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The Fate of Venusian Chlorine

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ABSTRACT

Venus and Earth are alike in size and composition and share similarly productive histories of volatile degassing. Yet while chlorine (Cl) is concentrated in Earth's surface reservoirs, it is nearly absent from the atmosphere of Venus. The fate of Venusian Cl offers insights into the evolution of magmatism and history of volcanic degassing on Venus and similar telluric planets. Here we explore the potential for sodalite ($Na_8Al_6Si_6O_{24}Cl_2$), crystallised from erupted volcanics, to act as a long-term Cl-sink in planetary crust. By estimating alkaline melt production on Venus and the partitioning of Cl between sodalite and Venusian silicate melts, we constrain Cl sequestration into sodalite-stable alkaline volcanics. We find that phonolites containing up to 7 mol% sodalite can be produced through 91–98% fractionation of low-degree partial melts originating from the Venusian upper mantle. Overall, just ~750 m of alkaline volcanic resurfacing has the capacity to sequester an Earth-like Cl budget within sodalite-bearing lithologies.

1. INTRODUCTION

Chlorine (Cl) is a hydrophilic and volatile element that behaves incompatibly during mantle melting, and, as a result, has been efficiently fluxed from Earth's interior over the course of planetary differentiation[1]. Today over half of Earth's Cl budget can be found in surface reservoirs, with ~25% held in the oceans alone[1,2]. By contrast, very little Cl has been detected in the surface environment of Venus, with radar imagery offering no signs of evaporite deposits[3] and HCl restricted to ~0.4–0.5 ppm[4,5] in the lower atmosphere. The thick, volatile-rich atmosphere is a sign Venus experienced extensive degassing in its past, yet if a terrestrial-like Cl budget was degassed to a similar extent as on Earth, its atmosphere should instead contain ~9 wt% Cl—or an amount equivalent to a global NaCl layer 80 m-thick.

Venus is compositionally and physically comparable to Earth, with a mass of ~0.82 Earth masses (M_e), yet its geodynamic regime and atmosphere are manifestly different[6]. Venus lacks plate tectonics[6], and its atmosphere is dense (92 bar at the surface), hot (with a mean surface temperature of 740 K), and, in comparison to terrestrial volcanic emissions, oxidized[7]. Driving much of Venus's divergent geologic behaviour is its loss of water, which is now almost entirely absent from the present-day Venusian atmosphere (~30 ppm)[8], and likely its interior[9]. Dynamical simulations of the early Solar System indicate that icy impactors from beyond the snow line could have delivered water to the terrestrial planets during later stages of planetary accretion[10,11], and an atmospheric D/H ratio 150 times greater than Earth's is a sign Venus was once water-rich[12]. Whether ancient Venus ever possessed liquid surface water is an open question. Models of the planet's long-term climatic evolution describe an early Venus that spans from dry across its entire crustal history, having lost its water inventory during a prolonged magma ocean phase, to a temperate and oceanic[9,13] past surface environment. Complicating matters, any morphological evidence of an ancient hydrosphere has been overplated by more recent volcanism, which covers ~80% of the exposed surface[7].

Given our limited knowledge of Venus's geological evolution, the present-day atmospheric composition and the near-complete absence of observable Cl deposits in its surface environment offer insights into both the evolution of the Venusian surface as well as its recent geological past[14–16]. Three end-member scenarios may explain the scarcity of atmospheric Cl on Venus: (1) Venus never accreted Cl in amounts comparable to Earth; (2) Cl degassing was inefficient, leaving a significant fraction in the mantle; or (3) Cl was initially degassed from silicate Venus, but has since been assimilated into erupted melts and sequestered by the crystallization of Cl-rich minerals (Fig. 1A).

The first scenario, that Venus never accreted a terrestrial-like Cl budget, lacks support from atmospheric and geochemical data. Accretionary simulations suggest that the volatile inventories of the terrestrial planets are broadly comparable[17–19], whilst *in-situ* measurements of K/Th and K/U ratios in Venusian basalts demonstrate that these ratios are similar in Venus and Earth, implying that the planets have similar volatile budgets[12]. The presence of a dense, CO_2- and N_2-rich Venusian atmosphere is also incompatible with a significant deficit in primordial volatiles[13,20–22].

In the second scenario, Venus's Cl budget has remained trapped in the mantle. However, in comparison with that of Earth, the thick, 92-bar Venusian atmosphere is enriched in atmophile and incompatible elements[12]. For instance, its CO_2 content is comparable to the carbon inventory of Earth's crust, and its N_2 inventory is roughly three times greater than that of Earth's atmosphere—both signs of efficient mantle outgassing[23,24]. Noble gas abundances in Venus's atmosphere further support arguments for extensive volatile release, with Ar-36, Ne-20, and elemental Kr measured in concentrations several times greater than in the atmosphere of Earth[14]. Venus's surface has also clearly experienced intense periods of global resurfacing, and so the extraction of incompatible elements from mantle feeder zones was

likely an efficient process in Venus's past. Collectively, these observations suggest that the Venusian mantle has undergone significant volatile depletion, making large-scale Cl retention in the interior unlikely.

