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Long-term changes in quiescent degassing at Mount Baker Volcano, Washington, USA; Evidence for a stalled intrusion in 1975 and connection to a deep magma source

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ABSTRACT

Long-term changes have occurred in the chemistry, isotopic ratios, and emission rates of gas at Mount Baker volcano following a major thermal perturbation in 1975. In mid-1975 a large pulse in sulfur and carbon dioxide output was observed both in emission rates and in fumarole samples. Emission rates of CO_2 and H_2S were ~950 and 112 t/d, respectively, in 1975; these decreased to ~150 and <1 t/d by 2007. During the peak of the activity the C/S ratio was the lowest ever observed in the Cascade Range and similar to magmatic signatures observed at other basaltic–andesite volcanoes worldwide. Increases in the C/S ratio and decreases in the CO_2/CH_4 ratio since 1975 suggest a long steady trend back toward a more hydrothermal gas signature. The helium isotope ratio is very high (>7 R_c/R_A), but has declined slightly since the mid-1970s, and $\delta^{13}C$ – CO_2 has decreased by $\geq 1\%$ over time. Both trends are expected from a gradually crystallizing magma. While other scenarios are investigated, we conclude that magma intruded the mid- to shallow-crust beneath Mount Baker during the thermal awakening of 1975. Since that time, evidence for fresh magma has waned, but the continued emission of CO_2 and the presence of a long-term hydrothermal system leads us to suspect some continuing connection between the surface and deep convecting magma.

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1. Introduction

Even during quiescence, many volcanoes emit significant quantities of gas (primarily H₂O, CO₂, and sulfur species) and sustain large heat fluxes without demonstrating other signs of unrest such as seismic swarms or surface deformation. Andres and Kasgnoc (1998) cite ~50 volcanoes that degassed SO₂, as well as CO₂, continuously from 1973-1998, many of which did not erupt during that time period including Vulcano, Italy (Chiodini et al., 1992; Harris and Maciejewski, 2000), Martin Volcano (Doukas and McGee, 2007), and Santa Ana, El Salvador (Olmos et al., 2007). In addition to these, many quiescent volcanoes emit CO₂ and H₂S with little to no measureable SO₂; these include volcanoes like Mount Baker, Washington (McGee et al., 2001), Illiamna Volcano and Mount Mageik, Alaska (Doukas and McGee, 2007), and Solfatara in Italy (Martini et al., 1984). The heat and gas released from volcanoes during inter-eruptive periods is ultimately tied to the state of activity and the amount of magma beneath the volcano, as well as geochemical reactions in overlying groundwater and hydrothermal systems (Giggenbach, 1987), and emission rates are commonly used as an indicator of the rate of magma supply to the volcano (McGee et al., 2000; Chiodini et al., 2002; Evans et al., 2002). However, there are relatively few quiescent degassing volcanoes with long-term geochemical records that show large changes in gas composition and emission rates without associated geophysical signs of unrest. Mount Baker, in the Cascade Range volcanic arc of northwestern USA, provides a good opportunity to study such a volcano.

Mount Baker has shown little evidence of seismic unrest, yet actively emits both CO₂ and H₂S (McGee et al., 2001), hosts fumaroles with temperatures above boiling point, and has some of the highest helium isotope ratios in the Cascade Range (Symonds et al., 2003b). Thermal activity and gas emissions increased in 1975, which suggested the possibility of a stalled magmatic intrusion, but the cause of the unrest has never been fully explained (Frank et al., 1977; Malone, 1979). We combine the long-term record of gas emission and chemistry measurements, starting in 1974, with new measurements collected in 2006 and 2007 to investigate non-eruptive degassing at Mount Baker, and to constrain the possible causes of elevated heat and gas output observed in 1975. The results provide a backdrop for assessing the changes in degassing characteristics at largely quiescent volcanoes such as those located in the Cascade Range, and show that regular monitoring of volcanoes with very low emissions and activity can be useful in interpreting deep magmatic processes ongoing beneath quiet volcanoes.

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2. Geologic setting and history of Mount Baker

Mount Baker is the youngest edifice in a large Quaternary volcanic field that has been continuously active for the past 1.3 million years (Hildreth et al., 2003). The volcano is located about 50 km east of the town of Bellingham in northwest Washington (Fig. 1) and reaches a height of 3286 m. Hildreth et al. (2003) have documented two Holocene eruptions: at 9.8 ka, basaltic–andesite lavas were extruded south of Mount Baker and covered at least 19 km² and at 6.5 ka, $\sim 0.1~{\rm km}^3$ of black andesitic ash was erupted in a sub-Plinian column from Sherman Crater, located approximately 1 km south of the summit. A hydrovolcanic explosion occurred at Sherman Crater in 1843 and may have been followed by phreatomagmatic activity (inferred from eyewitness observations), but by 1891 Sherman Crater was observed to be filled with ice (Scott and Tucker, 2003).

