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Crustal CO_2 liberation during the 2006 eruption and earthquake events at Merapi volcano, Indonesia

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[1] High-temperature volcanic gas is widely considered to originate from ascending, mantle-derived magma. In volcanic arc systems, crustal inputs to magmatic gases mainly occur via subducted sediments in the mantle source region. Our data from Merapi volcano, Indonesia imply, however, that during the April-October 2006 eruption significant quantities of CO_2 were added from shallow crustal sources. We show that prior to the 2006 events, summit fumarole gas $\delta^{13}C_{(CO2)}$ is virtually constant ($\delta^{13}C_{1994–2005} = -4.1 \pm 0.3‰$), but during the 2006 eruption and after the shallow Yogyakarta earthquake of late May, 2006 (M6.4; hypocentres at 10–15 km depth), carbon isotope ratios increased to $-2.4 \pm 0.2‰$. This rise in $\delta^{13}C$ is consistent with considerable addition of crustal CO_2 and coincided with an increase in eruptive intensity by a factor of ~3 to 5. We postulate that this shallow crustal volatile input supplemented the mantle-derived volatile flux at Merapi, intensifying and sustaining the 2006 eruption. Late-stage volatile additions from crustal contamination may thus provide a trigger for explosive eruptions independently of conventional magmatic processes. **Citation:** Troll, V. R., D. R. Hilton, E. M. Jolis, J. P. Chadwick, L. S. Blythe, F. M. Deegan, L. M. Schwarzkopf, and M. Zimmer (2012), Crustal CO_2 liberation during the 2006 eruption and earthquake events at Merapi volcano, Indonesia, *Geophys. Res. Lett.*, *39*, L11302, doi:10.1029/2012GL051307.

1. Introduction

[2] Arc magmas characteristically show petrographic and chemical traits that indicate addition of various amounts of crustal material during petrogenesis [*Hildreth and Moorbath*, 1988; *Davidson et al.*, 1990; *Thirlwall et al.*, 1996]. Generally, two fundamental models are considered: crustal material is mixed into the mantle source of arc magma (source contamination), i.e., it is derived from the subducted slab [*Hildreth and Moorbath*, 1988; *Gertisser and Keller*, 2003a; *Debaille et al.*, 2006], or it may be assimilated in the crust of the overriding plate when magma ascends to the surface (crustal contamination) [e.g., *Davidson*, 1985; *Thirlwall et al.*, 1996; *Chadwick et al.*, 2007]. On the basis of radiogenic isotope systems in continental arcs (ocean-continent subduction), both contamination scenarios appear significant [*Hildreth and Moorbath*, 1988; *Davidson et al.*, 1990]. For island arcs (ocean-ocean subduction), the former model is generally assumed to dominate [*Plank and Langmuir*, 1998; *Gertisser and Keller*, 2003a; Debaille et al., 2006], although crustal contamination has also been invoked in certain circumstances [*Arculus and Johnson*, 1981; *Davidson*, 1985; *Thirlwall et al.*, 1996].

[3] Merapi volcano (Central Java) is situated within the active volcanic front of the Sunda arc, resulting from the northward subduction of the Indo-Australian plate beneath Eurasia at a rate of about 6.5 to 7 cm/yr [*Tregoning et al.*, 1994]. Merapi is characterised by periods of dome growth and intermittent explosive events, and degasses continuously through high-temperature summit fumaroles [*Toutain et al.*, 2009]. Its recent eruptive activity is restricted to basaltic-andesite dome lavas and associated pyroclastic flows (block and ash flows, BAF). The upper parts of the crust underlying Merapi comprise a thick sequence (>10 km) of Cretaceous to Tertiary limestones, marls and volcanoclastic deposits [*van Bemmelen*, 1949]. These sequences outcrop in the immediate surroundings of Merapi and can be found as abundant meta-sedimentary calc-silicate xenoliths in Merapi eruptive deposits [*Camus et al.*, 2000; *Gertisser and Keller*, 2003a; *Chadwick et al.*, 2007].