In the third scenario, a significant fraction of Venus's Cl budget was previously outgassed, but was subsequently re-dissolved into erupted volcanics and sequestered within magmatic Cl-bearing minerals. As a volatile, incompatible element with high solubility in silicate melts[25] and yet with few mantle mineralogical hosts, the flux of Cl from the mantle is expected to be efficient[1]. In the absence of substantial mantle water—for instance, if Venus accreted under dry conditions—outgassed Cl would likely accumulate in the atmosphere or precipitate out in salt deposits such as those proposed on Io[26]. Much like Earth, Cl on a water-rich and temperate early Venus would have concentrated in surface water bodies, and with the onset of a runaway greenhouse effect and ocean evaporation, would have then deposited in widespread evaporite layers[27]. Subsequent volcanic over-plating of these salt pans would assimilate NaCl into the silicate melt and crystallise Cl-bearing minerals, effectively sequestering Cl in the crust (Fig. 1B). Atmospheric HCl concentrations, measured *in-situ*, support this hypothesis. Barring trace ClO_x and iron chloride aerosols (2.6 ± 0.5 ppb above 85 ± 2 km[28] and 17 ppbv in the clouds[29], respectively), HCl is the dominant Cl species in the Venusian atmosphere and is similarly distributed above and below the cloud deck (0.6 ± 0.12 ppm at 65 km[4], 0.5 ± 0.15 ppm at 15–30 km[30]), a sign it may be buffered by gas-mineral equilibrium reactions[31].

Among the potential Cl-bearing phases considered in Venus-relevant gas–mineral equilibrium studies, sodalite ($Na_8Al_6Si_6O_{24}Cl_2$), which forms in alkaline, SiO_2-undersaturated igneous systems[32], is considered the most promising stable Cl host[31,33–36]. The high surface temperatures and pressures on Venus enhance atmosphere-surface interactions, further facilitating Cl exchange between volcanic gases, melts, and the crust[37].

The potential for crustal sodalite to host significant fractions of Venusian Cl relies on two key factors: 1) the generation and emplacement of sufficient volumes of alkaline magma at or near the Venusian surface, and 2) the efficient uptake of Cl in sodalite-producing alkaline volcanics. To test whether Venus has in-gassed its surface inventory of Cl via assimilation by alkaline magmatism, we estimated the compositions of erupted volcanics by modelling isentropic (adiabatic) melting of the Venusian mantle, and the subsequent fractionation of these melts to 1 bar. Using modelled Venusian volcanic melt compositions, we experimentally determined the partitioning of Cl between sodalite and alkaline magma to constrain Cl compatibility in sodalite. We find that, as a consequence of Venus' widespread volcanism, atmospheric and surface Cl deposits could have been efficiently sequestered back into the Venusian crust via incorporation into Cl-bearing mineral phases.