In March 1975, an approximately order of magnitude increase in thermal activity and gas emission occurred at Sherman Crater. The area of snow-free ground increased by a factor of 3, heat flow estimates suggest an increase from 2 to 30 MW, and almost half of the glacier that filled Sherman Crater melted. Fumarole temperatures reached 131 °C in Sherman Crater in 1975 (Frank et al., 1977; Malone, 1979). Fine-grained ejecta were emitted by fumaroles throughout the

spring and summer of 1975 (to a distance approximately 100–300 m from fumarolic vents), but petrographic analysis revealed no juvenile material (Frank et al., 1977). Rates of gaseous sulfur emission, measured with a Meloy model 160-2 flame sulfur analyzer, were determined by airborne surveys in 1975, with 30 tons per day (t d^{-1}) measured on March 27 and 112 t d^{-1} on June 30 (Radke et al., 1976). An emission rate measurement on November 18 was similar to that of the June observation (Frank et al., 1977). The sulfur in the Radke et al. (1976) emissions measurements is assumed to be H₂S based on fumarole and plume compositions reported in Frank et al. (1977) and the fact that SO₂ has never been detected in any subsequent analyses of gas from Mount Baker (e.g., SO₂ <50 ppmv in 1980 samples). H₂S often results from the hydrolysis of magmatic SO2 by liquid water within a volcano's hydrothermal system (Doukas and Gerlach, 1995) and there was a well established hydrothermal system at Mount Baker prior to the 1975 increase in thermal activity.

Responses to the elevated activity in 1975 included deployments of additional seismometers and borehole tiltmeters, plus gravity-change and spirit-level surveys. Despite the increased seismic coverage in the area by late summer 1975, which lowered the detection threshold to ML = 0, few earthquakes were recorded in the Mount Baker region (Malone, 1976; Malone and Frank, 1976). Data from spirit-level

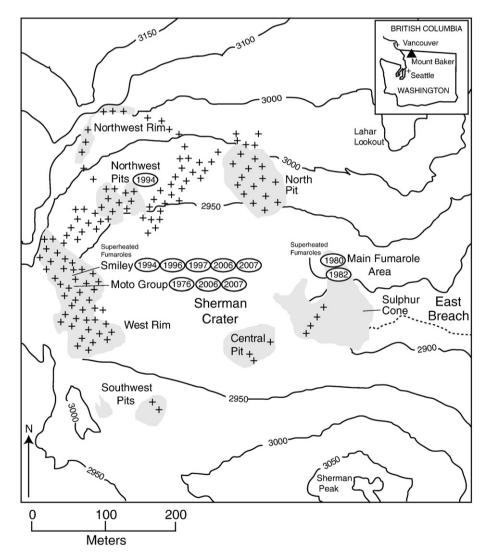


Fig. 1. Location Map of Sherman Crater. The grey shaded areas show regions of thermal ground mapped in 1975 (Frank et al., 1977), and the crosses show how the fumarole activity had migrated to the west by 1978/1979 (Shafer, 1980). The ovals contain the dates of sampling in each area. Over the years, most samples were collected in the East Breach and West Rim areas as these areas both had superheated fumaroles and the strongest emission (Table 1). The exact location of the 1994 sampling on the North Wall is not known. Locations for the 1978 samples are unknown.

measurements and borehole tiltmeters during the summer of 1975 were conflicting and of small magnitude, suggesting negligible deformation (Frank et al., 1977). Microgravity measurements, in contrast, showed a significant decrease at Sherman Crater between May 1975 and October 1976. Malone (1979) prefers a model of mass lost by fumarolic activity to explain the gravity results, but notes that an elevation increase at Sherman Crater could also explain the results. No deformation data exist to test that possibility, however, EDM surveys in 1981 and 1983 showed no signs of cm-scale or greater deformation (Chadwick et al., 1985), and seismicity in the region remained low through 2007, although airborne gas emissions and fumarole temperatures after 1975 continued to be elevated. High ³He/ ⁴He ratios (7.4–7.7) led Symonds et al. (2003b) to infer the presence of fresh magma at shallow (<5 km) depths, and to suggest that sustained high helium isotope ratios indicate continued additions of new magma at depth. However, there is no independent evidence to suggest that the magma is indeed shallow or that there has been periodic recharge to a shallow magmatic reservoir.

3. Measurements

Fumaroles in Sherman Crater (Fig. 1) were sampled for steam and gases such as CO₂, H₂, He, CH₄, H₂S, SO₂, HCl, HF, N₂, O₂ and Ar (Table 1). Samples collected between 1975 and 1997 were collected in evacuated bottles with and without NaOH using standard fumarole sampling techniques. 2006 and 2007 samples were collected using a titanium tube that was placed in the vent, while the surrounding crack was packed with mud to ensure good steam and gas flow and minimal air contamination. The titanium tube was fitted with temperatureresistant Tygon tubing to connect it to an evacuated bottle, which was placed in snow. Steam condensed within the tube and flowed down into the bottle, while other gases flowed directly into the bottle to fill up the head space. Constituents of the head space were measured by gas chromatography using both helium and argon carrier gases and a thermal conductivity detector. A vacuum inlet system allowed measurement of gas pressure within the sample tube and calculation of gas/steam ratios. All gas geochemistry samples (1976-present) were analyzed at USGS laboratories in Menlo Park, CA. Splits of gas were analyzed for C and He isotopes by mass spectrometry at several USGS laboratories.