[4] The long-term eruption record of Merapi suggests that frequent large explosive events have occurred in the past, and are likely to continue in the future [*Camus et al.*, 2000; *Newhall et al.*, 2000; *Gertisser and Keller*, 2003b]. Dome growth periods may last for years, but are interrupted by short-lived explosive events, lasting hours to days only [*Camus et al.*, 2000; *Newhall et al.*, 2000]. Based upon information of recent eruptive style and magma chemistry, Merapi is currently considered to be at the very beginning of a major phase of long-term increased activity [*Gertisser and Keller*, 2003b], which would pose formidable challenges to hazard mitigation efforts as >3.5 million people live in nearby Yogyakarta, ca. 25 km south of the volcano [*Newhall et al.*, 2000]. Here, we present $\delta^{13}C_{(CO2)}$ data obtained on high-temperature fumarole gas samples collected in 2002, 2003, 2005, 2006 and 2008 from the Woro fumarole field at Merapi summit (~3000 m a.s.l.). These data are supplemented by literature data from the same fumarole field, and $\delta^{13}C$ analyses of limestone samples from the basement

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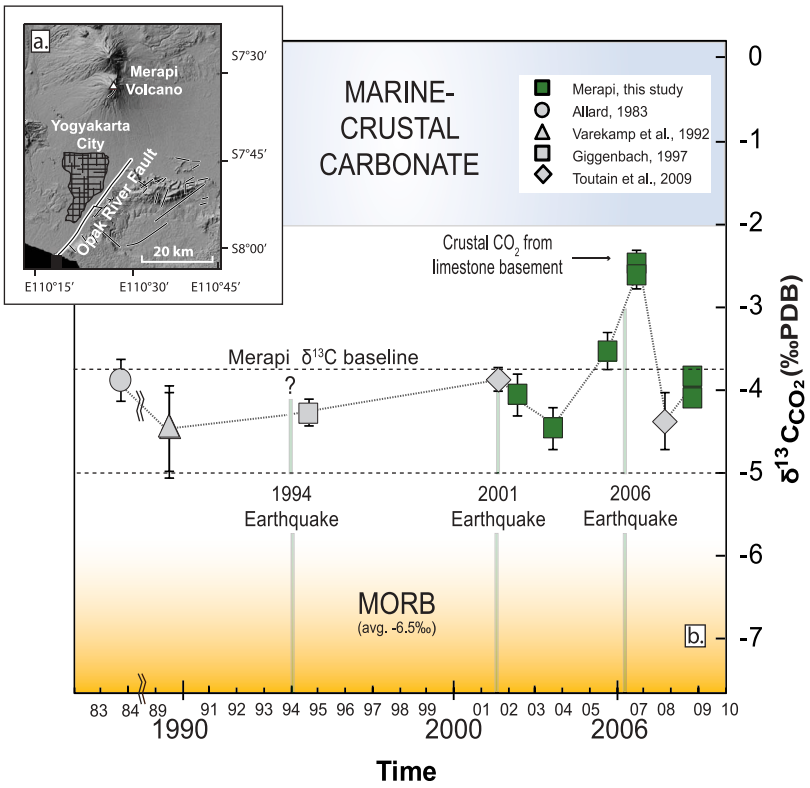

Figure 1. (a) Merapi and Yogyakarta location (after [*Walter et al.*, 2007]) and (b) variations in $\delta^{13}C_{CO2}$ in high-T Merapi fumaroles from the 1980s to 2008. Carbon isotope ratios are given in ‰ (per mil) relative to V-PDB. Green squares represent data from this study, grey symbols are values from the literature (see auxiliary material). The $\delta^{13}C_{CO2}$ values of baseline samples are considerably more positive than average mantle values (∼−6.5‰) and typical of subduction zones [*Sano and Marty*, 1995; *Hilton et al.*, 2002]. There is a marked increase in $\delta^{13}C$ during the 2006 eruption and after the May 26th 2006 earthquake, where values rise sharply relative to the baseline. This implies that a non-magmatic, high $\delta^{13}C_{CO2}$ crustal volatile input is associated with the 2006 Merapi earthquake and eruption. Local limestone provides the likely source for such a crustal volatile component.