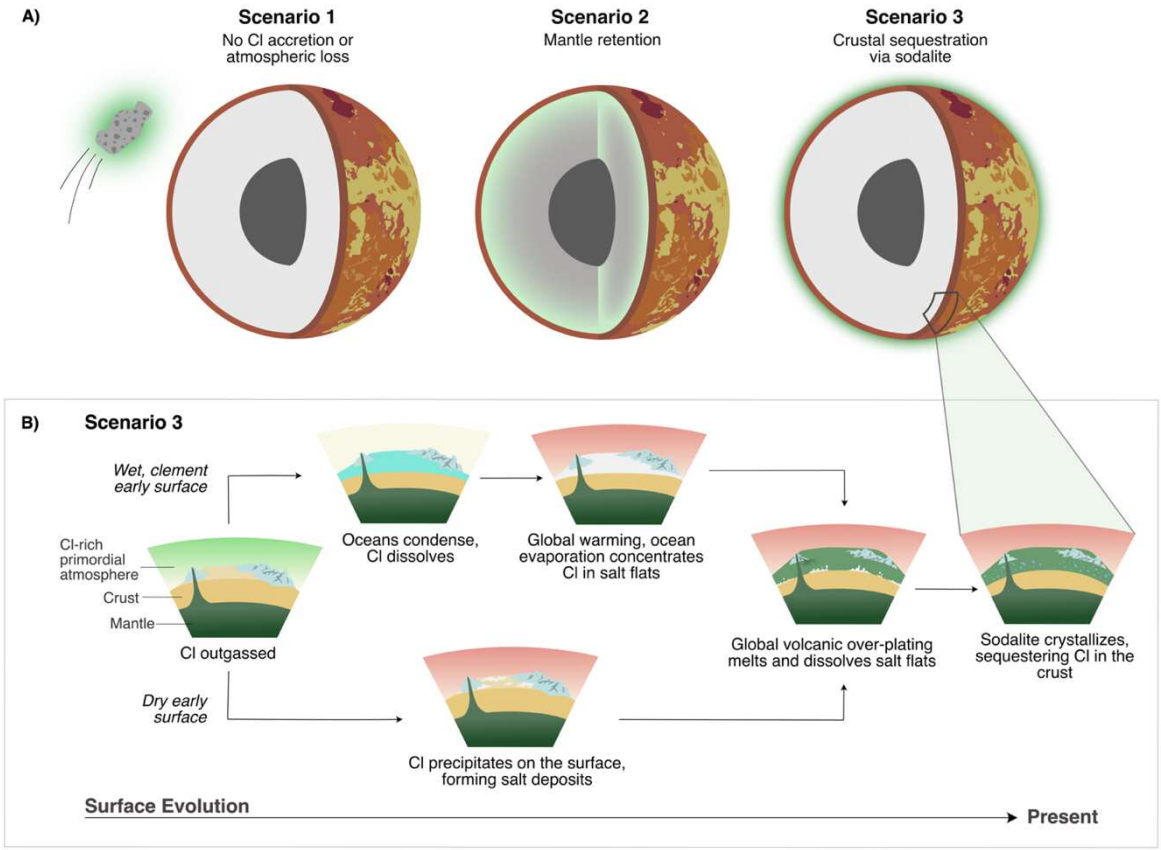


Figure 1. The possible fates of Cl on Venus. The Venusian surface environment appears to host very little Cl. Three plausible explanations may explain Venus's 'missing' Cl: accretion of Cl may have been insignificant on Venus (Scenario 1), Cl may be trapped in the Venusian mantle (Scenario 2), or Cl was outgassed in Venus's early history, but has since been bound in crustal minerals (Scenario 3). If previously outgassed, Cl may have precipitated on the planetary surface, either directly (in the case of an initially dry Venusian surface), or as evaporites deposited from primordial oceans. Subsequent volcanic overplating dissolved Venus's vast salt deposits, crystallizing Cl-bearing minerals such as sodalite.

2. RESULTS

2.1 Generation of alkaline magmas by mantle melting

If sodalite is the host of a significant proportion of Venus's Cl budget, SiO_2-undersaturated alkaline magmatism must be commonplace. Assuming the bulk composition of the mantle is comparable to that of Earth[21], we find that partial melts with >3 wt% Na_2O can be produced through low-degree (<7 wt%) partial melting in the upper mantle. Supplementary Fig. 1 depicts the stable mineral assemblages under upper-mantle to surface conditions and is contoured to show the alkalinity of melt (end-member fraction of the sodic melt component $NaSi_{1/2}$) within the suprasolidus fields. At a terrestrial-like upper-mantle temperature of 1250 °C, isentropic melting is initiated at 12.5 ± 1.3 kbar, corresponding to depths of roughly 30–40 km. Estimates of Venus's average crustal thickness range from 8 to 35 km[38–40] and may reach up to 70 km in some tesserae highlands[39], placing the locus of decompression melting near the base of the crust and coincident with conditions of rift-induced melting.

Upon cooling and decompression along the Venusian adiabat, these low-degree basaltic melts should undergo further fractionation to form highly alkaline volcanic rocks. The fractionation of these partial melts exhibits a notable sensitivity to pressure[41], with alkaline and SiO_2-undersaturated melts favoured when crystallization begins at pressures >3-5 kbar, where clinopyroxene is the first major crystallizing phase (rather than olivine, which is first to crystallise at pressures <3-5 kbar)[42]. By decompressing and cooling the partial melt composition generated by isentropic melting, starting at pressures ranging from 8.8–5.5 kbar and down to the 1 bar, melts undergo 91-98% fractionation to generate phonolites and trachytes with alkali contents of 10-15 wt% (Supplementary Fig. 2). These residual alkaline melts must then be extracted from crystal mushes and concentrated in some manner prior to eruption, as

commonly occurs in phonolite-producing magmatic systems on Earth[43]—possibly through compaction, filter-pressing[44], or in response to seismic disturbances[45]. Fractionating magma chambers and large-scale crystal mush systems in the Venusian crust have previously been proposed as sources of large volumes of evolved melts[46], and are consistent with arguments for plutonic heat-loss from the planetary interior[47].

