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## Chapter 21

# Controls on variations in cristobalite abundance in ash generated by the Soufrière Hills Volcano, Montserrat in the period 1997 to 2010

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**Abstract:** The Soufrière Hills Volcano (SHV) crystallizes cristobalite (crystalline silica) in its lava domes, and inhalation of cristobalite-rich ash may pose a chronic respiratory hazard. We investigate the causes of variation in cristobalite abundance (measured by X-ray diffraction) in ash from dome collapses, explosions and ash venting from 1997 to 2010.

Cristobalite abundance in bulk dome-collapse ash varies between 4 and 23 wt%. During periods of slow lava extrusion ( $<5 \text{ m}^3 \text{ s}^{-1}$ ), cristobalite is abundant (7–23 wt%), which we attribute to extensive devitrification in slow-cooling lava; it can also form rapidly (15 wt% in 2 months), but we find no correlation between cristobalite abundance and dome residence time (DRT). By contrast, during rapid extrusion ( $>5 \text{ m}^3 \text{ s}^{-1}$ ), cristobalite abundance is low (4–7 wt%, similar to that associated with Vulcanian explosions), and correlates strongly with DRT. We attribute this correlation to progressive vapour-phase mineralization or devitrification, and the lack of contamination by older lava. Cristobalite abundance is expected to be  $>7$  wt% for collapse of slowly extruded lava, for ash venting through a dome or for incorporation of hydrothermally altered edifice during explosions; cristobalite abundance is expected to be  $<7$  wt% for collapse of rapidly extruded lava, for ash venting without dome incorporation and from Vulcanian explosions at SHV.



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Cristobalite is a high-temperature, low-pressure crystalline silica polymorph that may crystallize as a meta-stable phase in dome lavas, and persists at ambient conditions. In industrial settings, the silica polymorphs of quartz, cristobalite and tridymite are capable of causing silicosis, a fibrotic lung disease. Crystalline silica is also classed as a Group 1 human carcinogen by the International Agency for Research on Cancer (IARC 1997). The discovery of cristobalite in the volcanic ash from the 18 May 1980 eruption of Mount St Helens, USA, prompted intensive research to determine the silicosis risk from inhaling the ash (e.g. Dollberg *et al.* 1986), but the evidence from a series of toxicological, epidemiological and clinical studies at the time was inconclusive on this point (see Horwell & Baxter 2006 for a review). In the event, exposure was short-lived and this substantially reduced public concern. Today, the chronic pathogenicity of cristobalite in volcanic ash is still under debate (Horwell *et al.* 2012).

The Soufrière Hills Volcano (SHV), Montserrat began its current eruption in July 1995. Lava dome growth started in late 1995 and has continued, intermittently, in a series of five phases (Wadge *et al.* 2014). Lava domes are inherently unstable, and are prone to partial or full collapses that generate pyroclastic density currents (PDCs) and associated co-PDC ash plumes. There have also been more than 100 Vulcanian explosions during the eruption. Baxter *et al.* (1999) observed that cristobalite is generated within the Soufrière Hills dome and is abundant in the co-PDC ashfall. Horwell *et al.* (2003) characterized the cristobalite in different grain-size fractions and found that the cristobalite was concentrated in the finest, respirable fractions ( $<4 \mu\text{m}$  diameter).

Over the course of the Soufrière Hills eruption, numerous mineralogical analyses and risk assessments for respiratory health have

been carried out, based on quantification of cristobalite abundance by X-ray diffraction (XRD) and <sup>29</sup>Si magic angle spinning nuclear magnetic resonance (MAS NMR) spectroscopy (see the review in Baxter *et al.* 2014). Initial analyses were carried out on ash from the first major dome-collapse PDC on 3 April 1996, where 25 wt% cristobalite was found in the sub-10  $\mu\text{m}$  fraction. Similar abundances were found in dome-collapse ash deposited in 1996 and 1997 (10–27 wt%, four samples), in 1999 (30 wt%, one sample) and in August–September 2001 (24–29 wt%, three samples) (Baxter *et al.* 1999, 2014). A routine assessment in 2009–2010 found that cristobalite abundance had dropped during dome growth in Phase V to  $<5$  wt% in the bulk ash. The current study was devised to quantify cristobalite abundance in samples spanning the entire eruption, using a single XRD technique for consistency, in order to determine whether this decrease in cristobalite abundance was real or an artefact of technique progression over time. Eruption parameters, such as dome residence time (DRT), lava extrusion rate and SO<sub>2</sub> flux, were then correlated with the data to investigate the causes of fluctuations in cristobalite abundance.