of Merapi as well as metamorphosed calc-silicate xenoliths contained in Merapi lavas (Table S1 of the auxiliary material).[1]

2. Analytical Techniques

[5] Whole rock samples of local limestones and calc-silicate samples were analysed for carbon isotopes at the Geochemistry Laboratory, Trinity College, Dublin, Ireland and GCA Laboratories, Sehnde, Germany, respectively. The carbon isotope ratio of the 2002–2006 gas samples was measured at GFZ-Potsdam, Germany and the 2008 gas samples were analysed at Scripps Institution of Oceanography, UC San Diego, USA. Whole rock samples of basaltic-andesite, calc-silicate xenoliths and limestones from the local basement of Merapi were analysed at IFM-GEOMAR, Kiel, Germany and Acme Labs, Vancouver, Canada. See auxiliary material for technical details (Text S1, section S1).

[6] All gas samples, except from 2006, were taken during quiescence degassing periods, i.e., during times of no eruptive activity. In contrast, the 2006 samples were taken during the eruptive events that lasted from April 25th to October 1st of that year [*Walter et al.*, 2007, 2008]. Sampling permission was approved on August 25th and samples were collected on September 2nd. Notably, a M6.4 earthquake occurred near Yogyakarta on May 26th. It caused ∼6500 fatalities and left about 0.5M people homeless [*Walter et al.*, 2008]. The eruption was hence ongoing for about one month prior to the earthquake and continued for about 4 months afterwards. Alert levels were eventually reduced to normal in early October 2006 [*Wilson et al.*, 2007].

3. Results

[7] Semi-continuous monitoring of Merapi gas emissions has now established a reliable long-term record of the isotopic composition of CO_2 released via summit fumaroles (last 10 years, Table S1). Prior to 2006, variation of fumarole carbon isotope ratios was limited ($\Delta\delta^{13}C_{1994–2005}$ = 0.8 ± 0.2‰) defining an average baseline value of −4.1 ± 0.3‰ (vs. V-PDB), which falls within the range of other subduction zones [*Hilton et al.*, 2002]. In 2006, however, carbon dioxide collected after the May 26th Yogyakarta earthquake showed a dramatic increase from baseline values to −2.4‰ ± 0.2‰. In 2007 and 2008, $\delta^{13}C$ values returned to background levels (Figure 1). The May 26th earthquake coincided with an increase in eruptive intensity and volcano seismicity by a factor of 3–5 for several weeks after the

[1]Auxiliary materials are available in the HTML. doi:10.1029/2012GL051307.

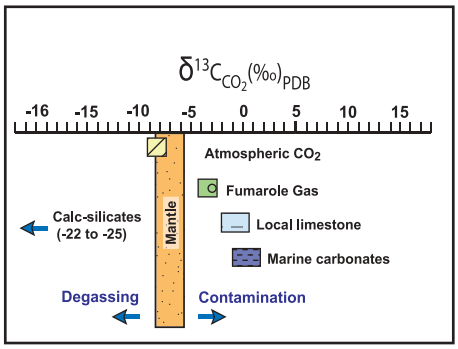

Figure 2. Plot of $\delta^{13}C_{CO2}$ of limestone basement and gas samples from Merapi; Reference fields from [*Goff et al.*, 1998] and [*Holloway and Blank*, 1994]. Degassing-related carbon isotope fractionation will shift values to the left [*Holloway and Blank*, 1994], consistent with very negative values in calc-silicate xenoliths (−22.2 to −25.0‰). Merapi fumarole samples, in turn, are elevated relative to average mantle values, suggesting a substantial addition of non-magmatic CO_2 to the magmatic system (contamination).

earthquake [*Harris and Ripepe*, 2007; *Walter et al.*, 2007, 2008].

