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- Gas output from the Central Volcanic Zone of northern Chile
- Identification of a common magmatic end-member of Chilean volcanism
- Comparison between measured and petrologically estimated carbon/sulfur fluxes

Supporting Information:
- Readme
- Table S1
- Table S2
- Table S3

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Gas emissions from five volcanoes in northern Chile and implications for the volatiles budget of the Central Volcanic Zone

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Abstract This study performed the first assessment of the volcanic gas output from the Central Volcanic Zone (CVZ) of northern Chile. We present the fluxes and compositions of volcanic gases (H_2O, CO_2, H_2, HCl, HF, and HBr) from five of the most actively degassing volcanoes in this region—Láscar, Lastarria, Putana, Ollagüe, and San Pedro—obtained during field campaigns in 2012 and 2013. The inferred gas plume compositions for Láscar and Lastarria ($CO_2/S_{tot} = 0.9$–2.2; S_{tot}/HCl = 1.4–3.4) are similar to those obtained in the Southern Volcanic Zone of Chile, suggesting uniform magmatic gas fingerprint throughout the Chilean arc. Combining these compositions with our own UV spectroscopy measurements of the SO_2 output (summing to ~1800 t d^{-1} for the CVZ), we calculate a cumulative CO_2 output of 1743–1988 t d^{-1} and a total volatiles output of >20,200 t d^{-1}.

1. Introduction

The Chilean Andes is a 4500 km long segment of the Andean Cordillera, extending from the Arica and Parinacota regions to Cape Horn (16°S to 56°S), with more than 200 active (Quaternary) volcanoes distributed into four distinct zones [*Stern*, 2004; *Thorpe and Francis*, 1979; *Thorpe*, 1984].

The Chilean segment of the Central Volcanic Zone (CVZ; 17°S to 26°S, Figure 1) comprises a ~600 km long volcanic district and contains ~34 active volcanoes [*Stern*, 2004]. Farther to the south, the Southern Volcanic Zone (SVZ; 34°S to 46°30′S) includes at least 60 historically active volcanoes [*Stern*, 2004] which volcanism is associated with a 70 km thick crust and high degrees of magma differentiation [*Hidreth and Moorbath*, 1988; *Davidson et al.*, 1990].

Despite the presence of several persistently degassing volcanoes, the present-day volcanic gas output of the CVZ remains poorly known. Gas composition data have been reported for several volcanoes [*Tassi et al.*, 2009, 2011; *Capaccioni et al.*, 2011; *Aguilera et al.*, 2012], but fluxes of major volcanic gas species (H_2O and CO_2) have not been reported. Compared to other, better-characterized arc segments (e.g., Central American Volcanic Arc (CAVA) [*Mather et al.*, 2006; *Freundt et al.*, 2014] and SVZ [*Voelker et al.*, 2014]), the paucity of gas flux measurements and poor knowledge of subducted sediment composition [*Plank and Langmuir*, 1998] result into an inadequate understanding of the recycling of volatiles along this sector of the Chilean volcanic front.

Here we report on the first combined assessment of volatiles emissions from five of the most active volcanoes of the CVZ in northern Chile. Using rapid deployment scanning Mini-DOAS (differential optical absorption spectroscopy) instruments [*Galle et al.*, 2010] and a dual UV camera system [*Kantzas et al.,* 2010], we obtained new data of the SO_2 output from the Putana, Láscar, Lastarria, Ollagüe, and San Pedro volcanoes (Figure 1 and Table S1 in the supporting information). These results were integrated with plume compositional data obtained using the INGV-type (Istituto Nazionale di Geofisica e Vulcanologia, Palermo, Italy) Multicomponent Gas Analyzer System (MultiGAS), filter packs, and direct fumarolic gas sampling (Tables S2 and S3 in the supporting information) to indirectly calculate—for the first time—the fluxes of the following other volcanic species: H_2O, CO_2, H_2, HCl, HF, and HBr. Comparison of our results with gas composition data from other Chilean volcanoes (e.g., Villarrica [*Shinohara and Witter*, 2005; *Sawyer et al.*, 2011]) suggests the presence of a common magmatic gas fingerprint across several thousands of kilometers of the arc. We further propose the first estimate of the total volcanic gas output from the CVZ in northern Chile.