Assessment of cristobalite abundance in volcanic ash is complicated by the presence of other minerals, in particular, plagioclase feldspar, which is often the dominant mineral in andesitic ash, and makes rapid quantification by XRD challenging owing to a peak overlap with cristobalite. Early studies of Mount St Helens and Soufrière Hills ash addressed this problem by employing the Talvitie method (Talvitie 1951), in which all minerals are dissolved except crystalline silica (Baxter *et al.* 1999). This method, which involves boiling the ash in phosphoric acid for 8 min, is difficult to perform consistently since the final mineral

**Table 21.1.** Sample information, cristobalite and quartz abundance, eruption phase (Wadge et al. 2014) and details of dome longevity for ash samples

Sample ID	Ash type	Location of collection	Distance from volcano (km)	Cristobalite (wt%)	Quartz (wt%)	Date erupted	Date collected	Phase (h = dome growth hiatus)	Maximum dome residence time*	Sample origin
MonExp	Vulcanian explosion	Various	3–8	6.1 <sup>†</sup>	0.9 <sup>†</sup>	25 September–6 October 1997	25 September–6 October 1997	I	None <sup>1</sup>	Ash from various Vulcanian explosion deposits
MVO421	Dome collapse	Next ghaut west of Castle Ghaut, SSH	3	12.8	1.0	4 November–26 December 1997	16 February 1998	I	2 months <sup>2</sup>	Major dome collapse (SW sector) including crater wall – Boxing Day event
MVO1038	Ash venting (and rock falls)	Northern Montserrat	7–8	17.5	0.4	1–14 February 1998	14 February 1998	I	2 months <sup>3</sup>	Ashfall weekly collection, from ash venting and rock falls
MBA5/6/99 (MVO1163)	Dome collapse	Lovers Lane	4	10.8 <sup>†</sup>	1.2 <sup>†</sup>	5 June 1999	5 June 1999	I h	18–25 months <sup>4</sup>	Small east and NE sector dome collapse
MVO1225	Dome collapse	Garibaldi Hill	6	15.9	1.4	29 July 2001	13 August 2001	II	16 months <sup>5</sup>	Major dome collapse of eastern sector of dome – removal of 95% of dome
MVO1226	Dome collapse	Vue Pointe Hotel	7	14.8	1.4	29 July 2001	2 August 2001	II	16 months <sup>5</sup>	Major dome collapse of eastern sector of dome – removal of 95% of dome
MBA12/7/03	Dome collapse	Olveston	7	8.4 <sup>†</sup>	1.6 <sup>†</sup>	12 July 2003	12 July 2003	II	24 months <sup>6</sup>	Total dome collapse – ~200 × 10 <sup>6</sup> m <sup>3</sup> – largest volume to date
MVO1307	Phreatic explosion	?	>3.5	10.5	4.7	28 June 2005	28 June 2005	II h	None <sup>7</sup>	Phreatic explosion
MVO1320	Ash venting	St Georges Hill	3.5	8.1	2.5	5 August 2005	10 October 2005	III	2 months <sup>8</sup>	Ash venting episodes over 2 months
25.11.2005	Dome collapse	Richmond Hill	5	10.4	0	25 November 2005	25 November 2005	III	4 months <sup>8</sup>	Small PDC/rockfall activity
MVO1343	Ash venting	Garibaldi Hill	6	7.2	0	24 January–10 February 2006	10 February 2006	III	6 months <sup>9</sup>	Mostly ash venting from 9–10 February 2006 and rockfalls from dome
MVO1348	Ash venting	Plymouth Police Station	4.5	8.4	2.1	24 January–24 February 2006	24 February 2006	III	7 months <sup>10</sup>	Ashfall collected in tray over 1 month. Mostly ash venting, and rockfalls from dome
21.4.2006	Dome collapse	St Georges Hill	3.5	16.4	0	21 April 2006	21 April 2006	III	9 months <sup>11</sup>	Small PDC/rock fall activity
MVO1498	Dome collapse	St Georges Hill	3.5	22.6	0	20 May 2006	23 May 2006	III	10 months <sup>12</sup>	Total dome collapse: base of ashfall sequence.
MVO1492	Vulcanian explosion	St Georges Hill	3.5	6.8	1.4	21 May 2006	23 May 2006	III	None <sup>13</sup>	Ashfall from Vulcanian explosion
MVO1510	Dome collapse	Not known	Not known	14.4	1.0	30 June 2006	30 June 2006	III	1 month <sup>14</sup>	Partial dome collapse