Emission rate measurements were made by aircraft downwind of the main crater in September, 1980 and 1983 using a COSPEC (correlation spectrometer) for SO₂ and a Miran for CO₂, but no SO₂ or anomalous CO₂ were measured on either occasion (T. Casedevall, personal communication). The Miran instrument, however, has a very large sample cell, which means that low emission rates (on the order of hundreds of tons per day) would be not detected. Emission rates since 2000 have been measured using a closed-path LICOR spectrometer (LI-6252, LI-COR Inc.) for CO₂; this instrument was calibrated using a zero and 496 ppm standard gas on the day of the flight in 2007. SO₂ and H₂S plume concentrations were measured with Interscan electrochemical analyzers (model 4172 and 4170, respectively, which are factory calibrated, e.g., McGee et al., 2001). Plume gas (i.e., external air) was pumped to the instruments in an open-flow arrangement with the tubing extended in front of the aircraft (Gerlach et al., 1997). Gas concentrations, GPS coordinates, and air temperature and pressure were logged at one sample per second. Transects were flown perpendicular to the wind direction with a roughly 60-m vertical spacing between transects. The average and standard deviation of background CO2 concentrations were calculated for each pass (i.e. elevation) from data collected outside the plume. The wind direction and speed were determined by performing neutral circles (Doukas, 2002). The method for calculating the gas emission rate in 2007 was identical to that described in Gerlach et al. (1997) and McGee et al. (2001).

4. Results

4.1. Declining emission rates

Magmatic gas discharge rates are a fundamental metric of volcanic activity. To compute CO_2 and H_2S emission rates prior to 2000, we used a combination of airborne measurements and fumarole gas ratio measurements. This procedure provides an extended time-series

Table 1
Gas geochemistry (in mol% and molar ratios) of fumaroles from Sherman Crater, Mount Baker, Washington (helium isotope ratios corrected for air contamination). SO₂ and HF were below detection limits in all samples analyzed, and HCl was rarely detected.

Date	Data ¹	Area ²	T °C	T °C depth³	CO ₂	H ₂ S	H ₂	CH ₄	Не	N ₂	02	Ar	Gas/steam	N ₂ /Ar	$\delta^{13}C_{CO2}$	$R_{\rm c}/R_{\rm A}$
1976	UD (1)	WR, Moto	89.6	231	81.9	12.5	0.04	0.002	0.004	5.1	-	0.06	_	85	-	-
1978	UD (2)	Unknown	_	225	79.5	19.1	0.003	0.011	0.002	1.3	_	0.02	_	59	_	7.62^4
		Unknown	_	222	81.1	9.4	0.087	0.04	-	8.8	_	0.15	_	59	_	_
		Unknown	-	218	92.6	5.5	0.17	0.14	-	1.4	_	-	-	-	-	-
1980	UD (3)	EB	99	142	78.4	9.2	0.0002	0.11	-	11.4	0.02	0.28	-	41	-	-
		EB	103	167	91.7	7.45	0.0019	0.83	0.0003	0.55	_	0.002	-	229	-	-
		WR	100	_	55.1	2.6	0.024	0.08	-	32.7	5.26	-	-	-	-	-
		WR	113	217	94.5	4.8	0.17	0.11	0.0006	0.47	_	< 0.001	-	-	-	-
1982	UD (4)	EB – Main	86	178	95.2	4.5	0.003	0.27	0.001	-	_	-	-	-	-	-
		EB – Main	104	181	94.2	5.5	0.003	0.26	0.0029	-	_	-	-	-	-	-
		EB – Main	106	171	97.2	2.5	0.003	0.27	0.0016	-	-	-	-	-	-	-
		EB – Main	117	171	94.1	5.6	0.002	0.29	0.0009	-	-	-	-	-	-	-
		EB – SE	134	167	88.9	10.8	0.001	0.22	0.0038	-	-	-	-	-	-	-
		EB – SE	134	190	89.9	9.8	0.003	0.21	0.0003	-	-	-	-	-	-	-
1994	(5)	WR, Smiley	135	227	93.8	5.1	0.31	0.23	0.00063	0.54	< 0.0005	0.0013	-	415	-6.1	7.42
		N. Wall	150	221	93.6	5.1	0.29	0.38	0.00091	0.59	< 0.002	0.0016	-	369	-5.9	7.49
1996	(5)	WR, Smiley	137	-	93.9	5.1	-	0.47	0.00074	0.47	0.0006	0.0014	-	334	-6.7	7.45
1997	(5)	WR, Smiley	134	217	93.0	5.2	0.25	0.44	0.00001	1.06	0.01	0.0012	0.018	852	-6.9	7.7^{5}
2006	(6)	WR	90	221	94.9	3.3	0.448	0.704	0.001	0.69	_	0.0013	0.013	517	-6.7	7.36
	(6)	WR	90	242	94.3	3.6	0.933	0.708	0.0008	0.54	-	0.0025	0.012	214	-6.7	7.31
2007	(6)	WR, Smiley	-	239	90.9	4.2	0.704	0.613	0.0009	2.9	0.61	0.025	0.025	-	-7.32	7.44
	(6)	WR, Moto	-	225	90.4	5.0	0.026	0.625	0.0009	3.25	0.68	0.033	0.010	361	-7.2	7.48
	(6)	WR, Moto	-	237	91.4	4.2	0.044	0.616	0.0009	3.11	0.61	0.03	0.005	349	− 7.2	7.34