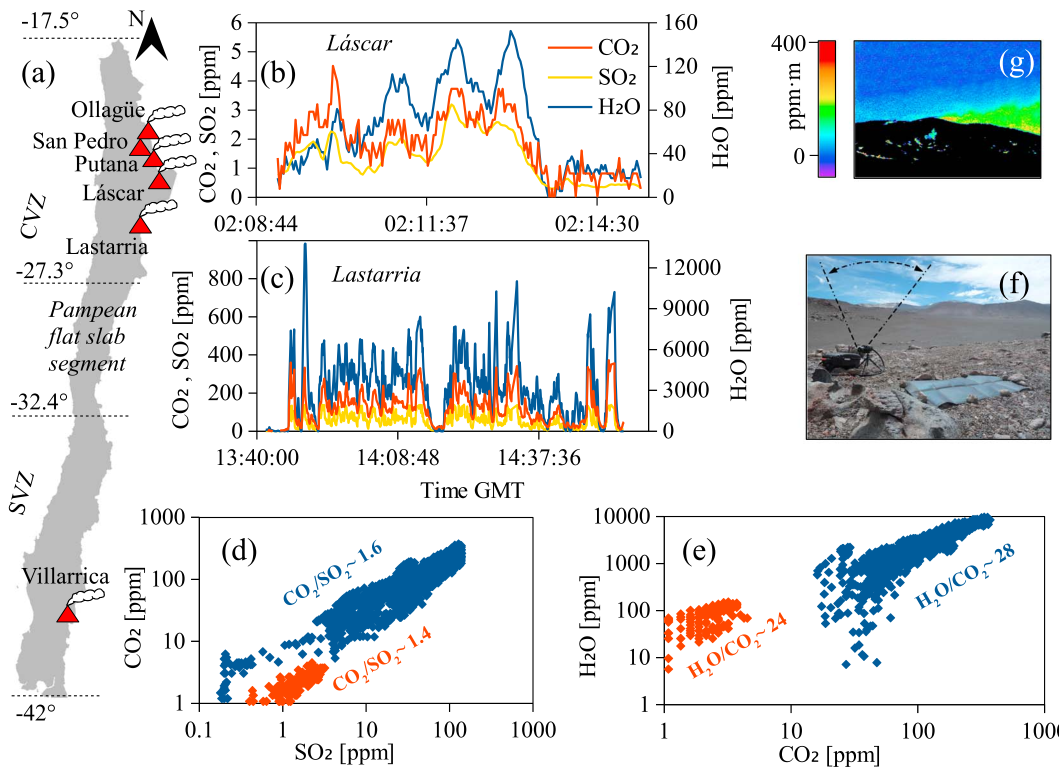

Figure 1. (a) Map of Chile showing the volcanoes of the CVZ and the SVZ targeted in this study. (b, c) Time series of CO_2, SO_2, and H_2O background-subtracted concentrations measured with the Multicomponent Gas Analyzer System (MultiGAS) at Láscar (Figure 1b) and Lastarria (Figure 1c). (d, e) Despite the significant temporal and spatial variabilities of concentrations, remarkably constant molar ratios were obtained at both Láscar (red) and Lastarria (blue). (f) Mobile scanning Mini-differential optical absorption spectroscopy (DOAS) instrument deployed at Lastarria. (g) SO_2 column densities measured at Putana volcano with an SO_2 camera on 5 December 2012.

