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Dynamics of carbon dioxide emissions, crystallization, and magma ascent: hypotheses, theory, and applications to volcano monitoring at Mount St. Helens

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Abstract Measurements of CO_2 fluxes from open-vent volcanos are rare, yet may offer special capabilities for monitoring volcanos and forecasting activity. The measured fluxes of CO_2 and SO_2 from Mount St. Helens decreased from July through November 1980, but the record includes variations of CO_2/SO_2 in the emitted gas and episodes of greatly increased fluxes of CO_2. We propose that the CO_2 flux variations reflect two gas components: (a) a component whose flux decreased in proportion to $1/\sqrt{t}$ with a CO_2/SO_2 mass ratio of 1.7, and (b) a residual flux of CO_2 consisting of short-lived, large peaks with a CO_2/SO_2 mass ratio of 15. We propose two hypotheses: (a) the $1/\sqrt{t}$ dependence was generated by crystallization in a deep magma body at rates governed by diffusion-limited heat transfer, and (b) the gas component with the higher CO_2/SO_2 was released from ascending magma, which replenished the same magma body. The separation of the total CO_2 flux into contributions from known processes permits quantitative inferences about the replenishment and crystallization rates of open-system magma bodies beneath volcanos. The flux separations obtained by using two gas sources with distinct CO_2/SO_2 ratios and a peak minus background approach to obtain the CO_2 contributions from an intermittent source and a continuously emitting source are similar. The flux separation results support the hypothesis that the second component was generated by episodic magma ascent and replenishment of the magma body. The diffusion-limited crystallization hypothesis is supported by the decay of minimum CO_2 and SO_2 fluxes with $1/\sqrt{t}$ after 1 July 1980. We infer that the magma body at Mount St. Helens was replenished at an average rate ($2.8\times10^6\,m^3\,d^{-1}$) which varied by less than 5% during July, August, and September 1980. The magma body volume (2.4–3.0 km^3) in early 1982 was estimated by integrating a crystallization rate function inferred from CO_2 fluxes to maximum times (20 ± 4 years) estimated from the increase of sample crystallinity with time. These new volcanic gas flux separation methods and the existence of relations among the CO_2 flux, crystallization rates, and magma body replenishment rates yield new information about the dynamics of an open-vent, replenished magma body.

Key words Volcanic gas · CO_2 · Volcano monitoring Eruption forecasting · Volcano/atmosphere interactions · Magma body replenishment · Magma crystallization rates

Introduction

How are the rates of release of CO_2 and SO_2 from open-vent volcanos related to dynamic processes in magma bodies? What quantitative information about magmatic systems can be inferred from volcanic gas flux measurements? These questions are the primary concerns in this paper.

Carbon dioxide is potentially one of the most advantageous volcanic gas species for monitoring subvolcanic magma bodies and for forecasting eruptive activity because of its low solubility in melts (Stolper and Holloway 1988; Fogel and Rutherford 1990; Pan et al. 1991) and because of evidence that it is exsolved and released from melt in subsurface magma bodies (Harris et al. 1981; Greenland et al. 1985; Gerlach 1986). In contrast, sulfur dioxide is lost chiefly from shallow magma bodies or when magma reaches the surface. Despite the po-

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tential of CO_2 for monitoring dynamic processes in magma bodies beneath volcanos, CO_2 fluxes have not been frequently measured.

This paper evaluates the potential of CO_2 flux measurements to detect magmatic activity below the vent. It uses the most extensive set of CO_2 flux data ever collected at a volcano (Harris et al. 1981). The open vent condition at Mount St. Helens in 1980 after the climactic 18 May 1980 eruption provided an opportunity to evaluate whether measurements of CO_2 fluxes and CO_2/SO_2 ratios reveal useful information about the sources and processes controlling gas emissions at the surface. This paper does not present new data or summarize old data. It makes a first attempt at a modern interpretation of old data. It is aimed at the use and interpretation of gas flux data, especially CO_2, at active, open-vent volcanos with relation to likely magmatic processes such as crystallization and magma ascent.

Gas flux measurements and gas collections

Carbon dioxide was reported in all gas samples from high temperature fumaroles at Mount St. Helens (Gerlach and Casadevall 1986a). Maximum fumarole temperatures were estimated to have ranged from 850 to 800 °C from July to November 1980 (Gerlach and Casadevall 1986b). The abundance of CO_2 in the fumarole gases decreased after September 1980 (Gerlach and Casadevall 1986a). The present paper discusses the CO_2 flux record for the period July 1980 through September 1980. Gerlach and Casadevall (1986b) showed that although the fumarole temperatures can be modeled by mixing magmatic volatiles and hydrothermal steam, the decrease with time of CO_2 abundance in fumarole gas could not be explained solely by a two-source mixing model. Finally, CO_2 in fumarole gas had $\delta^{13}C$ values of –10.4 to –10.8 per mil PDB and suggest a magmatic origin (Evans et al. 1981). We assume in this paper that all or nearly all of the CO_2 emitted by Mount St. Helens from July to November 1980 was magmatic.

The CO_2 flux from Mount St. Helens (Fig. 1) was measured 59 times from July to October 1980 using airborne plume profiling and infrared spectrophotometry (Harris et al. 1981). Sulfur dioxide fluxes were measured at the same times (Fig. 1) using an airborne spectrometric plume profiling method (Casadevall et al. 1981). The more than 1000 individual SO_2 flux measurements at Mount St. Helens followed a trend which decreased exponentially over a period of 8 years beginning after the 18 May 1980 eruption (McGee 1992; Fig. 2). Individual CO_2 flux measurements varied widely ($1.5–22.5 \times 10^6$ kg d^{-1}) while the monthly mean fluxes decreased from 11.5×10^6 kg d^{-1} in July to 5.3×10^6 kg d^{-1} in October (Harris et al. 1981). The amount of CO_2 released during noneruptive periods from July through October was approximately 0.9×10^9 kg (Harris et al. 1981). Approximately 35% of this total was emitted during nine anomalous periods labeled "peaks" in Fig. 1. These peaks represent large increases in the flux of CO_2, which last possibly for as long as a few days, although this is unknown from the data. The SO_2 fluxes sometimes increased during the

