for evaluating volcanic activity. Since no active vents exist in monogenetic fields until a new volcano erupts, no methods are presently used to monitor the gas emissions of such fields. A problem in monogenetic volcanic fields is recognizing the most active tectonic zones and therefore the most probable sites for the birthplace of a new monogenetic volcano, particularly in tropical settings where rapid soil development and intensive erosional processes, together with the young age of the volcanic products, act together to hide traces of active faults and fractures.

Using a portable West Systems soil CO<sub>2</sub> flux meter (accumulation chamber method) we propose to measure the base level of CO<sub>2</sub> flux from soils in the Michoacan-Guanajuato monogenetic volcanic field in the West-Central Trans-Mexican Volcanic Belt and recognize the CO<sub>2</sub> fluxes values at the two youngest cones of this monogenetic field. Routine measurement of these fluxes would allow discriminating areas with noticeable variations in CO<sub>2</sub> flux, which could be considered for the installation of permanent flux meters in order to correlate the gas measurements with possible seismic and/or volcanic activity.

In this work we present results of diffuse gas measurements made at Paricutin and Jorullo volcanoes and in some fixed points along the entire Michoacan-Guanajuato monogenetic field in April 2009, June 2011, May 2012 and November 2012. We measured more than 200 points at Paricutin volcano and over 150 at Jorullo volcano. Also, we present results for 30 fixed points that had been measured 4 times.



## Diffuse CO<sub>2</sub> and H<sub>2</sub>S degassing from the summit crater of Pico do Fogo

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Pico do Fogo volcano (2.800 m.a.s.l.) is the youngest and most active volcano of the Cape Verde archipelago and it is located to the east of the Bordiera semicircular escarpment at Fogo Island. Soil gas geochemical surveys in volcanic areas are useful tool to identify changes in volcanic activity related to magmatic processes. Among these studies, to monitor spatial and secular variations of soil CO<sub>2</sub> and H<sub>2</sub>S effluxes pattern can provide important information about the state of activity of the volcanic system. CO<sub>2</sub> has been one of the most studied gases in volcanic environments. Many studies have shown that significant amounts of CO<sub>2</sub> are released to the atmosphere by quiescent volcanoes and geothermal systems through soil diffuse degassing. Since the emission rate of diffuse CO<sub>2</sub> can increase dramatically prior to an eruption (Hernandez et al. 2001a; Carapezza et al. 2004), efforts have to be made to obtain a CO<sub>2</sub> flux baseline for a given volcanic system. However, and mainly due to analytical limitations, very few works on diffuse H<sub>2</sub>S emission have been carried out at volcanic-hydrothermal areas. With the aim of improving the geochemical surveillance program of Pico do Fogo, periodic soil degassing surveys have been performed at the summit crater of this volcano, focusing on the diffuse CO<sub>2</sub> and H<sub>2</sub>S emissions. Each diffuse CO<sub>2</sub> and H<sub>2</sub>S survey has been carried out following the accumulation chamber method at 65 sites homogeneously distributed at Pico do Fogo summit crater covering an area of about 0.142 km<sup>2</sup>. Soil CO<sub>2</sub> and H<sub>2</sub>S efflux distribution maps were constructed following a Sequential Gaussian Simulation (sGs) in order to distinguish areas with anomalous CO<sub>2</sub> and H<sub>2</sub>S emission rates and to compute the total gas emission from the studied area. The total diffuse CO<sub>2</sub> output released to atmosphere was estimated in the range 30-339 t/d for the period of study. Furthermore, the total H<sub>2</sub>S output was estimated in the range 2-68 Kg/d during the same period of study. Highest CO<sub>2</sub> and H<sub>2</sub>S efflux values were measured in an around western flank of the crater where the most evident fumarolic activity occurs. The 1999 survey was performed 4 years after the last eruption at Fogo (April 1995), and an emission rate of 918 ± 409 t/d was estimated. This value was followed by a drastic decrease in the CO<sub>2</sub> and H<sub>2</sub>S emission rates during the survey performed in May 2007. In the last survey (April 2012), CO<sub>2</sub> emission rate was estimated on 186 ± 34 t/d, which is still below the higher limit observed in 1999 whereas estimated H<sub>2</sub>S emission rate was 15 ± 11 Kg/d. This observed decreasing trend on diffuse CO<sub>2</sub> emission from the summit crater of Fogo seems to be related to its eruptive cycle. At the present, following the evolutionary model of gas release from volcanoes described by Notsu et al., 2006, Fogo is an inter-eruptive phase. Diffuse soil degassing in volcanic areas releases high amount of gases and its monitoring can be helpful for the mitigation of volcanic risk at Fogo Island.