¹ UD = unpublished data; sample collected by (1) Malone, (2) Johnston, (3) Casadevall and Greenland, (4) McGee & Sutton, (5) Symonds et al., 2003a,b, (6) this study; ² WR = West Rim, EB = East Breach; ³ 'depth' refers to the temperature calculated for the hydrothermal reservoir using the D'Amore and Panichi (1980) geothermometer, see text; ⁴ Poreda and Craig, 1989, ⁵ included in average in trend in Fig. 3. Most-recent samples: gas composition and d¹³C determined at USGS Menlo Park; helium isotopes at USGS laboratories.

estimate of emissions reaching back to 1975. For example, to estimate the 1976 emission rate of CO_2 the CO_2/H_2S wt. ratio from the 1976 fumarolic sample was multiplied by the H_2S emission rate, and likewise the H_2S/CO_2 wt. ratio from the 1997 fumarolic sample was multiplied by the CO_2 emission rate in 1998.

Total gas emissions have declined with time since 1976 (Fig. 2), with $\rm H_2S$ declining from 112 t d $^{-1}$ in 1975 to <1 t d $^{-1}$ in 2007, and $\rm CO_2$ declining from ~950 t d $^{-1}$ in 1975–76 to 150 t d $^{-1}$ in 2007. The detail of this decline is unknown because no measurements were made in the 1980s or early 1990s. Gas geochemistry (discussed below) suggests a rapid change in chemical composition, and thus emission rates might have also dropped substantially between 1975 and 1982. The measurements from 1998 to present suggest a gradual decline in both gases over this period (Fig. 2).

4.2. Gas geochemistry and temperatures

After water vapor the gases from Mount Baker are most enriched in CO₂ and H₂S, and have minor amounts of CH₄, He, and H₂. Species typical of shallow degassing of magma have not been detected (SO₂ and HF) or only rarely detected (HCl) in emissions at Mount Baker in the past (Symonds et al., 2003a,b), suggesting that there is efficient conversion from magmatic SO₂ to H₂S, and that HCl and HF remain dissolved in the magma because of its considerable depth. Scrubbing (i.e., the process where magmatic gas is removed from the gas stream by reacting with groundwater or precipitated as minerals) of magmatic volatiles has also been suggested for the lack of typical high-temperature magmatic species (SO₂ and halogens) at Mount Baker (Symonds et al., 2003a).

Abrupt changes in the H₂S/CO₂ ratio in fumarole samples occurred during and immediately following the 1975 unrest (Fig. 3). Between August 1974 and March 1975, the H₂S/CO₂ ratio of fumarole samples increased by only a fraction, but between March and August of 1975, the H₂S/CO₂ ratio increased from less than 0.01 to 0.2 (Fig. 3). These

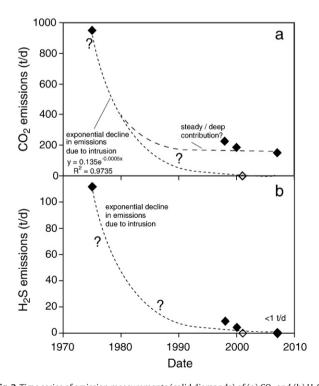


Fig. 2. Time series of emission measurements (solid diamonds) of (a) CO_2 and (b) H_2S at Mount Baker, 1976–2007 (the white open diamond was a high-wind day and emissions could not be measured). The dashed line is based on the decay of the H_2S/CO_2 molar ratio in fumaroles (Fig. 3) and has the form of $e^{-0.0005*t}$, where t is the time in days since the 1975 unrest.

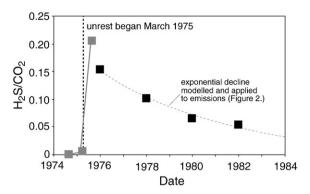


Fig. 3. Geochemical trends in Mount Baker fumaroles, 1974–1984. H₂S/CO₂, grey squares indicate data from Kiver and Steele, 1976, black squares indicate more complete fumarolic analyses (Table 1). The grey dashed line shows the exponential decline of the ratio. The black dashed line shows when the unrest began.

were the only two species measured at that time (Kiver and Steele, 1976). In the years since these data were collected, the H_2S/CO_2 ratio decreased and then stabilized at ~ 0.05 .