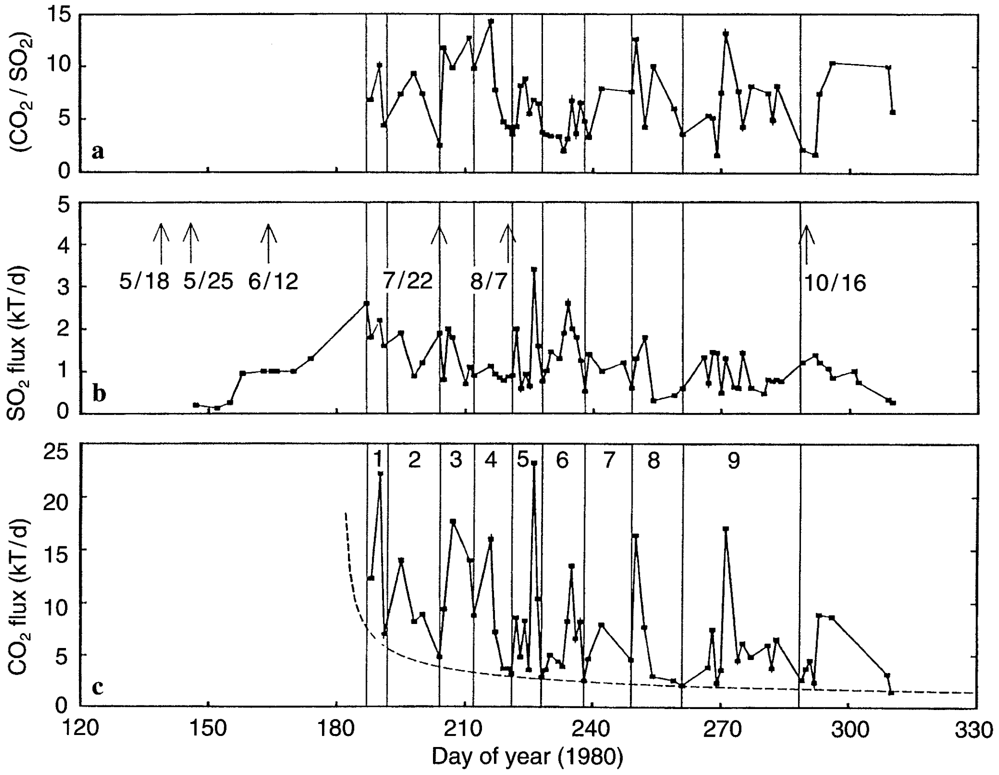

Fig. 1a–c Measured fluxes of CO_2 and SO_2 and CO_2/SO_2 ratios at Mount St. Helens from July to November 1980. **a** CO_2/SO_2 mass ratio; *dates adjacent to lines* indicate eruptions. **b** SO_2 flux from Casadevall et al. (1981). **c** CO_2 flux from Harris et al. (1981). *Vertical lines* mark boundaries between episodes (numbered) of anomalously high CO_2 emission rates involving magma ascent and replenishment of the magma body. The *dashed line* represents the baseline CO_2 flux resulting from crystallization (see Eq. (2))

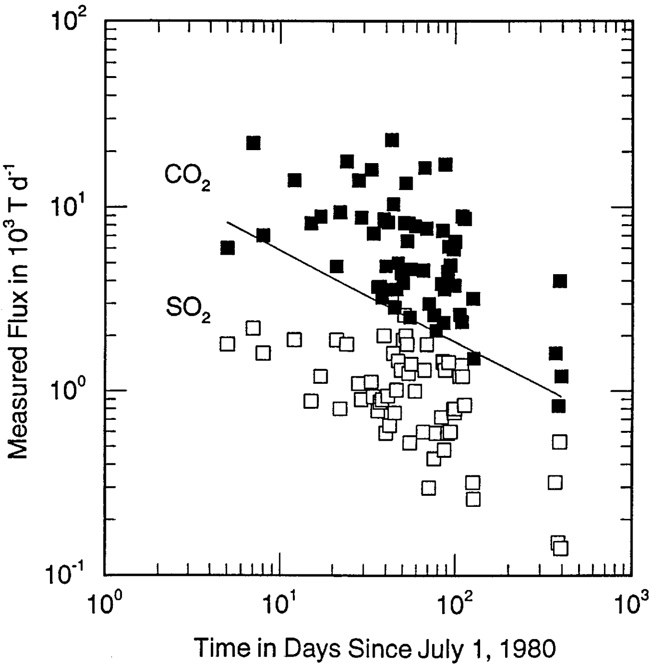

Fig. 2 Total fluxes of CO_2 (Harris et al. 1981) and SO_2 (Casadevall et al. 1981) vs time since 1 July 1980, plotted using logarithmic scales. The minimum fluxes of both species can be represented by a line with slope of –1/2, indicating that the minimum fluxes decrease in proportion to $1/\sqrt{t}$, where t is time since 1 July 1980. One kiloton per day (1 kT d^{-1}) is equal to 10^6 kg d^{-1}

anomalous periods (Fig. 1: peaks 5, 6, 8, and 9). The amount of CO_2 emitted during each of the anomalous periods (Table 1) was estimated by subtracting the expected emission represented by the baseline in Fig. 1 from the total flux. Although the peak shapes are uncertain, the amounts of CO_2 released during anomalies 5 and 6 differ by only 12–23% from the amounts released during the preceding anomaly (Table 1). Anomaly 9, which was longer than the others, is well defined and represents release of two times more CO_2 than was released during anomalies 4 and 5. The precision of individual flux measurements ranged from ±10 to ±40% (Harris et al. 1981) with the higher precision generally associated with larger fluxes and high wind speeds. The precision of average values and of peak areas derived from multiple measurements is greater than that of individual measurements. The differences between the high and low CO_2 flux measurements in the data set and in the peak areas cannot be attributed to random errors of measurement.

The CO_2/SO_2 ratios are more precise than the fluxes of either CO_2 or SO_2 alone because the flux ratio is independent of the wind speed. The CO_2/SO_2 mass ratios ranged from 1.7 to 15 (Fig. 1). The CO_2/SO_2 ratios reached relative maxima at, or close to, times of anomalously high CO_2 flux. The gas emission patterns and CO_2/SO_2 ratios bear no consistent relationship to the eruptions of 22 July, 7 August, and 16–18 October 1980. For example, the CO_2/SO_2 ratio increased from 5 to 15 over the period 9 July to 3 August 1980 (9 measurements), but decreased to near 5 over a period of several days before the eruptions on 22 July and 7 August 1980 (Harris et al. 1981). Similarly, the ratio increased from 5 to 10 between 17 September and 9 October (12 measurements), then decreased to 2 before the eruption on 16 October (Harris et al. 1981). There are positive and negative deviations superimposed on these trends. The latter time period was characterized by increasing CO_2

Table 1 Episodic releases of carbon dioxide at Mount St. Helens

Episode	Date of peak (1980)	CO_2 mass (10^6 kg)	Melt mass × distance (10^{12} kg m)	Intrusive mass for 10-km ascent (10^{10} kg)	Melt supply rate (10^6 m^3 d^{-1})	Magma supply rate (10^6 m^3 d^{-1})
1	8 July	26.5	196	1.96		
2	13 July	40.7	302	3.02		
3	25 July	72.3	536	5.36		
Intrusive mass (July 1980)				10.34	1.45	2.90
4	3 Aug	48.3	358	3.58		
5	13 Aug	43.0	319	3.19		
6	22 Aug	34.9	259	2.59		
7	29 Aug	>11	>82	>0.82		
Intrusive mass (Aug 1980)				10.18	1.43	2.86
8	6 Sept	35.7	265	2.65		
9	27 Sept	92.7	687	6.87		
Intrusive mass (Sept 1980)				9.52	1.38	2.76
Total (1 July to 1 Oct)		405.1	3000	30		