05.10.09	Ash venting	Garibaldi Hill	6	2.3	0.4	5 October 2009	5 October 2009	V	None <sup>15</sup>	Ash venting from early Phase 5 activity before any new dome growth
09.10.09	Dome collapse	Woodlands	8	5.0	0.6	9 October 2009	9 October 2009	V	1 day <sup>15</sup>	Moderate PDC activity
22.10.09	Dome collapse	Olveston	7	4.3	0.6	22 October 2009	22 October 2009	V	2 weeks <sup>15</sup>	Moderate PDC activity
08.11.09	Dome collapse	Olveston	7	5.5	0.9	8 November 2009	8 November 2009	V	1 month <sup>15</sup>	Moderate PDC activity
20.12.09	Dome collapse	Olveston	7	5.2	1.2	20. December 2009	20 December 2009	V	2 months <sup>15</sup>	Moderate PDC activity
04.01.10	Ash venting	Olveston	7	6.4	2.5	4 January 2010	4 January 2010	V	3 months <sup>15</sup>	Ash venting and PDCs
10.01.10	Vulcanian explosion	Olveston	7	5.6	0.46	10 January 2010	10 January 2010	V	3 months <sup>15</sup>	Vulcanian explosion ash
11.02.10	Dome collapse	Jack Boy Hill	7	6.6	2.44	11 February 2010	19 February 2010	V	4 months <sup>15</sup>	Major dome collapse

\*Maximum viable dome residence time given, but, in some circumstances, minor quantities of residual dome material may have been incorporated.

<sup>†</sup>Data from Horwell *et al.* (2010a). NB. The data in this chapter differ from Horwell *et al.* (2010a) due to the use of a different cristobalite standard here. Data from Horwell *et al.* (2010a) have been adjusted using a correction factor.

<sup>1</sup>Ash generated during Vulcanian explosions but may have incorporated a very small amount of edifice or residual dome material. The sequence of explosions was probably triggered by a dome collapse on 21 September 1997 (Druitt *et al.* 2002).

<sup>2</sup>Major dome-collapse events on 4 and 6 November 1997 removed about 10% of the lava dome leaving a broad crater. A lobe started to extrude at 4–5 m<sup>3</sup> s<sup>-1</sup> in the crater (Watts *et al.* 2002) on 4 November 1997 and collapsed in its entirety on 26 December 1997 leaving older sectors of the dome intact (Sparks *et al.* 2002). Ash is derived from the lobe and old, hydrothermally altered, crater wall material.

<sup>3</sup>Ash derived from venting through dome and rockfalls on a new lobe that started to extrude at 4–8 m<sup>3</sup> s<sup>-1</sup> on 27 December 1997 (Watts *et al.* 2002). Some venting and rockfalls may have occurred on the much older and more stable portion of the dome erupted pre-November 1997 and the crater walls.

<sup>4</sup>Dome growth continued until 10 March 1998, when it ceased until November 1999. Throughout this pause in extrusion there were dome collapses, explosions and ash venting (Norton *et al.* 2002). The east and NE parts of the dome were extruded between January and November 1997, the small portion removed in the dome collapse on 5 June 1999 probably included material from the 17 May 1997 and 22 October 1997 lobes.

<sup>5</sup>A large dome collapse on 20 March 2000 removed 95% of the dome, then lava extrusion began again almost immediately at 3–5 m<sup>3</sup> s<sup>-1</sup> (Cam *et al.* 2004) and persisted almost continuously for 16 months (Matthews *et al.* 2002). The 29 July 2001 collapse removed most of this dome.

<sup>6</sup>A new dome was observed growing on 3 August 2001 immediately after the 29 July 2001 dome collapse (Matthews *et al.* 2002). Small, partial collapses occurred throughout late 2001–2003 before the major 12–13 July 2003 collapse removed most of this dome, leaving a small ridge on the crater floor. The first Vulcanian explosion was on 13 July 2003, so not relevant for this sample (Herd *et al.* 2005).

<sup>7</sup>Ash was derived from the first of a series of five explosions 28 June–27 July 2005 in the crater left by the 2003 total dome collapse (Ryan *et al.* 2007). This first explosion generated ash comprising old, altered andesite with no juvenile glass.

<sup>8</sup>Slow dome growth was first observed on 8 August 2005, with pulsatory dome growth continuing at low average rates (<4 m<sup>3</sup> s<sup>-1</sup>) through January 2006 (Ryan *et al.* 2010).

<sup>9</sup>Vigorous ash venting, associated with the onset of a period of rapid extrusion rates (>12 m<sup>3</sup> s<sup>-1</sup>, with a peak rate of >20 m<sup>3</sup> s<sup>-1</sup> on 10–12 February 2006) (Ryan *et al.* 2010), which started on 9 February 2006 alongside the existing dome emplaced over the previous 6 months (Loughlin *et al.* 2010).

<sup>10</sup>Ash venting during a period dominated by very high average extrusion rates (>12 m<sup>3</sup> s<sup>-1</sup>, with a peak rate of >20 m<sup>3</sup> s<sup>-1</sup> on 10–12 February 2006) (Ryan *et al.* 2010).

<sup>11</sup>Rockfalls and ash venting from an active lobe at the summit of a 9 month-old dome (Loughlin *et al.* 2010).

<sup>12</sup>The 20 May 2006 dome collapse removed the entire 10 month-old lava dome and the small ridge of older, altered material (Loughlin *et al.* 2010). This basal ash sample was from the earliest ash venting and PDCs of the collapse.