2.2 Experimental calibration of Cl mineral-melt partitioning behaviour

To experimentally test the potential role of sodalite as a major mineral host of Cl in Venusian alkaline volcanics, we conducted mineral-melt Cl partitioning experiments, crystallizing sodalite from a phonolitic melt of the type which may have formed on Venus. The starting material composition had alkali and silica contents comparable to those produced through fractionation of our simulated Venusian mantle (Supplementary Table 1), and were saturated with 1 wt% Cl, added as NaCl. Sodalite-melt distribution constants ($D_{Cl}^{sodalite/melt}$) were calculated from the measured Cl contents of residual glasses (Extended Data Table 1). For a system in equilibrium, $D_{Cl}^{sodalite/melt}$ reflects the relative concentration of Cl in sodalite versus the residual melt, and the compatibility of Cl with sodalite.

The results of our mineral-melt partitioning experiments give Cl concentrations of 0.65 ± 0.12 wt% in our selected phonolite glass, which correspond to a $D_{Cl}^{sodalite/phonolite}$ value of 11.33 ± 2.19 and indicate that in an anhydrous alkaline silicate melt, Cl is highly compatible in sodalite (that is, $D_{Cl}^{sodalite/melt} \gg 1$) (Extended Data Table 1). The mineral assemblages of experimental charges include plagioclase, sodic pyroxene, iron and titanium spinels, trace chlorapatite, and ~3-5 μm euhedral crystals of sodalite. Excess undissolved Cl forms droplets of immiscible sodium and potassium chlorides. To verify equilibrium, a time-series of experiments was conducted with phonolite-halite mixtures held at final temperatures for 1 to 8

hours. Glass compositions from 4 hours onward remained consistent within the experimental uncertainty (Extended Data Fig. 1).

2.3 Constraints on Venusian Cl sequestration via alkaline overplating

While Cl is highly soluble in silicate melts and will readily dissolve in overplating alkaline volcanics, the long-term capacity of the Venusian crust to host Cl relies on the efficient production of Cl-bearing minerals. Sodalite formation can be expressed by the reaction[48]

$$6NaAlSiO_4 + 2NaCl = Na_8[AlSiO_4]_6Cl_2$$

Nepheline Sodalite

An estimate of sodalite production in a NaCl-saturated magmatic system can then be calculated from the proportion of nepheline in the Cl-free phase assemblage, with sodalite and nepheline forming in a 1:6 molar ratio. Calculating the CIPW normative mineral assemblage of our experimental phonolite composition, we find nepheline occurs in normative proportions of 31 mol% (Supplementary Table 1). With the knowledge that sodalite crystallisation is favoured over nepheline in Cl-rich silicate melts[48], we find that phonolite can yield a sodalite molar abundance of 7 mol%. This value places a lower limit on the proportion of sodalite produced in such a volcanic melt, and can be used to calculate the maximum volume of alkaline volcanic overplating needed to fully sequester an Earth-like Cl budget in Venus's crustal sodalite. In reality, alkaline volcanic melts overplating salt flats on the Venusian surface are likely to produce more sodalite than our estimates suggest, as the CIPW norm treats the crystallisation process as a closed system. This does not account for the saturation of early volcanics in Na from halite evaporites, which should stimulate additional nepheline crystallisation.

Mass-balancing Earth's estimated bulk Cl inventory (~1.04×10^{23} g, or ~2.93×10^{21} mol)[2,49] for Venus and assuming a similar degree of planetary Cl degassing (~55% of total Cl),

we find a presumed Cl load in Venus's surface environment of 1.38×10^{21} mol can be sequestered in 0.75 km of phonolitic overplating. Alkaline volcanism therefore presents an efficient mechanism for generating large sodalite-bearing Cl sinks within sufficiently alkaline deposits on Venus, requiring relatively modest volumes of alkaline melt production ($\sim 0.35 \times 10^9$ km^3).

3. DISCUSSION

3.1 Alkaline volcanism on Venus

On Earth, the global rates of alkaline magma production are poorly constrained, and likely account for less than 1% of the total igneous output[50,51]. Nevertheless, there is evidence to suggest that Venus may be more conducive to alkaline magmatism than Earth, potentially producing such melts across a broader spectrum of tectonic settings[46,52,53]. The estimated average thickness of Venus's volcanic plains is 8-35 km[38–40], such that the ~0.75 km upper limit of volcanic overplating required to sequester an Earth-like Cl budget is well within the planet's estimated magmatic output. The proportion of volcanic deposits on Venus that are alkaline and capable of stabilising sodalite, however, remains uncertain.

The tectonic regime of a planet exerts significant controls on the nature and distribution of global magmatism. The crustal dynamics of Venus and Earth are appreciably different, which should in turn impact their capacities for large-scale alkaline melt generation. On Earth, ~75% of new crustal production is associated with mid-ocean ridges, where melts are primarily basaltic and alkali-poor, while <5% is derived from intraplate sources—where the bulk of terrestrial alkaline volcanism occurs[54,55]. The geodynamics of Venus, with its single-plate lithosphere, are best compared to terrestrial intraplate tectonics[7] and so sources of alkaline melts may be more widespread than on Earth.