NOTE: The following assumptions can be made:

1. Solubility of CO_2 in melt given by: $[CO_2]_{melt} = 5 \times 10^{-12}$ (Pa^{-1}) P (Pa)
2. Lithostatic pressure gradient is 2.7×10^4 Pa m^{-1}
3. Magma ascent distance in 10 km
4. Replenished magma is 50% crystals

flux with time and variable SO_2 flux (Fig. 1; Casadevall et al. 1981). In this paper we use CO_2/SO_2 ratios to indicate periods of anomalous gas emissions, but it is important to point out that these can also be seen by looking solely at CO_2 flux measurements. SO_2 fluxes measured probably represent only a minimum value for sulfur gas fluxes because some sulfur may escape as another species or be removed in sublimates. This does not affect our interpretation in any way, because the CO_2 flux record provides an independent record which can be interpreted quantitatively.

Interpretations

In this paper we have tried to unscramble the CO_2 emissions record at Mount St. Helens by examining the idea that the gas emissions represent two components: (a) a long-term decreasing component similar to the overall trends observed for SO_2 at open-vent volcanoes after major eruptions (e.g., McGee 1992; Casadevall et al. 1994), and (b) an intermittent component recognizable by short periods of greatly increased flux superimposed on the long-term decreasing trend. The intermittent component is indicated in the record for Mount St. Helens by the numbered peaks of Fig. 1.

In this paper we propose hypotheses involving magmatic processes to explain CO_2 fluxes and we do not consider the possibility of a hydrothermal source, a possibility for which there is no strong evidence. We propose two hypotheses concerning the CO_2 flux variations at Mount St. Helens: Firstly, the minimum CO_2 flux (i.e., after subtraction of the peaks) was produced by crystallization of the magma body at rates governed by diffusion-limited heat transfer from the magma body to its surroundings. Secondly, the intermittent CO_2 flux component (i.e., the flux represented by the peaks) was produced by magma ascent of the type which replenished the deep magma body. We examine the consequences of these hypotheses in relation to the evidence. Our approach involves using observations to test some of the predictions of these two hypotheses. In our analysis we assume that there is a magma body beneath Mount St. Helens and that its minimum depth is approximately 8 km, based on experimental studies (Rutherford and Devine 1988; Rutherford et al. 1985; Rutherford and Hill 1993), distributions of earthquake hypocenters after eruptions (Scandone and Malone 1985), and vertical subsidence modeling (Scandone and Malone 1985).

Crystallization hypothesis

In order to fully interpret gas data in real time, a full suite of eruptive products (melt inclusions, solubilities, experimental data, etc.) should be described continually along with the flux data. Even though we could not do this in this case, we proceed to evaluate the hypothesis that gas release is controlled by crystallization. The hypothesis that gas release was controlled by crystallization at rates governed by diffusion-limited heat transfer makes three predictions which can be tested. Firstly, it predicts that the minimum flux of CO_2 should decrease with $1/\sqrt{t}$, where t is time after emplacement of the magma body. Secondly, it predicts that the minimum flux of SO_2 should vary with $1/\sqrt{t}$, assuming that sulfide fractionation is unimportant and that crystallization does not discriminate between dissolved S and CO_2 in terms of forcing them into the gas phase. Thirdly, it predicts that the amount of melt crystallized should increase in proportion to the square root of time.

The minimum CO_2 and SO_2 mass fluxes decreased with $1/\sqrt{t}$ after 1 July 1980 (Fig. 2). The minimum CO_2 flux from 5 July to 5 November 1980 satisfies the relation:

$$F_{CO_2}(t)\ \mathrm{kg\,d^{-1}} = 1.85\times10^{7}\,t^{-1/2} \tag{1}$$

with a coefficient of variation of 10.4% (Table 2), where t is time in days since 1 July 1980. This relationship suggests that the release of CO_2 from one source or process was governed directly or indirectly by a diffusion-limited process operating on the scale of the source. We identify this $1/\sqrt{t}$ decay trend as one component of the gas flux. We hypothesize that this component was caused by crystallization of magma beneath the vent and that the diffusion-limited process which governed the rate of gas release was heat flow from the magma body to its surroundings.

The CO_2 flux which is due to open-system crystallization of a stationary magma body with no change in the amount of gas stored (i.e., production rate equals release rate) should be equal to:

$$F_{CO_2}(t) = f_c(t)\, C^{m}_{CO_2}(P,t) \tag{2}$$

where $f_c(t)$ is the mass rate of crystallization and $C^{m}_{CO_2}$ is the concentration of dissolved CO_2 in the melt. The crystallization rate f_c and boundary heat flow for diffusion-limited heat transport into a half space are proportional to the reciprocal of the square root of time since emplacement (e.g., Ingersoll et al. 1948). If the minimum CO_2 flux is caused by crystallization at rates governed by diffusion-limited heat transfer, then the crystallization rate of the magma body (f_c) can be obtained from the observed baseline CO_2 flux variations (Eq. (1)) and the relation (Eq. (2)) involving the CO_2 flux, magmatic CO_2 concentration, and mass crystallization rate.

Petrographic measurements of glass (formerly melt) fractions in pumice and dome samples from Mount St. Helens (Melson 1983; see also Cashman 1992) could be used, in principle, to test the predicted linear increase in crystallinity with the square root of time after 1 July 1980. A prerequisite for using sample crystallinity data to test the hypothesis is absence of crystal fractionation in the sample set, which Melson (1983) demonstrated using geochemical data. However, because the total

Table 2 Low-CO_2 flux measurements at Mount St. Helens

Date (1980)	Days since 1 July[a]	Measured CO_2 flux (10^6 kg d^{-1})[b]	Measured CO_2 flux ($\times t^{1/2}$)[c]	Calculated CO_2 flux (10^6 kg d^{-1})[d]	Relative deviation from calculated flux (%)[e]
5 July	4	8.8	17.6	9.25	+ 5.1
10 July	9	7.0	21.0	6.17	−11.9
20 July	19	5.0	21.8	4.24	−15.2
30 July	29	3.8	20.5	3.44	− 9.5
9 Aug	39	3.1	19.4	2.96	− 4.5
19 Aug	49	2.5	17.5	2.64	+ 5.6
29 Aug	59	2.2	16.9	2.41	+ 9.5
8 Sept	69	2.0	16.6	2.23	+11.5
18 Sept	79	2.0	17.8	2.08	+ 4.0
5 Nov	127	1.4	15.8	1.64	+17.1
Mean relative error					+ 1.2
RMS relative error					10.4

[a] Time in days cince 1980 July 1

[b] Minimum CO_2 flux for date in column 1 based on lower curve in Fig. 1

[c] Product of baseline CO_2 flux and square root of time

[d] Calculated from Eq. (1)

[e] Ratio equal to 100 (calculated flux − measured flux)/measured flux

variation of the crystallinity values is small relative to the precision of individual measurements, Melson's (1983) data cannot be used to establish whether the crystallinity increased in proportion to time or to the square root of time (Fig. 3).