<sup>13</sup>A Vulcanian explosion on 21 May 2006 (Loughlin *et al.* 2010) covered all of Montserrat with grey accretionary lapilli.

<sup>14</sup>Dome growth continued following the 20 May 2006 collapse. The collapse of 30 June 2006 was composed of this new dome material (average extrusion rate 6.9 m<sup>3</sup> s<sup>-1</sup> on 25 May – 27 June 2006) (Ryan *et al.* 2010).

<sup>15</sup>Most of the material erupted in 2009–2010 was being shed from the active lava dome. Any older material incorporated into the 11 February 2010 collapse could derive from dome growth Phase 4b between 3 December 2008 and 3 January 2009. Older dome rocks (generated after the 20 May 2006 full collapse) remained undisturbed.

assemblage is very sensitive to small variations in the duration of the heating (Horwell 2002). Recently, this problem has been overcome with the advent of high-resolution diffractometers, aiding peak separation in bulk-ash samples, and the development of a rapid internal attenuation standard (IAS) technique by Le Blond *et al.* (2009), which allows quantification of a single mineral phase in a heterogeneous dust, without prior knowledge of bulk-sample composition. These improvements have facilitated more-rapid and, potentially, more-accurate assessment of cristobalite abundance than previously, and have been tested in ash from Soufrière Hills and other volcanoes (e.g. Merapi, Chaitén, Vesuvius, Rabaul, Eyjafjallajökull, Grímsvötn and Sakurajima) (Horwell *et al.* 2010a, b, 2013a; Le Blond *et al.* 2010; Hillman *et al.* 2012; Damby *et al.* 2013). This study systematically quantifies cristobalite abundance in a suite of 24 Soufrière Hills ash samples (1997–2010), using the IAS technique of Le Blond *et al.* (2009).

Horwell *et al.* (2013b) studied the nature and formation of cristobalite in the Soufrière Hills lava domes, and considered the likely processes of silica transport. Their observations support the mechanisms for cristobalite crystallization at Soufrière Hills proposed by MacGregor (1938) and Baxter *et al.* (1999): cristobalite is formed through vapour-phase crystallization in permeable networks of pores, cracks and vugs, and through devitrification of volcanic glass. Horwell *et al.* (2013b) showed that the morphology of vapour-phase cristobalite is variable: crystals are predominantly prismatic but platy, hexagonal crystals are also found in some samples. Devitrification cristobalite appears as ‘feathery’ crystallites in the groundmass, with more extensive devitrification leading to ‘fish-scale’ patches in thin section (characteristic of cristobalite). Where devitrification is complete, Horwell *et al.* (2013b) also found that subhedral quartz may be present instead of cristobalite; they proposed that the quartz forms by phase transition from cristobalite, enabled by heat from circulating fluids. Whilst the kinetics of formation of cristobalite through vapour-phase crystallization and devitrification are not well constrained, Williamson *et al.* (2010) demonstrated that cristobalite can form in the Soufrière Hills dome within hours to days of magma injection into the upper conduit or dome. Horwell *et al.* (2013b) found up to 11 wt% cristobalite in the Soufrière Hills dome rock.

## Methods

The ash samples, mainly sourced from the Montserrat Volcano Observatory (MVO) archives, are primarily the products of full or partial collapse of lava domes (14 samples) and are co-PDC ashfall; that is, from the plume that lofts from a PDC resulting from dome collapse. Ten further samples are investigated: three samples were produced by Vulcanian explosions (September–October 1997, 21 May 2006 and 10 January 2010); one sample was produced by a phreatic explosion (June 2005); six samples were produced by ash-venting events, four of which were collected over several weeks/months of activity (February 1998, August–October 2005 and January–February 2006); and two of which were collected immediately after single events (5 October 2009 and 4 January 2011). Three of the ash-venting samples may also contain some dome material from concurrent rockfalls that generated small PDCs (MVO1343 and MVO1348 from January–February 2006, and sample 04.01.10 from January 2010) (Table 21.1).

For each sample, we have obtained information on the location of collection and the distance from vent, date of eruption, date of collection and the phase of dome growth (according to Wadge *et al.* 2014). We then collated information supplied by MVO and from the MVO weekly reports ([www.mvo.ms](http://www.mvo.ms)) on dome growth and collapse in order to assign a maximum ‘DRT’ to each sample. In several instances, it was not possible to determine the

exact portion of dome that was removed during a collapse, so the data given are the maximum likely DRT. For some samples we recognize that small amounts of older dome lobes may have been incorporated, adding an additional component to the ashfall (see Table 21.1 for details). We also collated data on lava extrusion rates from published sources (Watts *et al.* 2002; Cole *et al.* 2010; Loughlin *et al.* 2010; Ryan *et al.* 2010).