In terrestrial intraplate settings, alkaline melts are typically associated with hotspot plumes or lithospheric rifting[56–58]. We suggest that alkaline partial melts can be produced on Venus through decompression melting near the base of the crust, a process characteristic of rift environments, and the formation of low-degree alkaline magmas is also achievable along the cooler margins of Venusian mantle plumes[7]. Other proposed sources of alkaline volcanism on Venus—such as plume-induced subduction zones, layered mafic intrusions, or melting initiated by crustal delamination[46,53,59]—may broaden the range of environments where sodalite-bearing volcanics are possible. Alkaline magmatism on Venus is further evidenced by both *in-situ* observations of alkali basalts at the Venera 8 and 13 landing sites, and channel-like flow features (*canali*), that have been interpreted as alkali carbonatites rich in halogens[60]. Moreover, given the solubility of Cl in silicate melts and its effect on lowering melt viscosity[25], the smooth, low topography of the Venusian basaltic plains may also partly be a consequence of Cl in-gassing.

To assess the timescales required to sequester an Earth-like Cl budget in the crust, we apply published estimates of past and present-day volcanic fluxes on Venus to model the rate of Cl uptake in alkaline melts. Present-day flux estimates—ranging from 0.01 to 4 km^3/year[61–63]—are too low to achieve appropriate levels of Cl sequestration within geologically meaningful timescales (Fig. 2). In Venus's past, however, resurfacing rates must have been significantly higher in order to produce the present global coverage of volcanic plains. Taking lower-limit volcanic flux estimates from the tail-end of Venus's most recent period of enhanced volcanic activity (~15 to 44 km^3/year in a pure catastrophic resurfacing scenario[64]), it would take ~100 to 300 million years to generate the necessary volume of phonolitic melt to accommodate the planet's Cl budget. These estimates are consistent with the proposed durations of Venus's magmatic epochs, which may have spanned hundreds of millions to billions of years[61,64].

Finally, given the degrees of partial melting and fractionation involved in generating our modelled alkaline volcanics, we estimate that 0.3–0.8% melting of the whole mantle would be required to produce a 750 m-thick phonolite layer on the surface of Venus, with an overall 7–17% acting as a feeder zone and undergoing partial melting (Supplementary Fig. 3). Such degrees of mantle processing could have been plausible during periods of mantle overturn in Venus's more volcanically-active past. In these events, the delamination of thickened crust[65] or destabilization of basaltic accumulations in the upper mantle[66] may have triggered whole-mantle convection, leading to large-scale volcanism. This process also produces a chemically heterogeneous mantle well-suited to act as a source of exotic melt compositions.

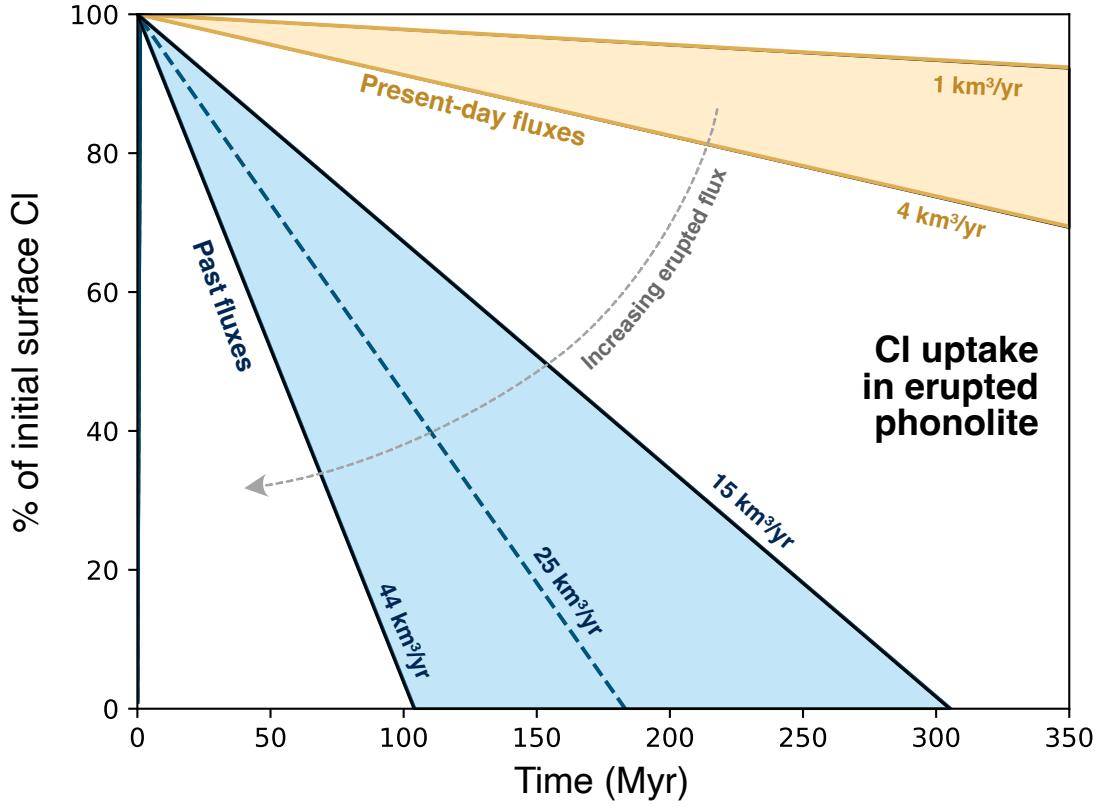
The melt volumes estimated here represent upper limits on the quantity of volcanism required to sequester an Earth-like Cl budget on Venus. In addition to the limitations of the CIPW norm as a calculator of sodalite production (see 2.3), our model assumes that all exsolved NaCl-rich immiscible fluids must be bound in sodalite to efficiently sequester in the crust. We presume that where an emplacement event is too small to entirely assimilate an evaporite deposit, the undissolved molten salts may ascend to the surface of the cooling melt body to be re-dissolved in subsequent overplating events. NaCl migration may not be so efficient, however, and could be trapped in pores or seams in the crystallised volcanics. In such a case, less alkaline volcanism would be needed to capture Cl in Venus's crust. In addition, while sodalite is the primary mineral host for Cl in SiO_2-undersaturated systems[48] and has previously been identified as the most promising mineral buffer of atmospheric Cl on Venus[31], it does not preclude alternate mineral hosts such as chlorapatite and scapolite. However, the presence of chlorapatite is limited by phosphorous availability, and a mass-balanced Earth-like inventory of crustal phosphorous would be able to sequester a maximum of just ~15% of Venus's presumed surface Cl budget. Given the high surface temperatures, it is also doubtful that carbonate minerals such as scapolite are stable on the Venusian surface[67].