The effective date of the intrusion (1 July 1980) may represent the time at which the cooling surfaces of the magma body reached their steady state configuration. We do not argue for instantaneous emplacement of the Mount St. Helens magma body on 1 July 1980. On the contrary, we suggest that, although the magma body grew before and after 1 July 1980, the crystallinity and the minimum gas fluxes were controlled by diffusion-limited heat transfer across cooling surfaces which reached their final configuration on or around 1 July 1980.

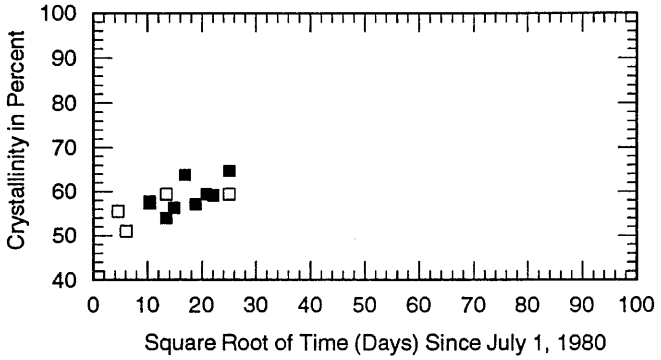

Fig. 3 Crystallinity of dacitic pumice and dome samples from Mount St. Helens eruptions from July 1980 to March 1982 (Melson 1983) vs $\sqrt{t}$. The crystallinity was calculated by Melson (1983) using K_2O in whole rock and glass samples which were not affected by crystal fractionation. The linear variation of crystallinity with $\sqrt{t}$ after 1 July 1980 is consistent with control of magma body crystallization rates by diffusion-limited heat flow from the magma body to its surroundings. Extrapolation of the trend suggests that 100% crystallization should occur at $\sqrt{t} = 76\text{–}96\ d^{-1/2}$, or 16–25 years after mid-1980 if no further replenishment occurs

Thus, there are two independent lines of evidence for crystallization control of one component of gas emissions at Mount St. Helens by diffusion-limited heat flow: the minimum CO_2 flux record and the minimum SO_2 flux record. In addition, the record of sample crystallinity vs time of eruption is consistent with the hypothesis, but precisely how sample crystallinity varies with time cannot be determined from the measurements.

Applications of the crystallization hypothesis

Inferred crystallization rate

The mass crystallization rate from 1 July 1980 onward may be inferred by substituting the CO_2 mass flux relation (Eq. (1)) into Eq. (2) and solving for f_c:

$$f_c(t)\ (\mathrm{kg\ d^{-1}}) = \frac{1.85\times10^{7}}{C^{m}_{CO_2}(P)\, t^{1/2}} \qquad (3)$$

where t is time in days since 1 July (1980) and the CO_2 concentration is expressed as a weight fraction.

There are several possible approaches for estimating the concentration of CO_2 in the melt under magmatic conditions. We use the estimated solubility of CO_2 in the melt as a proxy for the true (but unknown) concentration of CO_2 in the melt. This approach maximizes the CO_2 concentration in the melt and tends to underestimate the crystallization rate. We assume that the CO_2 solubility in dacitic melt on a mass basis is approximately $5\times10^{-12}\ \mathrm{Pa}^{-1}$. For comparison the pressure dependence of CO_2 solubility in rhyolitic melt, at 950 °C and pressures to 700 MPa, is approximately $6\times10^{-12}\ \mathrm{Pa}^{-1}$ (Fogel and Rutherford 1990). The solubility in a basaltic melt at 1200 °C and 100 MPa is 543 ppm (Pan et al. 1991). Using the assumed pressure dependence of the CO_2 solubility, Eq. (3) can be written in terms of pressure (in Pa):

$$f_c(t)\ (\mathrm{kg\ d}^{-1}) = \frac{1.85\times10^{7}}{5\times10^{-12}Pt^{1/2}} = \frac{3.7\times10^{18}}{Pt^{1/2}} \qquad (4)$$

For an assumed pressure of 200 MPa, the crystallization rate of the Mount St. Helens magma body declined from 3.7×10^{10} kg d^{-1} on 2 July to 0.33×10^{10} kg d^{-1} on 5 November 1980. The volume crystallization rates for this period decreased from 0.016 $km^3\ d^{-1}$ to 0.0014 $km^3\ d^{-1}$ for a density of 2300 kg m^{-3}.

Cumulative mass crystallized

The cumulative mass of melt crystallized $M_c(t)$ can be estimated from the integral of Eq. (4): $M_c(t) = 7.4\times10^{18}\,t^{1/2} / P$, where M_c is in kg, t is in days, and P is in Pa. The mass of liquid crystallized from 1 July to 5 November 1980 is approximately 0.42×10^{12} kg for a pressure of 200 MPa, and the volume is 0.18 km^3. The mass crystallized is a minimum value because it is based on the solubility of CO_2 in the melt, rather than on its actual concentration. The estimated cumulative mass crystallized at any time provides a lower bound on the size of the magma body.

The flux of CO_2 due to crystallization decreased by a factor of 10 from 1 July to 5 November 1980. The rapid decrease in CO_2 emissions related to crystallization emphasizes the need to measure CO_2 fluxes regularly as soon as possible after resumption of volcanic activity.

What caused the sharp CO_2 peaks?

The CO_2 fluxes in excess of the baseline values given by Eq. (1) and the observed oscillations in the CO_2/SO_2 ratios (Fig. 1a) require a cause other than crystallization in a stationary magma body. We hypothesize that relative maxima in CO_2 fluxes and in CO_2/SO_2 ratios are caused by degassing during episodic magma ascent. We also hypothesize that these events resulted in replenishment of a deep magma body approximately 8 km below Mount St. Helens.

The masses of CO_2 released episodically for the periods of anomalous gas emissions listed in Table 1 were calculated by integrating the individual peaks (Fig. 1c) and subtracting the baseline CO_2 emissions. The quantity equal to the product of magma mass and ascent distance ($M_m[Z_2–Z_1]$)was calculated by assuming a lithostatic pressure gradient of 2.7×10^{4} Pa m^{-1} and a pressure dependence of the CO_2 solubility in dacite of 5×10^{-12} Pa^{-1}. By substituting these values into Eq. (11) (see Appendix), we obtain

$$M_m[Z_2 - Z_1] = 7.4\times10^{6}(m)\int_{t_1}^{t_2} F^{a}_{CO_2}(t)\,dt \qquad (5)$$

which expresses a relation between the mass of CO_2 released and a measure of the magnitude of an intrusive event (melt mass times ascent distance). Equation (5) assumes no change in storage of exsolved CO_2.