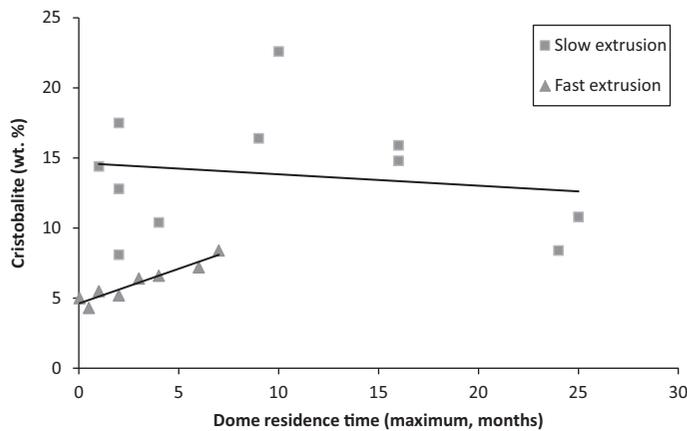
Cristobalite and quartz abundances were quantified for all bulk-ash samples using an Enraf-Nonius X-ray diffractometer with an INEL static, curved position-sensitive detector (PSD) at the Natural History Museum (NHM), London, according to the IAS method of Le Blond *et al.* (2009) with ZnO as the internal standard. Samples were first sieved to <1 mm so that data would be comparable with previous studies using the method. Cristobalite abundances were determined against a synthetic sample of alpha-cristobalite provided by D. K. Smith and kept at the NHM. We have determined this sample to be of a high degree of crystallinity and purity, probably higher than that of volcanic cristobalite, which has up to 3 wt% Al<sub>2</sub>O<sub>3</sub> substituted into its structure (Horwell *et al.* 2012). Consequently, the absolute abundances determined for the current samples may slightly underestimate the true value. This does not affect comparisons amongst samples in this study or the relative trends in abundance, and the data have an error of  $\pm 3$  wt%. The experiments were carried out without sample information to ensure no user bias in the quantification process.

## Results

Cristobalite abundance in the Soufrière Hills dome-collapse ash samples ranges from 4.3–22.6 wt% of the bulk ash (Table 21.1). Within this range, ash from slowly extruded lava ( $\lesssim 5 \text{ m}^3 \text{ s}^{-1}$ ) contained 8.4–22.6 wt% cristobalite, whereas ash from rapidly extruded lava ( $\gtrsim 5 \text{ m}^3 \text{ s}^{-1}$ ) contained 4.3–6.6 wt% cristobalite. The rapidly extruded samples were generated in 2009 and 2010 during a period of elevated extrusion rates (average of  $7 \text{ m}^3 \text{ s}^{-1}$  but a maximum of  $35 \text{ m}^3 \text{ s}^{-1}$ : Cole *et al.* 2010), and the cristobalite content is markedly lower than throughout most of the previous 13 years of eruption, confirming the results of the routine analyses.

There is no correlation between cristobalite abundance and DRT (Pearson’s product–moment correlation coefficient =  $-0.169$ ; two-tailed  $p$ -value = 0.619) for samples from collapses of dome material extruded during periods with normal, slow extrusion rates ( $\lesssim 5 \text{ m}^3 \text{ s}^{-1}$ ) (Fig. 21.1, Table 21.1). By contrast, there is a strong, positive correlation between cristobalite abundance and DRT (Pearson’s product–moment correlation coefficient = 0.954; two-tailed  $p$ -value =  $2.28 \times 10^{-4}$ ) (Fig. 21.1) for samples generated during periods of rapid extrusion ( $\gtrsim 5 \text{ m}^3 \text{ s}^{-1}$ ). Therefore, longer DRT does not necessarily result in dome-collapse ash samples with more abundant cristobalite. For example, greater abundances of cristobalite are found in collapses mobilizing dome rock with a DRT of 8–9 months (16.4 and 22.6 wt%) than eruptions mobilizing dome material with a DRT of 16–24 months (8.4–15.9 wt%). Likewise, collapses mobilizing fresh dome material (e.g. DRT <2 months) are capable of generating cristobalite-rich co-PDC ash when lava is slowly extruded (e.g. 14.4 and 12.8 wt%).

Two ash samples collected from the 20 May 2006 lava-dome collapse and a subsequent explosion on 21 May 2006 (MVO1498 and MVO1492, respectively) allow direct comparison of cristobalite abundance from an old dome (DRT of *c.* 10 months) and a Vulcanian explosion (DRT of 0 months). MVO1498 was collected from the base of the ashfall sequence on 20 May and contains 22.6 wt% cristobalite, whereas MVO1492 was collected from the fall deposit of the subsequent explosion and contains 6.8 wt% cristobalite. All of the Vulcanian explosion samples contained approximately 6 wt% cristobalite (5.6, 6.1 and 6.8 wt%).



**Fig. 21.1.** Cristobalite content v. DRT for two data series: (i) dome-collapse ash samples collected during slow extrusion rates; (ii) dome-collapse ash samples collected during fast extrusion rates. Both datasets include ash-venting samples, which incorporated dome material (or concurrent rockfall/PDC material).