Long-term trends in the geochemistry (from 1975–2007) are more easily visualized by observing the $\rm CO_2/CH_4$ and $\rm CO_2/H_2S$ ratios (Fig. 4). Following the unrest, $\rm CO_2/CH_4$ was very high, indicating a

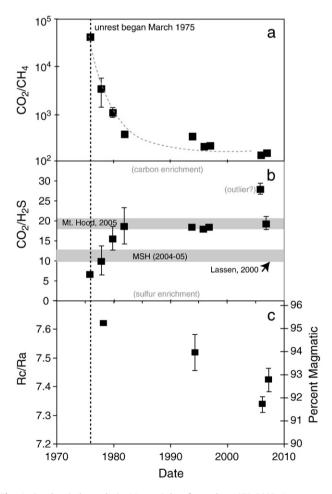


Fig. 4. Geochemical trends in Mount Baker fumaroles, 1976–2007. Averages and standard errors for molar ratios of (a) CO_2/CH_4 (b) CO_2/H_2S (or C/S) and (c) He isotopic signatures. The grey bands indicate C/S ratios calculated from airborne plume work at the various volcanoes (Gerlach et al., 2009). The left axis on (c) is the R_c/R_A ratio and the right axis is the percent of magmatic component assuming endmembers of mid-ocean ridge basalts ($R_c/R_A = 8$) and crustal components ($R_c/R_A = 0.02$). The point in the mid-1990s is the average of four analyses.

dramatic enrichment in CO₂. Since ~ 1978, there has been increasing CH₄ and decreasing H₂S in gas samples, both absolutely and relative to CO₂ (Fig. 4, Table 1). The absolute concentration of CH₄ has increased from 0.002 to ~0.7 mol% and H₂S has decreased from roughly 12 to 4 mol%. CO₂/CH₄ ratios have decreased from ~41,000 to 150 and CO₂/H₂S has increased from 7 to ~19 (Fig. 4a and b). Finally, the carbon isotopic signature of the CO₂ has decreased slightly in the last 10 years or so (δ^{13} C_{CO2} = -5.9% in 1997, -6.7 in 2006, and -7.3 in 2007).

The helium isotopic signature measured at Mount Baker in 1978 ($R_{\rm c}/R_{\rm A}\!=\!7.62$) is one of the highest measured in all of the Cascade Range, exceeded only by He observed in spring waters near Three Sisters, Oregon, ($R_{\rm c}/R_{\rm A}\!=\!7.8$, Evans et al. (2004). Average He isotopic values fell from ~7.6 to 7.4 between 1978 and 2007 (Fig. 4c.). If we assume a magmatic endmember of mid-ocean ridge basalts (MORB=8 R/Ra) and crustal rocks (0.02 R/Ra), this would suggest that the magmatic contribution of He has decreased only a few percent since 1976, from >95% to roughly 92% (Fig. 4c).

The fumaroles at Mount Baker have remained at high temperatures for an extended period of time. However, prior to the 1975 disturbance, all fumarole temperatures were at or below 90 °C (Frank et al., 1977). In contrast, during the 1975 unrest Frank et al. (1977) measured temperatures of ~ 130 °C in the main fumarole area in the East Breach region of the crater (Fig. 1). These temperatures remained elevated through subsequent years, and superheated fumaroles also appeared in the West Rim region (Fig. 1) by the early 1980s. West Rim fumarole temperatures of 132–135 °C were documented in 1982 and 1994, 1996, and 1997 (Symonds et al., 2003a,b). No superheated fumaroles were observed in the West Rim area in 2006; temperatures were ~ 90 °C. In 2007, technical difficulties prevented temperature measurements, so it is not known if superheated fumaroles still exist anywhere within Sherman Crater.

If the D'Amore and Panichi (1980) geothermometer is used to calculate hydrothermal reservoir temperature, the results (Table 1) suggest that gases have equilibrated at roughly the same temperature since the first full samples were analyzed in 1976. The highest equilibrium temperatures for all years fall in the range of 217–242 °C; all of these samples were collected from the West Rim location. Samples from the East Breach region, however, suggest temperatures between 142 and 190 °C (Table 1, Fig. 1). Multiple reservoirs with different temperatures have also been proposed at Mount Hood, another Cascade Range volcano of comparable height (Nathenson, 2004)

In contrast to steady reservoir temperatures over time, evidence suggests that the heat flux has decreased since the 1970s, or that the distribution of hot zones in Sherman crater has reorganized since that time (Fig. 1). Shafer (1980) noted that the fumarolic/thermal activity in 1975 in and around Sherman Crater was most vigorous in the East Breach/Main Fumarole area and in the Central Pits (Fig. 1), but by March 1979 the activity was most intense along the West and Northwest Rims, had reduced in the East Breach/Main Fumarole area, and was absent from the Central Pit region. These patterns have remained relatively constant since that time (Fig. 1). Specifically, the Central Pit and one of the Southwest Pits are no longer active and are now filled with snow, and there is also evidence that the West Rim fumarole area has reduced in size.