The product of melt mass times gas-saturated ascent distance for the anomalies in Table 1 ranges from approximately $2–7\times10^{14}$ kg m. The intrusive masses required for individual peaks are $2–7\times10^{10}$ kg for assumed ascent distances of 10 km. The equivalent volumes of melt are approximately $8.7–30\times10^{6}$ m^3. The total melt mass times ascent distance inferred from episodic CO_2 emissions from 1 July to 16 October 1980 is 3×10^{15} kg m (Table 1). The intrusive mass and volume of melt (crystal-free) required are 3×10^{11} kg and 0.13 km^3, based on 10 km of ascent. This is the estimated amount of melt emplaced between 1 July and 16 October 1980, without correction for periods of missing data or entrained crystals. The magma supply rate, assuming 50% crystals in the magma, would be twice the supply rate of the melt.

The mean magma supply rate ($2.8\times10^{6}\ m^3\ d^{-1}$), which we suggest was added to the deep magma body, varied by less than 5% from July to October 1980 (Table 1). The supply rates inferred here are one to two times larger than the rate ($1.3\times10^{6}\ m^3\ d^{-1}$) estimated by Moore and Albee (1981) for growth of the shallow intrusion before the 18 May 1980 eruption.

Separating the two components in the CO_2 flux

We have two ways to separate the components: (a) subtract the "baseline" trend of long-term decrease (in situ crystallization) from the whole, to get the signal of the peaks (magma ascent) alone; and (b) use the variations of CO_2/SO_2 to separate the volcanic gas flux into contributions from sources producing gas with different CO_2/SO_2 ratios. Each separation method yields an estimate of the flux contributed by each source and the fraction of the total CO_2 flux contributed by each source as a function of time.

The CO_2/SO_2 ratio of a gas mixture (R_m) generated by mixing gas from two sources with CO_2/SO_2 ratios R_1 and R_2 is:

$$R_m = a_1 R_1 + (1 - a_1)\,R_2 \qquad (6)$$

where a_1 is the fraction of the total gas flux from source 1 and (1-a_1) is the fraction from source 2. We rearrange Eq. (6) to obtain $a_1 = (R_m–R_2)/(R_1–R_2)$. The CO_2/SO_2 ratios of the mixed gas (R_m) ranged from 1.7 to 14.3 during the observation period. Assuming $0\le a_1<1$, we infer $R_1>14.3$ and $R_2\le1.7$. Using $R_1=1.65$ and $R_2=15$, we obtained the gas flux separation result shown in Fig. 4b. The separation of flux components obtained by using the CO_2/SO_2 ratios in the volcanic gas yields a pattern similar to that obtained by the "baseline" separation method (Fig. 4a).

The estimated CO_2 fluxes contributed by magma ascent (obtained using the baseline separation method)

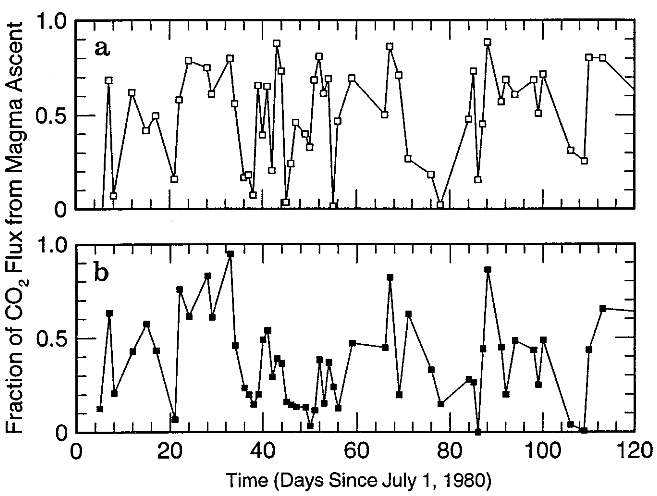

Fig. 4a, b Fraction of total CO_2 flux at Mount St. Helens contributed by magma ascent estimated by using two CO_2 flux separation methods. **a** The baseline CO_2 flux separation results are shown in the *upper graph*. **b** The CO_2/SO_2 flux separation results are shown in the *lower graph*. The two methods yield similar patterns, suggesting the equivalence of the two flux separation methods for Mount St. Helens data

and the CO_2 flux contributed by a gas source with high CO_2/SO_2 (obtained using the CO_2/SO_2 separation method) agree within the precision of these two methods (Fig. 5). This finding suggests that the CO_2 flux in excess of the baseline values and the modulation of the CO_2/SO_2 ratios above minimum values could have been controlled by a single process. Evidently, the gas generated by the hypothesized magma ascent process or intermittent source must have had CO_2/SO_2 ratios which were *higher* than in the gas produced by crystallization. This observation supports the hypothesis that magma ascent involved replenishment of the magma body from a source *deeper* than the crystallizing magma body. The basis for this conclusion is that higher fugacities of CO_2 relative to other species cause silicate melts undergoing decompression to reach saturation with respect to CO_2 (gas) before the melt reaches gas saturation for any other species. This is a consequence of the low solubility of CO_2 in silicate melts (e.g., Stolper and Holloway 1988; Pawley et al. 1992; Fogel and Rutherford 1990; Pan et al. 1991) and the assumption that the solubility of S was higher than that of CO_2 (e.g., Carroll and Webster 1994).

Alternatives to magma body replenishment

A single, nonreplenished magma body cannot explain the observed CO_2 flux measurements shown in Fig. 1c. Intermittent fracturing of the enclosing rock could release gas stored in the magma, but should not change the CO_2/SO_2 ratio. Alternatively, episodic ascent of magma toward the surface would release gas, but such a process would tend to release gas with a CO_2/SO_2 ratio close to that of gas generated by crystallization of the magma body, with relatively more SO_2 released as the ascending magma neared the surface, causing the CO_2/SO_2 ratios to decline, contrary to observations. Accordingly, we reject a single, nonreplenished magma body hypothesis.