[8] In addition to fumarole monitoring, we analysed local limestone basement rocks and calc-silicate xenoliths for their $\delta^{13}C_{CO2}$ characteristics together with representative whole rock compositions (see Tables S1 and S2). Basaltic-andesite lava samples show low CO_2 concentrations of ≤0.02 wt. % (Table S2) and $\delta^{13}C_{CO2}$ values of −23.3 to −27.5‰ [*Donoghue et al.*, 2009]. At such low concentrations, lava samples are extremely susceptible to organic carbon contamination and the $\delta^{13}C$ is likely a mixture of relict magmatic and post-eruptive organic carbon [*Macpherson et al.*, 1999; *Donoghue et al.*, 2009]. Calc-silicate xenoliths show somewhat higher CO_2 concentrations (∼0.06 to 0.3 wt. %) (Table S2), but these are still low compared to the limestones, implying that the calc-silicate xenoliths, like the lavas, have experienced severe CO_2 degassing (Figure 2) [cf. *Holloway and Blank*, 1994]). Calc-silicates do contain some relict carbonate inclusions [e.g., *Deegan et al.*, 2010] so their higher CO_2 concentrations imply that any post-eruptive organic contamination is unlikely to be significant. Calc-silicate xenoliths exhibit $\delta^{13}C_{CO2}$ values of −22.2 to −25.0‰, consistent with values expected from strongly degassed rocks [cf. *Allard*, 1983; *Holloway and Blank*, 1994]. Local limestones from the sub-volcanic basement [*van Bemmelen*, 1949; *Gertisser and Keller*, 2003a; *Chadwick et al.*, 2007], in turn, have Loss on Ignition (LOI) values of ∼42 to 43 wt. %, which if balanced with CaO to make $CaCO_3$, yields concentrations ∼43.5 wt. % CO_2 (Table S2). The $\delta^{13}C$ of the local limestones ranges between −0.8 and −2.2‰ (Table S1), i.e., typical values for marine and biogenic carbonates [*Sano and Marty*, 1995].

4. Discussion

[9] The $\delta^{13}C_{CO2}$ values used for our fumarole gas baseline (1994–2005) range between −3.5 and −4.4‰, consistent with other published analyses from Merapi [*Allard*, 1983; *Varekamp et al.*, 1992; *Giggenbach*, 1997; *Toutain et al.*, 2009], and defining an average of −4.1 with a variation of ±0.3 (1σ sd). The average of all data available (except 2006) is also −4.1‰ ± 0.3. Notably, these baseline data are significantly higher than pure magmatic, i.e., mantle-derived, CO_2 ($\delta^{13}C$ = −6 to −9‰; [*Javoy et al.*, 1986; *Marty and Tolstikhin*, 1998]) and coincide with typical subduction zone values [*Sano and Marty*, 1995; *Hilton et al.*, 2002]. Fluctuations within the baseline range are likely caused by unsteady and low-level background contributions from local limestone crust (e.g., from active contact aureoles) [*Allard*, 1983]. The origin of the baseline CO_2 can therefore be explained by a mixture of essentially subducted sediment and mantle wedge-derived CO_2 with variable, but small, additions from the crust [cf. *Allard*, 1983; *Zimmer and Erzinger*, 2003; *Johnston et al.*, 2011]. However, the 2006 $\delta^{13}C$ values are considerably higher than values normally associated with subduction zones. Such high $\delta^{13}C$ values are not produced by either open or closed system volcanic degassing, which both act to lower values [*Holloway and Blank*, 1994] (Figure 2), implying that a considerable fraction of CO_2 in the 2006 Merapi fumarole emissions must be derived from a non-magmatic, high-$\delta^{13}C$ source (Figure 1). Sedimentary carbonate basement underneath Merapi provides such a high-$\delta^{13}C$ source (Figure 2 and Table S1).