As is the case in all studies of Venus's geologic evolution, our interpretations are constrained by limited geochemical and physical data. Upcoming missions such as VERITAS, DAVINCI, and EnVision should advance our understanding of the planet's surface composition, internal structure, and volcanic and tectonic processes[68,69]. These missions may help determine the plausibility of alkaline igneous complexes or extensive lava fields as effective Cl reservoirs in the Venusian crust.

4. CONCLUSIONS

We find that alkaline volcanics can be produced through the fractionation of low-degree partial melts in the Venusian upper mantle, in magmatic regimes thought to operate on Venus. Less than 1 km of phonolitic overplating can entirely subsume an Earth-like surface Cl budget in primary magmatic sodalite.

Despite current data limitations, this study provides experimental support for sodalite stability on Venus[31] and strengthens the case for its role as a mineralogical sink for Cl. The immiscibility and migration behaviour of salt-rich fluids in silicate systems also provides insights into the dynamics of volatile retention and phase separation, and the capacity of sodalite to sequester volatiles may offer constraints on volatile redistribution in stagnant-lid and water-poor worlds, in turn informing broader models of differentiation and crust formation on rocky planets.



283

284 **Figure 2.** The rate of Cl sequestration in Venusian volcanics is dependent on both the
285 composition of the erupted melt and the magmatic flux. Using global magmatic flux estimates
286 of past- and present-day Venus, 15-44 km^3/yr of phonolitic volcanism can feasibly draw down
287 a terrestrial-like surface Cl budget (~1.38×10^{21} mol NaCl) from Venus's surface environment
288 in ~100-300 My—a time interval consistent with projected durations of Venus's resurfacing
289 epochs[61,64].

290

291 ## 5. METHODS

292 ### 5.1 Thermodynamic modelling

293 To model alkaline partial melting and fractionation in Venus's mantle, we used the Mineral
294 Assemblage Gibbs Energy Minimizer (MAGEMin) modelling package[70] in conjunction with
295 the recently published alkaline melt thermodynamic dataset of Weller et al.[71]. This dataset is

296 constructed in the Na_2O-CaO-K_2O-FeO-MgO-Al_2O_3-SiO_2-TiO_2-Fe_2O_3-Cr_2O_3 system, and
297 expands on the peridotite to granite thermodynamic models of Holland et al.[72] by incorporating
298 new activity-composition (*a-X*) relations for alkaline minerals, including nepheline.