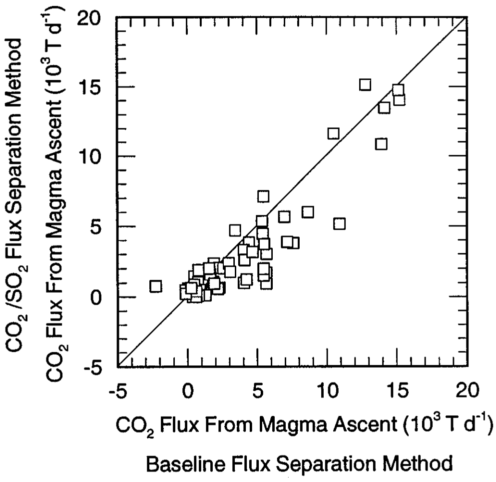

Fig. 5 Comparison of the CO_2 fluxes contributed by magma ascent estimated by the two flux separation methods. The abscissa value is the CO_2 flux (1 kT $d^{-1} = 10^6$ kg d^{-1}) due to magma ascent inferred by using the baseline CO_2 flux separation method. The ordinate value is the same quantity inferred by using the CO_2/SO_2 flux separation method. The results scatter about a line through the origin with slope 1. This graph shows the equivalence of these two methods for estimating the CO_2 flux from magma ascent for a range of conditions at Mount St. Helens. The negative value may be an artifact of a poorly defined baseline flux and large measurement error in early July 1980

The volcanic gas record also supports an alternative hypothesis involving two magma bodies and no magma ascent. In this case the volcanic gas data require that one magma body released gas continuously with CO_2/SO_2 ratio near 1.7 and a second magma body released gas with CO_2/SO_2 ratio near 15 only intermittently. We are not able to reject this hypothesis.

Our preferred hypothesis is that the fluxes of CO_2 and SO_2 were controlled by open-system crystallization and episodic replenishment of a magma body. The gas produced by magma body crystallization must have had a CO_2/SO_2 ratio near 1.7, whereas the gas produced by magma ascent must have had a CO_2/SO_2 ratio near 15. The much higher CO_2/SO_2 ratio of gas produced by magma ascent requires a source deeper than the crystallizing magma body (i.e., it requires a CO_2–rich source component).

The magma body replenishment hypothesis could be disproved if the CO_2/SO_2 ratio of the gas phase of Mount St. Helens dacitic magmas does not increase with pressure above 200 MPa or if relatively more sulfur was lost from such melts at high pressures than at low pressures. The replenishment hypothesis implicitly requires magma mixing, although the evidence produced by small mixing ratios of two compositionally related magmas might be impossible to recognize. The replenishment hypothesis, therefore, could be disproved by evidence that magma ascent from a deep source did not occur, that the magma body was not replenished, and that magma mixing did not occur before the 7 August and 16–18 Oct. 1980 eruptions.

Is magma ascent to the conduit detectable by gas emissions?

The Mount St. Helens dacite crystallized before eruption in a magma body at a pressure near 220 MPa (depth ca. 8 km) with approximately 4.6 wt.% H_2O in the liquid and approximately 67 mole% H_2O in the gas (Rutherford et al. 1985; Rutherford and Devine 1988; Rutherford 1993). The presence of thin reaction rims on a significant fraction of the amphibole phenocrysts requires three conditions: (a) pre-eruptive ascent from a magma body within the amphibole stability field, (b) mixing within the conduit of magmas with different storage histories, and (3) short residence times (typically less than 10 days) at depths of less than 6.5 km (Rutherford and Hill 1993).

The inferences from amphibole reaction rims about magma storage history and ascent rates (Rutherford and Hill 1993) are consistent with the related inferences from volcanic gas data presented herein. The volcanic gas fluxes require significant contributions from a crystallizing magma body and from magma ascent. However, the magma ascent contribution to the total CO_2 flux and the modulation of the CO_2/SO_2 ratio arise from replenishment of the deep magma body, not from pre-eruptive magma ascent, which produced the amphibole reaction rims. This raises an important issue: why is there no evidence in the volcanic gas record of pre-eruptive magma ascent into the conduit (PAC)?

Detection of PAC in the volcanic gas record requires that the signal from PAC be large relative to the background fluxes caused by magma body crystallization and replenishment. If the signal-to-background ratio for CO_2 emissions associated with PAC is too small, such an event would not be detectable by monitoring CO_2 emissions. The best conditions for detecting PAC occur when the signal due to magma body replenishment is smallest (zero). In this case the background volcanic gas emissions would be due only to crystallization. Assuming the melt loses all of its CO_2 during either crystallization or PAC, the maximum signal-to-background ratio is equal to the magma supply rate to the conduit divided by the crystallization rate. The magma supply rate to the conduit is approximately equal (to the nearest order of magnitude) to the eruptive mass divided by the length of the repose period. For the repose periods before the eruptions of 7 August and 16–18 October 1980, the signal-to-background ratios would have been 0.14 and 0.035, respectively (Table 3). These signals were simply too small to detect with the gas flux measurement methods and conditions

Table 3 Estimated replenishment, crystallization, and eruptive masses for 3 repose periods

Period	Replenishment			Crystallization		Eruptions		
	CO_2 released (10^6 kg)[a]	Mass × distance (10^{12} kg m)[b]	Intruded mass (10^{12} kg)[c]	CO_2 released (10^6 kg)[d]	Mass solidified (10^{12} kg)[e]	Mass erupted (10^{12} kg)[f]	Percent of intruded mass[g]	Percent of solidified mass[h]
Repose period before 22 July eruption								
13 June–1 July	51.9	385	0.076	Small	Small			
1–22 July	67.2	498	0.100	144	0.144			
13 June–22 July	119.2	883	0.176	170	0.17	0.0127	28	7.5
Repose period before 7 August eruption								
23 July–7 Aug	120.6	894	0.178	58	0.058	0.0083	19	14
Repose period before 16–18 Oct eruption								
8–29 Aug and 5 Sept–16 Oct	217.3	1612	0.322					
30 Aug–4 Sept	20.7	154	0.030					
8 Aug–16 Oct	238.0	1766	0.352	157	0.157	0.0055	6.2	3.5

[a] CO_2 released episodically during repose period (see Table 1)

[b] Melt mass × ascent distance from Table 1

[c] Mass of intruded magma (50% crystals) is equal to two times the mass of intruded melt in Table 1

[d] CO_2 released during repose periods calculated from integrated form of Eq. (2)

[e] Incremental mass solidified during repose period calculated from Eq. (4)

[f] Total eruptive masses include air-fall ash (Sarna-Wojcicki et al. 1981), pyroclastic flows (Rowley et al. 1981), and lava domes (Moore et al. 1981)

[g] Eruptive mass as percentage of mass of magma intruded during repose period

[h] Eruptive mass as percentage of mass of magma solidified during repose period

[i] Estimated contributions during periods with no measurements were calculated using rates measured during same repose period and adjusted for length of data gap

at Mount St. Helens. The signal was obscured by the large CO_2 fluxes due to magma body replenishment. However, if magma body replenishment were absent and crystallization rates were much smaller, increased gas fluxes and decreases in the CO_2/SO_2 ratio might be useful for detecting PAC.

Magma body volume, crystallization, and end of eruptive series

The volume of a magma body, its crystallization rate, its replenishment rate, its degassing history, and the effectiveness of crystal fractionation should govern the character and duration of an eruptive series (e.g., Tait et al. 1989). We consider whether the crystallization rates estimated from CO_2 fluxes and crystallinity data from petrographic studies (Melson 1983) can be applied to estimate the size of the magma body. We also suggest a possible reason for the apparent end of the eruptive series in October 1986.