However, the 28 June 2005 phreatic explosion is notably different, containing 10.5 wt% cristobalite and 4.7 wt% quartz (the Vulcanian explosions contained <1.5% quartz).

The six ash-venting samples have varied cristobalite abundance (2.3–17.5 wt%). Five samples contained abundant cristobalite (6.4–17.5 wt%), and were all either erupted when a dome was present or their eruption was accompanied by a small dome-collapse PDC. Sample 05.10.09 contained only 2.3 wt% cristobalite and was erupted just before Phase V dome growth began, when the vent was likely to be unobstructed by older dome material.

Quartz abundance throughout the sample suite is lower than cristobalite abundance and varies less, ranging from about 0 to 5 wt%, with most samples having  $\ll 2.5$  wt%.

## Discussion

Our dataset demonstrates considerable variation in cristobalite abundance in ash throughout the Soufrière Hills eruption. The highest value measured was approximately 23 wt% in a sample of bulk co-PDC dome-collapse ash, but all other samples have <18 wt% cristobalite. From analysis of just 14 dome-collapse ash samples, we cannot conclude that 23 wt% represents an upper limit for cristobalite in bulk ash at Soufrière Hills. However, we do note that this figure is in broad agreement with published data: Baxter *et al.* (1999, 2014) found up to 30 wt% cristobalite in the sub-10  $\mu\text{m}$  ‘thoracic’ fraction over 5 years of analyses. The differences between these values probably originates, at least in part, from the known enrichment of cristobalite in the fine fractions relative to bulk co-PDC ash (Horwell *et al.* 2003), but there may also be some disparity in abundance due to the differences among techniques used to quantify it. Nevertheless, these data support the values used by Hincks *et al.* (2006) (5–25 wt% cristobalite for dome-collapse ash) for their risk assessment of long-term cumulative exposure to cristobalite from Soufrière Hills volcanic ash using Monte Carlo simulation.

The ash studied contains little quartz, and, where abundance is greater than around 2.5 wt%, we attribute this to the formation of quartz as a devitrification product (Horwell *et al.* 2013). Murphy *et al.* (2000) found <0.5 wt% quartz in dome lava indicating that, as with cristobalite, quartz may be enriched in dome-collapse ash due to fractionation of minerals between the co-PDC plume and the block-and-ash flow deposit (Horwell *et al.* 2001). Tridymite was not found in Soufrière Hills ash samples in this study, and Horwell *et al.* (2013) did not observe it in Soufrière Hills dome lavas. It is not clear why tridymite is

absent; at Mount St. Helens, it is seen as acicular crystals within the dome groundmass (e.g. Cashman *et al.* 2008).

We find cristobalite in all three Vulcanian explosion samples investigated (5.6–6.8 wt%). Since cristobalite is not a primary magmatic phase at Soufrière Hills, its presence in these samples indicates that it must have been incorporated into the eruption column by erosion of existing dome material or from entrainment of upper-conduit plug material or hydrothermally altered edifice, all of which are suitable environments for cristobalite formation. In the single phreatic explosion sample, incorporation of hydrothermally altered edifice material is a reasonable explanation for the presence of both cristobalite (10.5 wt%) and quartz (4.7 wt%), since the quartz is likely to have formed through a reconstructive phase transition from cristobalite, aided by hydrothermal fluids (Horwell *et al.* 2013).

The data allow us to investigate the causes of variation in cristobalite abundance. Below, we discuss seven potential controls.

### Controls on cristobalite variation

**Dome rock mineralization.** The primary control on the variation in cristobalite (and quartz) abundance in ash is presumed to be the variation in its abundance in the dome rock from which the ash forms. Horwell *et al.* (2013) found that cristobalite abundance in Soufrière Hills dome lava varies widely (in the range 1–11 wt%; nine samples). The lowest abundance was found in vesicular dome lava samples, which were interpreted to have been injected into the base of the dome shortly before collapse (Williamson *et al.* 2010); the highest abundance was found in samples that contained abundant vapour-phase cristobalite. We speculate that ash containing the highest cristobalite abundance derives from portions of the dome where extensive vapour-phase crystallization has taken place. Horwell *et al.* (2013) proposed that extensive vapour-phase crystallization of cristobalite can occlude pore spaces, leading to progressive reorganization of the degassing pathways. Through this process, different parts of the dome may be differentially mineralized. Parts of the dome that are not exposed to silica-bearing gases will be cristobalite-poor; other parts may experience total infill of porosity by vapour-phase mineralization. This latter scenario suggests that there may be a maximum fraction of vapour-phase cristobalite that a portion of dome rock can acquire before the supply of silica-bearing gases becomes self-limiting. Other processes, such as compaction in the lower dome–upper conduit (Michaut *et al.* 2009), may also affect the routing of gases through the dome and the subsequent distribution of vapour-phase mineralization.