299 The composition of the Venusian mantle is considered comparable to that of Earth, with
300 the caveat that it is nearly anhydrous[21]. Here, we used the composition of the spinel lherzolite
301 KLB-1, which is commonly used in similar petrological modelling studies and approximates
302 the terrestrial primitive mantle[73], to act as a proxy for the Venusian mantle. We adopt the
303 approach of Dyck et al. (2021)[74] and Weller et al. (2019)[75] and assume that mantle
304 decompression melting occurs isentropically, that thermal equilibrium is maintained between
305 solid and melt phases, and that melt is efficiently extracted at low melt fractions (3%)—within
306 the range expected for melts fractionating beneath mid-ocean ridges[76].

307 The thermal regime of Venus's mantle is thought to be similar to that of Earth, despite its
308 higher surface temperatures[7,46]. For this reason, we modelled upper mantle melting on Venus
309 at temperatures commensurate with those on Earth, using a mantle potential temperature of
310 ~1250 °C[77]. Following the rationale of Shellnutt (2013)[46], the oxidation state of the Venusian
311 mantle was assumed to be similar to that of Earth.

312 We first simulated adiabatic decompression melting of KLB-1, beginning at 1250 °C and
313 13 kbar, to establish the conditions of incipient mantle melting. This was followed by fractional
314 crystallisation modelling of partial melt compositions generated at varying pressures along the
315 isentropic decompression path (P = 8.8, 7.5, 6.2, 5.5 kbar) down to 1 bar, to simulate magma
316 fractionation through the crust and to the surface and establish the possible range of eruptive
317 melt compositions. The final compositional range of modelled alkaline volcanics informed the
318 selection of appropriate experimental starting compositions. A phonolite within this
319 compositional range and with a high sodium (Na_2O) activity was obtained from tables of

alkaline volcanic compositions listed in Carmichael et al. (1974)[78] for further modelling and experimental work (Supplementary Table 1).

To determine the modal abundance of sodalite in these melts, we calculated CIPW normative phase proportions of our selected phonolite, and obtained a nepheline ($NaAlSiO_4$) modal abundance of its sub-solidus mineral assemblage. In high aCl_2 systems, nepheline reacts with NaCl to form sodalite[48,79], as expressed by Reaction 1. The maximum sodalite content of the selected phonolite assemblage was therefore determined using the stoichiometric ratio 6 nepheline : 1 sodalite, and occurs when nepheline is entirely consumed by Reaction 1.

$$6NaAlSiO_4 + 2NaCl = Na_8[AlSiO_4]_6Cl_2 \quad \text{Reaction 1}$$

Nepheline Sodalite

5.2 Experimental

The selected phonolite starting material was synthesized by intimately mixing reagent-grade oxide (SiO_2, TiO_2, Al_2O_3, FeO, MnO, and MgO), carbonate (Na_2CO_3 and K_2CO_3), and phosphate ($Ca_3(PO_4)_2$) powders for 2.5 hours under ethanol with a mortar and pestle. The mixture was decarbonized overnight in an $800\ °C$ box furnace and reground to a fine powder. The starting material was then re-mixed under ethanol with sodium chloride.

To measure Cl partitioning between sodalite and phonolite in a low-pressure system, sodalite was crystalised *in-situ* from phonolite starting material doped with 1 wt% Cl. Experiments were conducted in sealed vertical tube furnaces using alumina crucibles (8 mm outer diameter, 6 mm inner diameter), which were vacuum-sealed in silica glass enclosures to

minimise Cl loss and prevent contamination. Each enclosure was constructed from a 17 cm-long silica glass tube (11 mm O.D., 9 mm I.D.) sealed at one end and packed with a base of silica powder. The crucible was placed atop this base, followed by a 1 mm-thick alumina lid and a 6 cm-long inner glass rod (8 mm diameter) (Supplementary Fig. 4). The assembled tubes were evacuated under vacuum and sealed by fusing the outer tube to the inner rod using an oxy-acetylene torch.

Vacuum-sealed glass assemblies were suspended in the furnace hotspot using Pt wire attached to a glass hook and rapidly quenched at the end of each run by dropping them into a water bath. Furnace calibration was performed using a B-type thermocouple before and after the experimental campaign. Each experiment was first held at 1200 °C, above the liquidus of the melts, for 1 hour to ensure complete homogenisation. The temperature was then decreased at a controlled rate of 1 °C every 5 minutes to final temperatures of 800 °C. To assess equilibrium in the phonolite–halite system, a time-series of experiments was conducted in which samples were held at a final temperature of 800 °C for durations of 1, 4, and 8 hours. After quenching, experimental charges were extracted from the sealed glass enclosures using a slow-speed diamond saw to preserve crystal textures and minimise mechanical disruption.

5.3 Analytical

Compositional data of experimental run products were collected on the CAMECA SX-Five-Fe field emission electron probe micro-analyser (EPMA) at the University of Oxford Department of Earth Sciences. Alumina crucibles containing sample material were mounted in epoxy, cut in two with a slow-speed diamond saw, and remounted prior to polishing. To limit Cl dissolution during sample preparation, experimental charges were polished to 1 μm grit under isopropanol with an oil-based diamond paste.