5. Discussion

At least four hypotheses might be invoked to explain the 1975 activity and continuing high fumarole temperatures and degassing rates: (1) magma began intruding to shallow levels in 1975 and continues to do so (Symonds et al., 2003b); (2) the 1975 activity resulted from a stalled intrusion, with little subsequent magmatic activity (e.g., (Roman et al., 2004), and emissions since 1975 represent gas lost from the stalled intrusion plus hydrothermal gases; (3) convection in a deep

(~10 km) magma source, to which a path was opened in 1975, supplies thermal energy and gas to the surface (e.g., Williams-Jones et al., 2003; Werner et al., 2008); and (4) boiling in the hydrothermal reservoir increased, either because of a more permeable connection to the surface (depressurization) or a better connection to a deep magmatic heat source that does not allow passage of new magmatic gas. These four general scenarios are discussed below.

We consider first scenario 4, increased boiling in a hydrothermal system, because increases in gas and heat emissions would be expected, but this does not necessarily require new input of magmatic gas. Because of the different solubilities of H₂S, CO₂, and CH₄ in hydrothermal waters (Giggenbach, 1980; Chiodini et al., 2002), an increase in H₂S/CO₂ as occurred in 1975 (Fig. 3), and a simultaneous increase in CO₂/CH₄, as might be inferred from Fig. 4, could have resulted from increased boiling where vapors became enriched in the more soluble gases (H₂S>CO₂>CH₄). An increase in boiling could have occurred by increasing the temperature of the hydrothermal aquifer (without addition of magmatic volatiles) or by depressurization of the reservoir. If such a permeability enhancement invoked in scenario 4 then gradually disappeared due to hydrothermal mineralization or other conduit processes, many of the long-term trends observed since 1975 (Fig. 4a and b) would be expected or at least possible.

However, simple boiling of the hydrothermal system is difficult to reconcile with the very low CO₂/H₂S and CH₄/CO₂ ratios in fumarolic gas (Symonds et al., 1994; Fischer et al., 1997) and the magnitude of CO₂ and H₂S emissions in 1975–1976 (Tables 1 and 2). The fumarolic C/S ratio in 1976 was one of the lowest ever measured in the Cascade Range (Fig. 3b), even lower than the ratio in eruptive gases at Mount St. Helens (Gerlach et al., 2009). Such a low ratio would not be expected to result from increased boiling in a hydrothermal system because S is typically lost to groundwaters and hydrothermal mineral phases (Symonds et al., 1994, 2003a). The low ratio is, however, similar to that for magmatic gases observed at other active arc volcanoes. Typically magmatic gases at andesitic arc volcanoes have C/ S ratios <10, whereas hydrothermal gases typically have values >10 (Symonds et al., 1994; Fischer et al., 1997; Scaillet and Pichavant, 2003). Furthermore, the 950 t d⁻¹ CO₂ emission rate calculated for 1975-76 would also require the pre-existing hydrothermal reservoir to be extremely CO₂-rich. Yet, evidence for such a reservoir is virtually absent in the groundwater system. No CO2-rich springs occur at Mount Baker although such springs are frequently found on the flanks of other western USA volcanoes where CO₂ leaking up from depth dissolves in overlying groundwater (e.g., at the Mammoth and Lassen volcanic centers; Evans et al., 2002).

The alternate hypothesis, that Mount Baker received an injection of relatively S-and CO₂-rich magmatic gas from depth, is favored. In this hypothesis (scenarios 1–3), increases in H₂S would be directly related to the high content of sulfur gases in the magmatic component, namely SO₂ and H₂S, while CH₄ would decrease relative to CO₂ because CH₄ has a low concentration in magmatic gases and is typically formed in hydrothermal environments (Giggenbach, 1980;

Table 2Gas emissions from Mount Baker, Washington.

Date	Reference	H_2S (t d^{-1})	CO ₂ (t d ⁻¹)
1975	Radke et al. (1976)	112	950*
1998	Gerlach et al. (2009)	9.7*	225
2000	McGee et al. (2001)	5.5	187
2001	This study	nd	nd
2007	This study	0.3	~150

^{*} The emission rates were determined by ratio with the fumarole chemistry. The 1975 CO_2 emission rate was determined using the 1976 fumarole ratio of CO_2/H_2S and the 1998 H_2S was determined using the 1997 fumarole ratio of H_2S/CO_2 , nd = not detected, likely due to high wind velocities during measurement.