The volume of the Mount St. Helens magma body can be estimated. We assume that (a) the crystallization rate function (Eq. (4)) may be extrapolated to complete crystallization, (b) the crystallinity of the magma body increased with $\sqrt{t}$ at the same rate suggested by samples plotted in Fig. 3, and (c) the initial magma and replenishing magma were 50% crystalline (by mass) based on Fig. 3. The total crystallization time (20 ± 4 years) of the magma body was estimated by linear extrapolation of the trend in Fig. 3 to 100% crystallization ($\sqrt{t}=86\pm10\ \mathrm{d}^{-1/2}$, or 16–25 years). The mass of melt crystallized ($2.8\text{–}3.5\times10^{12}$ kg; 1.2–1.5 km^3) was obtained by integrating Eq. (4) using $P=200$ MPa, corresponding to $[C^{m}_{CO_2}]=0.001$ by weight. The estimated magma body volume, assuming 50% crystallinity in the initial and replenished magmas, would be equal to twice the volume of melt crystallized, or 2.4–3 km^3. Our result is somewhat smaller than the volume (5–7 km^3) estimated by Pallister et al (1992) based on a review of geophysical, gas emissions, and petrologic data, and associated models for estimating magma volumes. As noted above, we assumed that the concentration of CO_2 in the melt was equal to the pressure-dependent solubility of CO_2 in the melt, causing our estimated volume to represent a minimum value. If $X^{m}_{CO_2}$ were 0.3–0.5, then our estimated volume could be easily increased by a factor of 2–3, and our estimate would be similar to that of Pallister et al. (1992).

The last eruption of Mount St. Helens (and presumably the last one in the eruptive series) occurred when the extrapolated crystallinity would have been 75–80% ($\sqrt{t}=48$; Fig. 3). We suggest that the eruptive series ended because the magma became extremely rigid when the magma crystallinity reached 75%. For magmatic systems in which crystal fractionation is not important, crystallinity trends (e.g., Melson 1983) plotted against the square root of time since intrusion may be useful in forecasting the minimum expected duration of an eruptive series. Replenishment of a mostly crystalline magma body by hotter magma could decrease the crystallinity, thereby allowing new eruptions. These potential applications to forecasting the bulk crystallinity of magma bodies and the minimum duration of an eruptive series need to be assessed in more detail.

Conclusions

The CO_2 flux measurements at Mount St. Helens from July through November 1980 represent the most robust data set of their kind from an open-vent volcano. Although the data have low precision (±10–40%) and display great variability, SO_2 flux measurements which were made at the same time aid our interpretation. The flux/time plots display trends which can be interpreted as the sum of two components: (a) a component decreasing with $1/\sqrt{t}$ with low CO_2/SO_2 ratios and (b) large peaks, which last a few days at most and have high CO_2/SO_2 ratios. We propose that these two components reflect two contrasting causes: (a) crystallization of a stationary magma body (depth ca. 8 km and volume 2.4–3.0 km^3 in March 1982) releasing gas at rates controlled by diffusion-limited heat transfer from the magma body, and (b) episodic release of gas from ascending parcels of magma ($9\text{–}30\times10^{6}\ \mathrm{m}^3$ of melt), which replenished the larger static magma body. This interpretation enables the crystallization and replenishment rates of the magma body to be estimated by analysis of volcanic gas flux measurements.

The measurement of CO_2 fluxes at volcanos is rare, and the quality and frequency of measurements in this case were not ideal. We have interpreted these data retrospectively to evaluate the possible value of such data for monitoring volcanos and forecasting activity. The Mount St. Helens case shows that volcanic gases can reveal quantitative information about replenishment and crystallization rates of magma bodies. Such knowledge may be useful in understanding and predicting the behavior of open-system magma bodies beneath active volcanos. The capability to infer magma body crystallization and replenishment rates in near real-time by separating the total CO_2 flux into contributions from different processes should lead to improvements in volcano monitoring and eruption prediction. It should also allow testing of hypotheses concerning magma transport and eruption triggering mechanisms. These capabilities, however, require increased emphasis on automated volcanic gas measurement techniques and process-based methods for analyzing volcanic gas data within the volcano monitoring community.

Appendix: mass balance equations for degassing of magma bodies

An idealized magmatic system which releases gas to a vent is illustrated in Fig. 6. We consider two magmatic

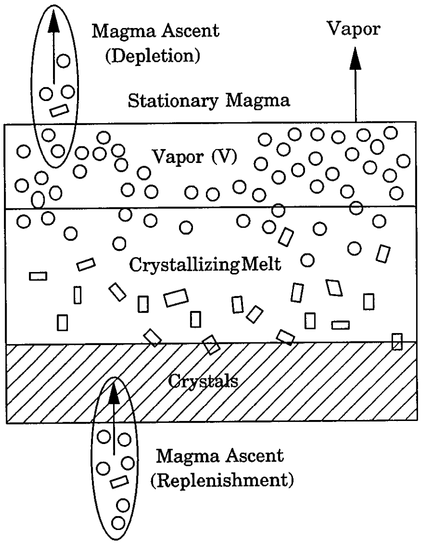

Fig. 6 Illustration of several sources contributing to volcanic gas emissions at the same time. The CO_2 flux from a stationary magma body may include contributions from crystallization, decompression, and changes in the amount of gas. Magma ascent may result in replenishment of the magma body from a deeper source, or it may diminish the amount of magma in the magma body. Quantitative interpretation of volcanic gas observations requires separation of the total flux into contributions from identifiable sources and processes

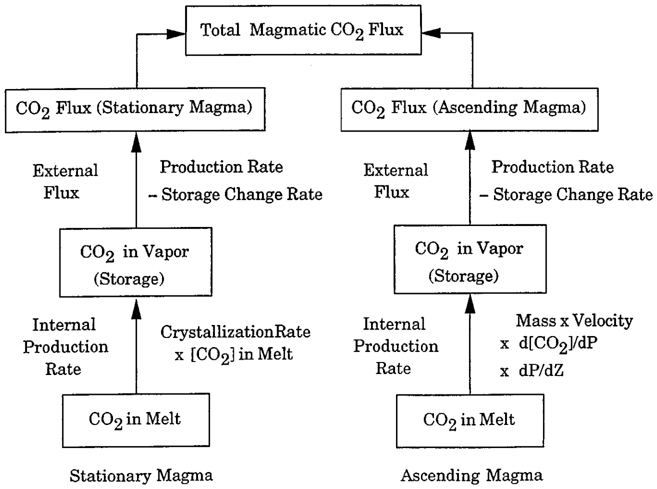

Fig. 7 Flow chart showing how the total CO_2 flux from magmatic sources is the sum of contributions from two source types: a stationary magma body and an ascending magma. In both cases the external flux from the source is the difference between the CO_2 gas production rate and the storage change rate. The two source types differ, however, in their internal production rates of CO_2 gas because the dominant process in each source type is different (crystallization vs decompression)

sources for CO_2 in volcanic gas: (a) a stationary magma body and (b) an ascending magma body. Both are assumed to be three-phase (crystals, liquid, and gas) open-system magma bodies. Our objective is to show how the total flux of CO_2 at open-vent volcanos can be related theoretically to rates of magma ascent and crystallization (Fig. 7).