[10] In this respect, it is notable that the 1994, 1998, 2006 and 2010 eruptive deposits, like most other recent eruptions of Merapi [*Gertisser and Keller*, 2003a; *Chadwick et al.*, 2007; *Deegan et al.*, 2010], contain abundant calc-silicate xenoliths that display conspicuous vesicular degassing textures in the associated volcanic material and frequently exhibit well-developed reaction rims (Figure 3 and Text S1, section S2). The calc-silicate xenoliths comprise classic skarn mineral assemblages (diopside, wollastonite, anorthite, ±garnet, ±tremolite, ±quartz) (Figure 3) [*Camus et al.*, 2000; *Chadwick et al.*, 2007] and thus provide petrological evidence for on-going interaction between magmas and the thick succession of Cretaceous to Tertiary carbonate basement rocks beneath Merapi. The conversion of limestone to diopside + wollastonite (i.e., skarn) releases CO_2 [$CaCO_3$ (limestone) + SiO_2 (silica) ⇔ $CaSiO_3$ (wollastonite) + CO_2 ⇑ (carbon dioxide)] [*Mollo et al.*, 2010], which is added to the magmatic volatile budget. This is consistent with the very low volatile concentrations in the calc-silicate xenoliths relative to their limestone protoliths (Table S2), the low $\delta^{13}C_{CO2}$ isotope values (∼−24‰) and a general increase of diopside over wollastonite towards the rims of the xenoliths that is coupled with preferential loss of CO_2 from inclusions rims (Figure 3 and Tables S1 and S2). This latter feature implies an advanced state of magma-xenolith interaction, with progressive conversion of xenoliths to a more "magma-like" mineral composition (i.e., wollastonite + magnesium = diopside [*Bowen*, 1928]). Moreover, crystal isotope stratigraphy (CIS) of plagioclase in recent-erupted Merapi basaltic-andesites has identified carbonate assimilation and skarn recycling as processes which have markedly affected phenocryst compositions [*Chadwick et al.*, 2007]. Specifically, there is evidence of i) plagioclases with albite cores mantled by anorthite rims (almost An_{100}), with rims having high (crustal) $^{87}Sr/^{86}Sr$ ratios, indicating the presence of a Ca-rich, crustally-derived liquid during late crystallisation, and ii) plagioclases with anorthite cores (again up to almost An_{100}) and crustal $^{87}Sr/^{86}Sr$ ratios in the cores, underpinning their crustal inheritance [*Chadwick et al.*, 2007]. Calc-silicate (skarn-derived) crystal matter is frequently