Symonds et al., 2003a). Similar trends in gas chemistry were observed at Nisyros Island in Greece (Chiodini et al., 2002), where the H_2S/CO_2 ratio increased on average from 0.15 to \sim 0.35 (as compared to the increase from 0.01 to 0.2 at Mount Baker) and CO_2/CH_4 increased from 200 to 500. At Nisyros, these changes occurred gradually (over 4 years), and were attributed to increases in magmatic fluids introduced to the hydrothermal system. At Mount Baker, the initial input is thought to have occurred over a 3-month period (Fig. 3).

After the initial changes observed in 1975-1976 where H₂S and CO₂ increased (Fig. 3), the geochemistry of Mount Baker fumaroles has gradually trended toward higher relative contents of hydrothermal components, such as CH₄ (Fig. 4), and the heat flux has decreased (as inferred from decreasing areas of thermal ground in Fig. 1) from 1975 to 2007. Decreases in CO₂/CH₄ (Fig. 4a.) likely reflect multiple processes. First, the initial rapid drop of CO₂/CH₄ from near 41,000 to ~600 between 1976 and 1978 was probably caused by the passing of a large CO₂ pulse associated with deep magma injection. The equilibrium reaction between CO2 and CH4 depends on redox conditions and is also temperature dependent, i.e., CH₄ increases relative to CO₂ with decreasing temperature (Fiebig et al., 2004). The value of log (X_{CH4}/X_{CO2}) in every sample since 1975–80 falls in the range of -2.5to -2.17, which, at reservoir temperatures of $\sim 150-240$ °C, falls on the D'Amore and Panichi (1980) equilibrium line (see Fig. 7 in Fiebig et al., 2004). Therefore, the slow decrease in CO₂/CH₄ since 1979 most likely indicates more reaction of CO₂ to CH₄ as equilibrium between the two species is gradually approached over time.

Although CH₄ can also indicate a sedimentary or shallow organic component in the gas phase, no sediments are known to underlie Mount Baker (Hildreth et al., 2003) and the summit is devoid of vegetation. Also, Symonds et al. (2003b) point out that the $\delta^{13}\text{C-CH}_4$ values (-21.7 to -24.3%) in gas samples collected during the 1990s are the heaviest in the Cascade Range and seem to be incompatible with solely a sedimentary source.

The gradual increase in CO₂/H₂S (or C/S) from 7 to 19 between 1976 and 1982 (Fig. 4b.) could represent a number of processes expected to follow the passing of a large pulse of magmatic gas rich in C and S. CO₂ is relatively conservative in high-temperature hydrothermal environments, while S is more easily removed (Giggenbach, 1996; Symonds et al., 2001, 2003a). For instance, the increasing presence of groundwater as heat flux declines would cause more H₂S to dissolve, and deposition of hydrothermal minerals such as native sulfur, alunite, pyrite in the conduit would cause decreases in S in emissions (Giggenbach, 1996; Symonds et al., 2001, 2003a). Between 1982 and 2007, the fumarolic ratio essentially stabilized near a value similar to that of Mount Hood, perhaps indicating that the hydrothermal systems in each of these Cascade Range volcanoes have similar effects on the magmatic gases that rise up through the volcanic edifices. Regardless, the substantially higher CO₂/H₂S ratios in plume emissions relative to fumarolic gases in more recent years would argue that the processes described above are active at Mount Baker.

Isotopic analysis of carbon and helium assists in identifying gas sources and how those sources have changed over time. The gradual decline in the δ^{13} C of CO₂ (~ -5.9 in 1994 to -7.3 in 2007) is consistent with progressive, Rayleigh-process CO₂ loss from a cooling intrusion, as discussed by Gerlach and Taylor (1990), and is similar in magnitude to the decline in δ^{13} C that has occurred at Mount St. Helens since the 1980 eruption (Bergfeld et al., 2008). The isotopic signature of He at Mount Baker is one of the highest found in the Cascades (up to ~7.7 R_c/R_A , where R_c is the corrected ratio of ³He to ⁴He relative to is the ratio in air, R_A), but the data suggest that perhaps the mantlederived magmatic component has been declining slightly over the last 30 years. Similar decreases have been observed at other Cascade Range volcanoes over similar timeframes (e.g. Mount Shasta decrease by 0.7 R_c/R_A units over 24 years, Symonds et al., 2003b), which could indicate a reduction in the mantle-derived magmatic component in both cases. The high helium isotopic value led Symonds et al. (2003a,b) to suggest constant input of magma beneath the volcano, but in this situation (our scenario 1) we might also expect the gas emissions to stay more constant, and the magmatic signature of the gas geochemistry to remain unchanged for long periods, which hasn't been the case. The changes in gas chemistry and isotope data, however, do support scenario 2 – a stalled intrusion – in the mid-1970s.