The total flux of magmatic CO_2 is the sum of the fluxes from a stationary magma body and the flux from ascending magma (Fig. 7). We regard the total flux of magmatic CO_2 to be controlled by the entire magmatic system, not solely by the rate of crystallization or the rate of magma ascent. The external flux of CO_2 (i.e., the flux entering or leaving a magma body) is equal to the difference between the CO_2 production rate and the rate of change of the amount of CO_2 stored in the magmatic gas phase. Changes in the amount of gas held in a magma body may decouple the external flux of volatile components from their internal production rates.

Total mass of CO_2 in a magma body

The total mass of CO_2 in a magma body at time t is:

$$M_{CO_2}(t) = M_m(t)\, C^m_{CO_2}(P,t) + M_g(t)\, X^g_{CO_2}(P,t)\frac{W_{CO_2}}{W_g} \qquad (7)$$

where M is mass, P is pressure, C is concentration, X is mole fraction, W is molecular weight, and t is time. The subscripts and superscripts refer to gas (g), melt (m), and CO_2. We ignore solid phases in the mass balance for CO_2.

The total flux of CO_2 from a magma body (F_{CO_2}) is obtained by differentiating Eq. (7), giving $F_{CO_2}(t) = -\mathrm{d}M_{CO_2}(t)/\mathrm{d}t$. Differentiating Eq. (7), we obtain

$$\begin{aligned} F_{CO_2}(t) = & -M_m(t)\frac{\partial C^m_{CO_2}}{\partial P}\frac{\mathrm{d}P}{\mathrm{d}Z}\frac{\mathrm{d}Z}{\mathrm{d}t} \\ & -M_m(t)\left(\frac{\partial C^m_{CO_2}}{\partial t}\right)_Z - C^m_{CO_2}\frac{\mathrm{d}M_m}{\mathrm{d}t} \\ & -\left[M_g(t)\frac{\mathrm{d}X^g_{CO_2}}{\mathrm{d}t} + X^g_{CO_2}\frac{\mathrm{d}M_g(t)}{\mathrm{d}t}\right]\frac{W_{CO_2}}{W_g} \qquad (8) \end{aligned}$$

where Z is depth of the magma body and $\mathrm{d}P/\mathrm{d}Z$ is the lithostatic pressure gradient. In Eq. (8), the first term represents CO_2 exsolution due to magma ascent; the second term represents CO_2 exsolution due to changes in CO_2 in the melt at constant depth; the third term represents CO_2 exsolution due to crystallization; and the last two terms represent changes in the CO_2 mass in the gas phase due to changes in the composition and amount of the gas phase. We consider two special cases of Eq. (8).

CO_2 flux from a stationary magma body

We assume for a stationary magma body that (a) the concentration of CO_2 dissolved in the melt is constant, and (b) the composition of the gas phase is constant. We justify these assumptions below. After substituting the magma crystallization rate ($\mathrm{d}M_x/\mathrm{d}t$) for $-\mathrm{d}M_m/\mathrm{d}t$

into Eq. (8) and eliminating terms, we obtain a mass balance equation for the flux of CO_2 across the boundary of an open-system stationary magma body:

$$F^s_{CO_2}(t) = C^m_{CO_2} \frac{dM_x}{dt} - X^m_{CO_2} \frac{dM_g(t)}{dt} \frac{W_{CO_2}}{W_g} \tag{9}$$

The first term is the internal gas production rate by crystallization, and the second term is the storage change rate. Equation (9) is summarized on the left side of Fig. 7.

Validation of Eq. (9) using field data is not possible. However, the validity of Eq. (9) depends only on conservation of mass and the assumptions that the concentrations of CO_2 in the melt and gas are invariant. A detailed model of petrological and geochemical evolution might be used to evaluate some of the terms in Eq. (8). However, the errors caused by compositional changes are probably small relative to errors arising from the imprecise measurement of volcanic gas fluxes.

CO_2 flux from an ascending magma body

We assume that during magma ascent most of the CO_2 released is generated by decompression, rather than crystallization. We also assume that the composition of the gas phase is constant. The total error of the first assumption is believed to be small relative to the uncertainty of CO_2 flux measurements. The assumption of invariant gas compositions during magma ascent is reasonable based on the much smaller fugacities of H_2O, SO_2, and H_2S relative to the fugacity of CO_2 (using realistic volatile abundances) for magma ascent at pressures greater than 200 MPa.

The flux of CO_2 across the boundary of an ascending isothermal magma body is obtained by substituting the ascent velocity ($V = dZ/dt$) into Eq. (8) and by using the above assumptions to eliminate terms. The result is:

$$F^a_{CO_2}(t) = -M_m(t)\, V(t) \frac{\partial C^m_{CO_2}}{\partial P} \frac{dP}{dZ} - X^g_{CO_2} \frac{dM_g(t)}{dt} \frac{W_{CO_2}}{W_g} \tag{10}$$

where the superscript *a* refers to magma ascent contributions to the total CO_2 flux.

Validation of Eq. (10) using field data is not possible. Equation (10) will tend to overestimate the mass of ascending melt in proportion to the amount of CO_2 generated by crystallization relative to the amount generated by decompression. Equation (10) is exact, however, for magma ascent with negligible crystallization and invariant gas composition.

If there is no change in the mass of CO_2 in the gas (i.e., the second term in Eq. (10) is zero), we can integrate Eq. (10) to obtain

$$\frac{\int_{t_1}^{t_2} F^a_{CO_2}\, dt}{\dfrac{-\partial C^m_{CO_2}}{\partial P} \dfrac{dP}{dZ}} = M_m \int_{t_1}^{t_2} \frac{dZ}{dt}\, dt = M_m [Z_2 - Z_1] \tag{11}$$

The numerator on the left side is the mass of CO_2 released by an ascending magma body from time t_1 to t_2. The first and second terms in the denominator, respectively, are the pressure derivative of the CO_2 concentration in the melt and the lithostatic pressure gradient.

The right side of the equation is the product of the mass of ascending melt and the change in vertical position. Because Eq. (11) assumes that the melt is gas-saturated, the term (Z_2–Z_1) represents the gas-saturated magma ascent distance. After the CO_2 flux due to an ascending magma body is obtained (e.g., by one of the flux separation methods described above), then it is possible to use the mass of CO_2 released during episodic magma ascent and the left side of Eq. (11) to infer the product of melt mass and ascent distance.

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