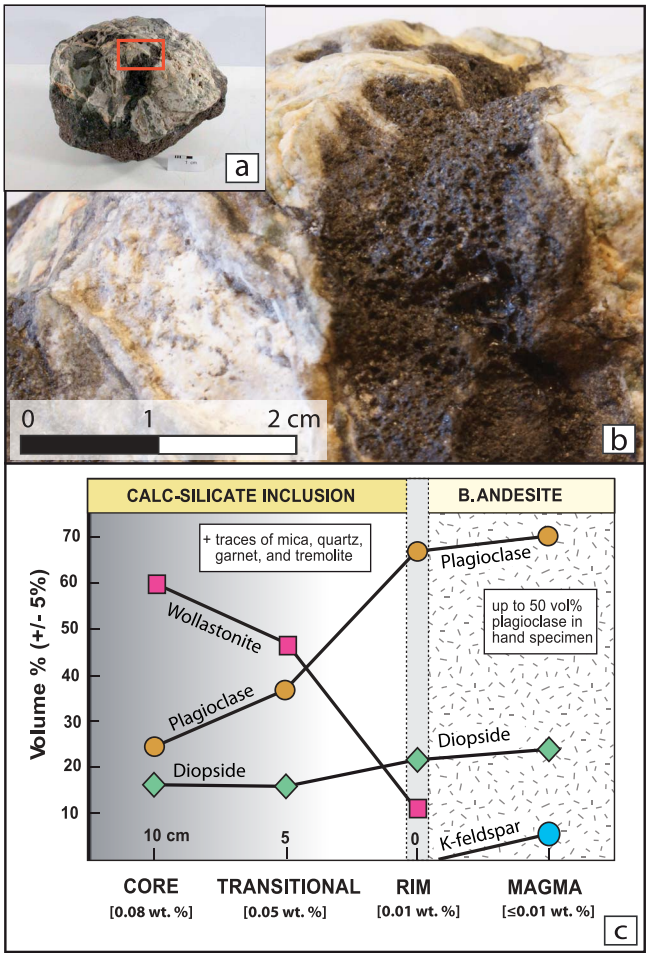

Figure 3. (a) Representative Calc-silicate xenolith (sample M-XCS-0) with wollastonite + diopside mineralogy and infiltrating andesite vein [see also *Deegan et al.*, 2010]. (b) Close up of Figure 3a (red square) shows the infiltrating magma to be strongly vesicular at the magma-xenolith contact, indicating gas liberation due to chemical interaction between magma and xenolith. (c) Mineralogical core to rim profile through same calc-silicate xenolith. Note the decrease in wollastonite and CO_2 from the core to the rim, but an increase in plagioclase and diopside, indicating an advanced state of conversion of the xenolith to a more 'lava-like' composition and is consistent with progressive CO_2 degassing of the xenolith.

identified in volcanic systems emplaced within carbonate crust, e.g., Vesuvius, Italy [*Mollo et al.*, 2010] and Popocatépetl volcano, Mexico [*Goff et al.*, 2001; *Schaaf et al.*, 2005], which, like Merapi, are prone to short-lived explosive behaviour. Petrological experiments on carbonate assimilation carried out using Merapi samples [*Deegan et al.*, 2010] demonstrate that decarbonation of limestone can produce substantial amounts of CO_2 in short time-scales (minutes to hours).

[11] Using a simple mass balance approach based upon $\delta^{13}C$ values, it is possible to quantify the contribution of crustal carbon sources to the magmatic volatile output of the fumaroles at Merapi [cf. *Iacono-Marziano et al.*, 2009]. Using an average of the 1994 to 2005 data as the baseline value (i.e., −4.1‰) and our Javanese carbonate values of −0.8 and −2.2‰, between ~50 and 80% of the CO_2 emitted during the 2006 events owes its provenance to crustal limestone (Table S3 and Text S1, section S3). Significantly, following the shallow crustal May 26th earthquake, the number of pyroclastic avalanches increased by a factor of 3 to 5, as did dome growth, reaching maximum dome volumes >150,000 m^3 [*Walter et al.*, 2007]. This is the period corresponding to the additional crustal input of CO_2. In contrast, the 2001 earthquake located deep in the subducting slab (~130 km depth) was only accompanied by a mild increase in fumarole temperature [*Walter et al.*, 2007]. The available carbon isotope data for that year do not reflect any significant increase of $\delta^{13}C$ (see Table S1).

[12] It is most unlikely that the changes in 2006 reflect mantle source variations, as they operate on timescales of 10^4–10^5 years [*Turner et al.*, 2000]. The sharp increase in $\delta^{13}C$ in 2006, its transient duration, the crustal depth of the earthquake hypocentres, and the link with eruptive and seismic intensity are, in turn, all consistent with addition of CO_2 from mid- to upper-crustal depths. Such late crustal additions of CO_2 to subduction zone baseline fluxes likely modify volatile budgets of ascending magmas at Merapi considerably [cf. *Goff et al.*, 2001; *Schaaf et al.*, 2005; *Chadwick et al.*, 2007]. We infer that magmatically-induced CO_2 liberation from long-term crustal storage reservoirs, such as the thick limestone basement of Merapi, may be a process that is triggered and/or amplified by external mechanisms, such as seismic events. It is likely that the May 26th, 2006, Yogyakarta earthquake, in conjunction with the ongoing eruption, caused stress changes in the upper crust that resulted in thermal and dynamic fracturing, release of trapped gas pockets, magma injection, xenolith entrainment and disintegration, which acted together to create a multitude of new reaction surfaces that temporarily accelerated the rate of magma-carbonate interaction [cf. *Deegan et al.*, 2010].