Gas emissions complement and support the above hypothesis that a magmatic intrusion occurred beneath Mount Baker sometime prior to August 1975, and likely coincident with the March 1975 increase in thermal activity. The level of gas emission reached in 1975 (112 t d^{-1} H_2S and 950 t $\Bar{d}^{-1}\,CO_2)$ is similar to that of other hypothesized 'stalled intrusions' in Alaska (Roman et al., 2004; Werner and Doukas, 2008), but low compared to eruptive activity. Studies have shown that emissions from eruptive events at similar ice-clad andesitic volcanoes typically reach levels of CO₂ emission in excess of 2000 t d⁻¹, and often greater than 10,000 t d^{-1} (Hobbs et al., 1991; Doukas and Gerlach, 1995; McGee et al., 2006). These studies also show that SO₂ emission is typically greater than 1000 t d⁻¹. H₂S emission has not been measured extensively during eruptive activity at these volcanoes, but the measurements that exist suggest very low H₂S relative to SO₂ emission (McGee et al., 2006), and no SO₂ has been detected at Mount Baker. Emissions of CO₂ and H₂S from Mount Baker have been sustained at a low level since 1975 (Radke et al., 1976; McGee et al., 2001), but the nature of the initial decline is unknown. In most cases of unrest (with or without eruption) at the Alaskan Cook Inlet volcanoes, emissions of CO₂ and H₂S decline rapidly the first year and are below detection limits after ~ two years (Casadevall et al., 1994; Roman et al., 2004; McGee et al., 2006).

The sampling frequency of emission rates at Mount Baker does not allow us to know whether or not the emissions dropped off rapidly or gradually after 1975. If we model an emission-rate decay that is consistent with the geochemical trends shown in Figs. 3 and 4, this suggests a decline with the form of $e^{-0.0005*t}$, where t is the time in days, and results in a pattern of declining emissions over a period of many years. While not common, a gradual decline (~30 years) in CO₂ emissions from a single intrusion is possible as shown by the trend observed at the Horseshoe Lake tree kill area at Mammoth Mtn. where CO_2 emissions dropped from an average of 250 t d^{-1} between 1995 and 97 to 38 t d^{-1} in 2006 (Lewicki et al., 2008). However, an alternate hypothesis is that the continued CO_2 emission at 150–200 t d^{-1} since 2000 is the result of a connection to a deeper (perhaps convecting) magma source as proposed in scenario 3. In this scenario, the rapid geochemical changes in 1975 are the result of the emplacement of a shallow magma, but the long-term, sustained CO2 emissions and mantle-type helium isotopic signature are driven by a connection to a deep convecting magma. Other evidence that supports this scenario is the long-term stability of hydrothermal reservoir temperatures implied by geothermometry, and the existence of a long-lived hydrothermal system at Mount Baker.

If an intrusion was emplaced beneath the volcano in 1975, its size can be roughly determined from the integrated amount of $\rm CO_2$ it released. If we subtract the steady source of 'deeply-derived' $\rm CO_2$ emissions (~150–200 t d⁻¹) measured in recent times (since 2000) from the exponential trend in Fig. 2a, a total of ~1750 kilotons of $\rm CO_2$ have been released over 32 years from the intrusion alone (although the sum of the contribution since 1990 would only be 5% of this total). Assuming the intruding magma body was andesitic with density of 2700 kg m⁻³, and a (maximum estimated) $\rm CO_2$ content of 0.6 wt.% (Wallace, 2005), this suggests degassing and/or crystallization of a ~0.1 km³ magma body. This is a minimum estimate because the magma remained at depth and would still retain some $\rm CO_2$, and also the initial $\rm CO_2$ content of the magma could be less.

If we attribute the long-term degassing rate of \sim 150–200 t d⁻¹ CO₂ to deep convecting magma, about 0.003–0.004 km³ yr⁻¹ of this magma must be degassed. This is roughly one quarter to one fifth of the minimum amount of magma degassed annually at White Island Volcano

in New Zealand ($0.015 \text{ km}^3 \text{ yr}^{-1}$, Werner et al., 2008), an open-system volcano with significant evidence for magma convection driving degassing. While magma convection cannot be convincingly invoked for Mount Baker, the amount magma being degassed is large enough to suggest that a connection to a large magma source at depth is reasonable.

6. Conclusions

Gas geochemistry and emissions from Mount Baker during 1974–2007 show evidence for initial increases in magmatic gases rich in S and CO₂, followed by long-term trends towards a more hydrothermal gas endmember, accompanied by decreasing gas and heat flux. Four processes were considered to explain these changes including continuous supply of new magma beneath the volcano, emplacement of a shallow magmatic intrusion, connection to a deep convecting magma body, and increased boiling of the hydrothermal system. The gas geochemistry and emission rates provide convincing evidence of an injection of magmatic gas into the hydrothermal system prior to August 1975 as the result of a small magmatic intrusion at depth. Isotopic data also suggest progressive degassing of a magma body. However, the long-term nature of the decline of the gas emissions and heat flux, which differs from the characteristics of stalled intrusions observed at other volcanoes, suggests that there also could be connectivity to a deeper magma source.

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