## Diffuse CO<sub>2</sub> and He Emission From Sao Miguel Volcanic Systems, Azores.

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Azores archipelago forms a 650 km WNW ESE lineament across the Mid-Atlantic ridge between 37 and 40 N and is situated close to the triple point junction of the African, American and Eurasian plates. Sao Miguel is the largest (747 km) of the nine volcanic islands that were formed by a stillactive hotspot volcanism and are mainly composed of alkali basalt and its differentiated products. Six different volcanic zones have been identified in Sao Miguel Island. The oldest unit, the Northeast volcanic complex, in the easternmost part of Sao Miguel, was followed a few hundreds of thousand years ago by the trachytic stratovolcanoes of Sete Cidades, Agua de Pau and Furnas. During latest Pleistocene and Holocene time, eruptions occurred in the two zones between these three volcanoes. To contribute to a better understanding of the volcano-structural characteristics of the different volcanic systems, a soil CO<sub>2</sub> and soil helium survey was performed in the summer period of 2011. CO<sub>2</sub> is the second most abundant gas typically released into the atmosphere from volcanic systems and helium is considered an ideal geochemical tracer due to its properties: chemically inert, physically stable and practically insoluble in water under normal conditions. These properties together with its high mobility on the crust make the presence of helium anomalies on the surface environment of a volcanic system to be related to deep fluid migration controlled by volcano-tectonic features of the area and provide valuable information about the location and characteristics of the gas source and the fracturing of the crust. The survey covered the total surface of the island with 1471 homogenously distributed sampling points. At each sampling site, soil CO<sub>2</sub> efflux measurements have been performed by means of portable NDIR sensors according to the accumulation chamber method. At each sampling site, soil gas samples were collected at 40 cm depth for helium content analysis. Helium content in the soil gases was analyzed within 24 hours by means of Quadrupole Mass Spectrometer, model Pfeiffer Omnistar 422. The highest soil CO<sub>2</sub> flux and helium enrichments were measured at Fogo and Furnas fumaroles areas. The main CO<sub>2</sub> anomalies areas were found it at Fogo and Furnas volcanic systems in the central area<sup>1</sup> of the island, while the most important soil helium enrichments, were observed at the westernmost volcanic complexes, where the younger materials are present. Significant soil helium enrichments were measured in a NE-SE trend at Sete Cidades, Dos Picos and Fogo volcanic systems, in agreement with the direction of Terceira Rift. The CO<sub>2</sub> and helium emission rates estimate for the entire island amounted for 19709 3000 t d and 45 15 kg d respectively.

## Diffuse CO<sub>2</sub> And 222Rn Degassing From Faial Volcano System, Azores

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The island of Faial is the nearest of the central Azorean islands to the Mid-Atlantic Ridge. The main morphological feature of the island is a 2-km-wide summit caldera (Caldeira) in the summit of a complex large andesitic-to-trachytic stratovolcano. The formation of the caldera was followed by construction of fissure-fed basaltic lava fields and small volcanoes that form a peninsula extending to the west. The WNW-ESE Pedro Miguel graben is the most relevant tectonic structure observed in the island, being composed by two families of faults dipping NE and SW, respectively. Two historical eruptions have occurred at the island, being the most recent the Capelinhos eruption in 1957-58. This eruption created a new island that soon merged with the western peninsula.

With the aim of estimating the total CO<sub>2</sub> emission rate at the entire island during an inter-eruptive period and to delineate the relation between soil diffuse degassing and the main volcano-tectonic structures, soil CO<sub>2</sub> afflux and 222Rn activity were measured at 432 homogeneously-spaced sampling sites. Spatial distribution maps of both parameters were constructed by means of sequential Gaussian Simulation (sGs). Diffuse CO<sub>2</sub> emission values ranged between non detectable values to 270 g m<sup>-2</sup> d<sup>-1</sup>, with an average value of 19 g m<sup>-2</sup> d<sup>-1</sup>. Highest soil CO<sub>2</sub> efflux values were measured at the East side of the island and at the North flank of the Caldeira. The total CO<sub>2</sub> emission rate estimated for the entire island was 3,418 ± 71 t d<sup>-1</sup>. In the case of 222Rn, it ranged between non detectable to 49 kBq m<sup>3</sup>, with an average value of 4 kBq m<sup>3</sup>. Anomalous high 222Rn activity values were observed along the East and South of the island, and at the Pedro Miguel graben. These data indicate a deeper source for the gas emission at Pedro Miguel graben. These data correspond to the first study of surface gas geochemistry conducted on the island of Faial and will serve as a basis for future studies of volcano monitoring.