Analyses were calibrated with the standards albite (Na, Si, Al), Durango apatite (P), fayalite (Fe), magnesium oxide (Mg), manganese metal (Mn), sodium chloride (Cl), sanidine (K), titanium dioxide (Ti), and wollastonite (Ca). All major elements were analysed with minimum 20 s peak counting times. Sodium proved highly mobile in both glass and sodalite crystals, so a 'time-zero' correction was applied to Na and Si glass analyses to extrapolate measurements to the counts acquired in the first few seconds of each analysis.

Sodalite crystals were numerous, though small in size, with diameters in the range of ~3-5 μm. To limit beam damage whilst reducing the likelihood of including small subsurface sodalite crystals in glass analyses and hence skewing Cl measurements, glass phases were analysed with a 15 kV accelerating voltage and 20 nA beam current, with a defocused spot size of 10 μm for Cl measurements and 15 μm for major element analyses.

Compositional data were collected with CAMECA PeakSight software, and a labradorite and St. John's olivine were analysed as secondary standards. Cl in Durango apatite was also measured as a secondary assurance of Cl calibration. All secondary standard analyses were in excellent agreement with known compositions.

5.4 Calculation of Cl Distribution Constants

The Cl distribution coefficient between sodalite and silicate melt ($D_{Cl}^{sodalite/melt}$) is given by Equation 1:

$$D_{Cl}^{sodalite/melt} = \frac{X_{Cl}^{sodalite}}{X_{Cl}^{melt}} \quad \text{Equation 1}$$

Where X_{Cl}^{α} is the concentration of Cl (wt%) in phase α. Due to the small size of sodalite crystals in experimental charges and concerns over beam damage, direct measurement of $X_{Cl}^{sodalite}$ was not feasible. However, as the experiments were conducted under anhydrous and

sulphur-free conditions, significant substitution of Cl^- by OH^- or SO_4^{2-} within the sodalite structure was considered extremely unlikely. Therefore, $X_{Cl}^{sodalite}$ was assumed to approximate that of stoichiometric sodalite (7.32 wt%[32]). Cl partition coefficients were then calculated using this value alongside measured concentrations of Cl in the melt phase (X_{Cl}^{melt}).

6. ACKNOWLEDGEMENTS

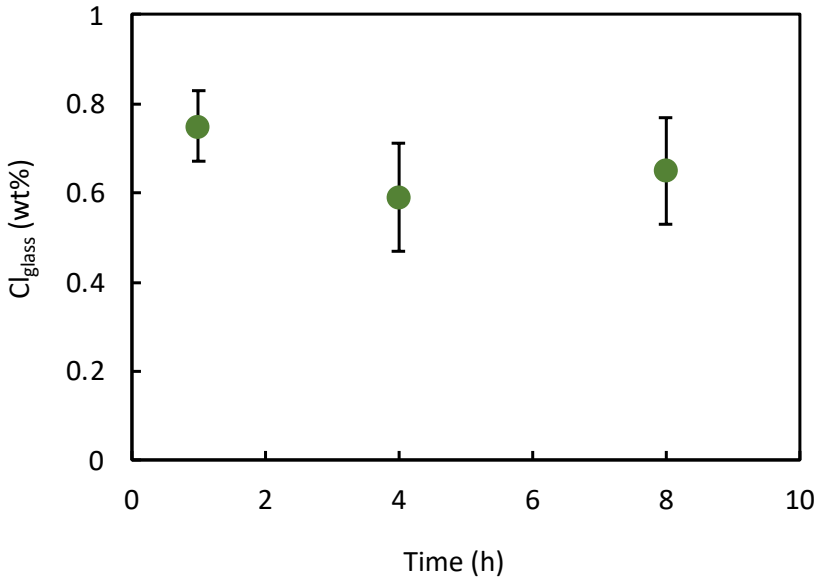
We are grateful to A. K. Matzen for discussions and access to the Oxford University Earth Sciences Department EPMA. K.A.B. was supported by the STFC and the Clarendon Fund Scholarship.

7. EXTENDED DATA

Extended Data Table 1. Experimental conditions and Cl glass concentrations.

Experiment	Added Cl (wt%)	Time at Final temperature (h)	Final temperature (°C)	Measured Cl_{glass} (wt%)	$D_{Cl}^{sodalite/melt}$
KB-37	1	8	800	0.65 (0.12)	11.33 (2.19)
KB-43	1	4	800	0.59 (0.12)	12.51 (2.51)
KB-42	1	1	800	0.75 (0.08)	9.75 (1.10)

2 standard deviations in ().



Extended Data Figure 1. Chlorine concentration (wt%) in phonolitic experimental glasses as a function of time held at a final run temperature of 800°C.

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