**Lava extrusion rate.** We propose that ash from slowly extruded dome lava is richer in cristobalite (8–23 wt%) than that from rapidly extruded dome lava (4–7 wt%) because it is more extensively devitrified. Devitrification depends on diffusion through the glass of species that are ‘crystal nutrients’ (Castro *et al.* 2008); hence, it proceeds more rapidly at higher temperatures. For exogenous domes, such as those at Soufrière Hills, the magma cools rapidly on eruption. Slowly erupted magma spends more time travelling through the upper conduit–dome system, implying that it spends more time at the elevated temperatures and low pressures that favour devitrification. This model is proposed by Scott *et al.* (2013) to explain their observation of variable devitrification in dome lavas at the Santiaguito dome complex (Guatemala). Textural data from Soufrière Hills dome lava presented in Sparks *et al.* (2000) and Murphy *et al.* (2000) offer further support: they found that rapidly extruded dome rock is glass-enriched (25–30 vol.%) and cristobalite-poor in comparison to slowly extruded samples (5–15 vol.% glass). The model predicts that abundance of cristobalite of devitrification origin should correlate inversely with extrusion rate; this hypothesis could be tested against analysis of dome rock samples (see below) since the origin of the

cristobalite (devitrification v. vapour phase) is more-readily determined in dome rock thin sections than in the ash derived from it. An additional explanation for the greater abundance of cristobalite in ash derived from slowly extruded lava is that the lava could be exposed to a higher flux of silica-bearing gases per unit volume of lava than would more-rapidly extruded lava. This assumes that the total flux and spatial concentration of such gases in the dome is independent of extrusion rate. This model predicts that the abundance of vapour-phase cristobalite should correlate inversely with extrusion rate and could be tested as above.

**Dome residence time.** The abundance of cristobalite in bulk ash derived from rapidly extruded lava correlates strongly with DRT (Fig. 21.1). According to the first model proposed above, these samples are cristobalite-poor because they contain little devitrification cristobalite. The variation in total cristobalite could, therefore, reflect variation in vapour-phase cristobalite content. Since we expect vapour-phase mineralization to be progressive, the correlation between cristobalite abundance and DRT for rapidly extruded samples is consistent with this hypothesis. Alternatively, or additionally, it could be that slow, but progressive, devitrification is partly responsible for the correlation with DRT. We know that, during the period of rapid extrusion in 2009–2010, PDCs were largely generated from calving of freshly erupted dome lava rather than from deeper portions of the dome, and Sparks *et al.* (2000) did not observe vapour-phase cristobalite in their young, glassy dome samples. Although devitrification will be more rapid in the hotter conditions deeper in the dome, we still expect that it will occur at shallower levels. Further work on dome rock is required to discriminate conclusively between these two hypotheses. A suitable analytical methodology was developed by Horwell *et al.* (2013), who demonstrated that a combination of Scanning Electron Microscope (SEM)–Raman and SEM–cathodoluminescence can be used to identify submicron crystallites that are characteristic of early-stage devitrification cristobalite. Image analysis can then be used to quantify devitrification v. vapour-phase cristobalite in samples of known extrusion rate and DRT.

The abundance of cristobalite in bulk ash derived from slowly extruded lava does not correlate with DRT (Fig. 21.1). According to the models proposed above, this could be because cristobalite abundance for these samples is dominated by their degree of devitrification, which correlates inversely with extrusion rate rather than DRT. Alternatively (or additionally), the nature of the dome-collapse event that originates the ash may be influential. Unlike the immediate calving of fresh dome rock during rapid extrusion, larger collapses following periods of variable extrusion rate will mobilize more heterogeneous source material; this may eliminate any correlation between cristobalite abundance and maximum DRT for these samples.

**Type of eruptive event.** Our data also indicate that the cristobalite abundance in ash is related to the type of event that generates the ash. Samples that were formed by Vulcanian explosions, by ash venting in dome-free parts of the crater and by collapse of lobes of the dome during periods of high extrusion-rate all contain <7 wt% cristobalite; we designate these samples, and the events that produce them, ‘cristobalite-poor’. We speculate that ash derived from passively generated rockfalls and PDCs from dome talus or the carapace might also fall into this category (see the discussion in the subsection ‘Carapace conditions’ later). Samples that were formed by collapse of dome material during periods of low extrusion rate, as is the norm at Soufrière Hills (Watts *et al.* 2002; Loughlin *et al.* 2010; Ryan *et al.* 2010), all contain >7 wt% cristobalite. Ash formed during ash venting through a dome, and phreatic explosions – that probably incorporate hydrothermally altered edifice – also fall into this category, which we designate ‘cristobalite-rich’. This classification system, summarized in Table 21.2, provides a guide for assessing the