[13] Recently, it has been advocated that CO_2 lubricates slip planes of crustal faults, thus aiding fault rupture [*Miller et al.*, 2004]. Therefore, volcanic activity and associated CO_2 liberation may, in turn, also represent a potential trigger for increased regional seismicity. The 2006 eruption of Merapi was already ongoing for six weeks (i.e., since mid-April) prior to the May 26th earthquake. Thus, we suggest the possibility that shallow-level crustal CO_2 degassing at the volcano released CO_2 into crustal weak zones, thereby changing the regional stress regime to promote the 2006 Yogyakarta earthquake elsewhere along the fault system. In this way, we envisage a chain of events whereby earthquake and volcano interacted in a positive feedback loop. This hypothesis is supported by the slightly elevated $\delta^{13}C$ values observed in the year prior to the eruption (2005), potentially heralding increased crustal CO_2 production due to fresh magma entering the volcano edifice.

[14] Irrespective of the actual cause of the earthquake, the gas isotope evidence combined with petrological and other information on lavas and calc-silicate xenoliths, identify a crustal CO_2 end-member as the most likely source of the 'excess' CO_2 in 2006 [cf. *Allard*, 1983; *Chadwick et al.*, 2007; *Deegan et al.*, 2010]. Limestone degassing is very efficient and occurs on timescales of minutes to hours [*Goff et al.*, 2001; *Deegan et al.*, 2010; *Mollo et al.*, 2010]. Interaction between limestone and liquid magma would promote formation of a free CO_2 vapour phase, as re-dissolution of CO_2 back into the melt is extremely unlikely at shallow crustal levels [*Holloway and Blank*, 1994]. The liberated

CO_2 would add significantly to the volatile output of the volcano. In this respect, the high rates of CO_2 degassing envisaged for Merapi in 2006 may provide a modern-day analogue for the suggestion by *Johnston et al.* [2011] that increased rates of de-carbonation (outgassing) in the Cretaceous is due to contamination of arc magmas by upper crustal platform carbonates. Temporarily high rates of crustal CO_2 emissions would also directly increase the explosive character of an eruption and thus contribute to seemingly erratic eruptive events that give very limited forewarning. It is conceivable that many such $\delta^{13}C$ gas excursions go unnoticed, as they would be most pronounced during ongoing 'CO_2-fueled' eruptions, i.e., when fumarole data are effectively lacking due to safety concerns. A means to acquire CO_2 data remotely and transmit such data in real-time would add considerably to hazard mitigation efforts.

5. Conclusions

[15] We conclude that crustal volatiles intensify ongoing eruptions at Merapi independently of conventional magmatic processes, such as crystal fractionation or convective mixing, and may even be a factor in promoting regional seismic activity. From a hazard mitigation viewpoint, sudden overpressurisation due to (i) chamber wall instabilities and consequent interaction with limestone, and (ii) the release of trapped crustal gas pockets due to fracturing and dyking would provide weak shallow seismic warning signals only. Moreover, these signals would be located very close in time to an event i.e., only hours to days prior to an erratic explosive outburst. An analogy may be drawn between Merapi and similar explosive volcanoes elsewhere where carbonate forms at least part of the sub-volcanic basement (e.g., Vesuvius, and Popocatépetl [*Goff et al.*, 1998, 2001; *Schaaf et al.*, 2005; *Iacono-Marziano et al.*, 2009; *Mollo et al.*, 2010]). "Popo", for example, situated on a carbonate platform in thick continental crust, erupts abundant calc-silicate inclusions, shows elevated (excess) $\delta^{13}C_{CO2}$ values, and is characterised by sudden CO_2 outbursts, making it a prolific threat to Mexico City and surroundings. Merapi acts in a similar fashion to Yogyakarta.

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