**Table 21.2.** Eruption styles at SHV, classified by approximate cristobalite content

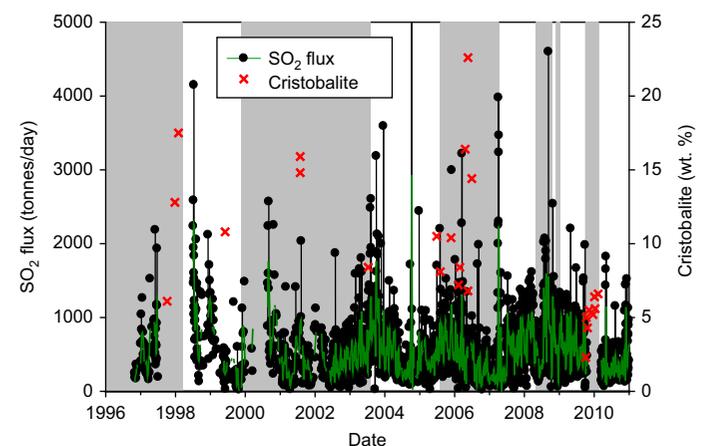
Cristobalite-rich (<7 wt%)	Cristobalite-poor (>7 wt%)
Dome collapse during periods of high extrusion rate	Dome collapse during periods of normal extrusion rate
Ash venting (without dome incorporation)	Ash venting (with dome incorporation/associated PDCs)
Vulcanian explosions (with or without dome incorporation)	Phreatic explosions (inclusion of edifice)

likely cristobalite abundance in ash from future eruptions of Soufrière Hills.

**Source variations in composition and flux of silica-bearing gases.** The kinetics of cristobalite crystallization may be affected by fluctuations in the composition and flux of silica-bearing gases circulating in the dome. Such fluctuations may result from, for example, pauses in magma extrusion, and in response to changes in volatile exsolution from magma at depth. It is known that some major dome collapses are immediately followed by a marked increase in SO<sub>2</sub> content (Christopher *et al.* 2010), suggesting that magmatic gas is trapped beneath the dome prior to collapse, then subsequently released. This may result from permeability occlusion at depth, which may, in turn, lead to pressurization beneath the dome, contributing to its failure (Horwell *et al.* 2013). However, we find no systematic relationship between SO<sub>2</sub> flux following a collapse and cristobalite abundance in dome-collapse material, but note that a weak correlation might not be apparent with so few samples (Fig. 21.2).

**Carapace conditions.** The rubbly nature of the talus/carapace in the top 5–10 m of the dome could be unfavourable for cristobalite crystallization through vapour-phase transport in pore networks as transport is likely to be predominantly around the talus blocks with little internal pore flow within individual blocks. In addition, penetration of meteoric water into the carapace and talus could also block the movement of vapours through the pore network, again leading to unfavourable crystallization conditions for cristobalite.

**Fractionation of the ash.** The cristobalite content of the ash itself will be affected by: the variable fractionation of minerals into the lofting co-PDC plume, depending on fragmentation efficiency, and the resulting particle size and density (Horwell *et al.* 2001); plume dynamics that will affect whether fragmented dome portions are widely dispersed or deposited at specific localities; the



**Fig. 21.2.** Cristobalite content of dome-collapse samples v. SO<sub>2</sub> flux with periods of continuous dome growth shown in grey bands.

distance from the vent, although, on Montserrat, ash collection on land is not feasible more than about 8 km from the vent and there is no evidence to suggest that shorter distances significantly influence the results (Table 21.1); and entrainment of older pyroclastic material during PDC emplacement.

## Conclusion

Whilst cristobalite formation in lava domes is clearly spatially and temporally heterogeneous, cristobalite abundance in ash can be linked systematically with eruption style, allowing inferences about its formation mechanism. The results of this study show that cristobalite abundance in Soufrière Hills volcanic ash, erupted between 1997 and 2010, does not correlate with lava DRT during periods of slow lava extrusion, but does correlate strongly during rapid extrusion phases. Abundant cristobalite can form rapidly during periods of slow extrusion rate (as seen for samples with low DRT), but, during high extrusion rates, cristobalite abundance is substantially lower, at a level similar to that observed during Vulcanian explosions. The patterns in cristobalite abundance for different eruption styles that we observe here support those seen in previous studies at SHV (Baxter *et al.* 1999; Horwell *et al.* 2003). These relationships should be considered during future activity, and may help to inform any new health-risk assessment undertaken at Soufrière Hills as part of the strategy of ash sample collection and analysis of cristobalite (see Baxter *et al.* 2014). Through collaboration between risk managers and the volcano observatories, the eruption style (e.g. dome collapse, Vulcanian explosion or ash venting) can be rapidly established, as well as information on whether collapsing dome portions are fresh, aged or heterogeneous, and whether lava extrusion rate is low or high. The data also clarify that ash venting and explosion-generated ash can contain cristobalite if lava dome material is mobilized.

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