2. Volcanic Activity

Láscar is a calc-alkaline stratovolcano located east of Salar de Atacama (23.37°S, 67.73°W) and is the most active volcano of the Chilean segment of the central Andes [*Oppenheimer et al.*, 1993]. Several small-to-moderate vulcanian eruptions have been recorded from Láscar since the midnineteenth century (some recently registered eruptions occurred in 2013, 2006–2007, 2005, 2002, 2000, 1994–1995, 1994, 1993–1994, 1993, and 1991–1992), most of which created ash columns with heights of up to a few kilometers above the summit. The activity of Láscar has been cyclic since 1984 [*Matthews et al.*, 1997]; in each cycle a lava dome is extruded into the active crater, accompanied by vigorous degassing through high-temperature fumaroles distributed on and around the dome. The Quaternary Lastarria volcano is located on the Chile-Argentina border at 25.17°S, 68.50°W and rises to 5706 m above sea level (asl). The north-northwest-trending edifice contains five nested summit craters. The youngest ($<$0.3 Myr) volcanic feature is a lava dome on the northern crater rim [*Naranjo*, 1986]. Although no historic eruption has been recorded, the youthful morphology of deposits and the ongoing uplift (~2.5 cm yr^{-1}) that began in 1996 [*Pritchard and Simons*, 2002; *Froger et al.*, 2007] support the active nature of Lastarria. Putana volcano is a 600 km^2 cone of andesitic-basaltic to dacitic lavas with a summit crater that is ~0.5 km in diameter at 5890 m asl. It has been characterized since the nineteenth century by persistent active degassing from four main summit fumarole fields, which feed a sustained steam plume [*Casertano*, 1963]. San Pedro (21.88°S, 68.14°W) is a composite basaltic andesite-to-dacitic volcano with several recorded historic eruptions, most recently in 1960 [e.g., *O'Callaghan and Francis*, 1986]. Steady fumarole activity on the upper western volcano flank has been documented from at least 2012 (Observatorio volcanológico de los Andes del Sur Monthly Reports). Ollagüe (21.30°S, 68.18°W) is also a composite volcano, consisting of high-K calc-alkaline rocks of dominantly andesitic to dacitic composition, but also comprises basaltic andesites [*Feeley et al.*, 1993]. The last eruption occurred in 1903, and the present-day persistent fumarole activity is associated with a summit lava dome.

Table 1. Summary Table of Derived Volatiles Output

Volcano		H_2O	CO_2	SO_2	H_2S	HCl	HF	HBr	Total ($t\,d^{-1}$)
Putana ($t\,d^{-1}$)				68.5					68.5
Lastarria ($t\,d^{-1}$)		11,059	973	884	174	385	5.8	0.6	13,480
Láscar ($t\,d^{-1}$)		5,192	534	554	30	199	9.4	0.15	6,517
Ollagüe ($t\,d^{-1}$)				150					76
San Pedro ($t\,d^{-1}$)				161					81
Total[a]	($t\,d^{-1}$)	16,251	1,506	1,818	204	604	15.2	0.76	>20,220
	($t\,yr^{-1}$)	6×10^6	5.5×10^5	6.7×10^5					
Total CVZ[b]	($t\,d^{-1}$)	$2.2–3.3 \times 10^4$	1,743–1,988	1,818					
	($t\,yr^{-1}$)	$8–12 \times 10^6$	$6.4–7.3 \times 10^5$	6.7×10^5					
Extrusive + Intrusive[c] ($t\,yr^{-1}$)			$3.3–8.1 \times 10^5$	5.3×10^{-5}					

[a]Calculated from measured gas compositions and fluxes.
[b]H_2O and CO_2 fluxes calculated using molar $H_2O/CO_2 = 30–40$ and $CO_2/SO_2 = 0.9–2.2$ for Putana, Ollagüe, and San Pedro volcanoes.
[c]Calculated petrological volatiles output.

3. Results

3.1. Láscar

At the time of our field survey of Láscar in 2012 (during 4–7 December), a sustained steam plume was produced by the persistent fumarole field, which is located ~200 m below the rim of its active central crater. The MultiGAS was deployed on the crater's rim in order to measure the bulk plume composition. We detected a dilute plume with strongly correlated volcanic H_2O, CO_2, and SO_2 mixing ratios (Figure 1b). The CO_2/SO_2 and H_2O/CO_2 molar ratios ranged from ~1 to ~1.7 and from ~12 to ~34, respectively, during our monitoring period (Figure 1d). Sets of base-treated filter packs were simultaneously collected to derive the in-plume halogen/SO_2 ratios. Our 2012 results ($HCl/SO_2 \sim 0.7$, $HF/SO_2 \sim 0.06$, and $HBr/SO_2 = 2.2 \times 10^{-4}$) indicate a more F-poor gas than in 2003 ($HCl/SO_2 \sim 0.6$ and $HF/SO_2 \sim 0.5$ [*Mather et al.*, 2004]). The Láscar plume was measured for several hours on 3–7 December 2012 and on 2–4 and 8 December 2013 by placing one or two scanning Mini-DOAS stations at various distances from 2.5 to 11 km downwind of the summit. The overall SO_2 output over the two measurement periods was $554 \pm 217\,t\,d^{-1}$ (mean ± standard deviation). Although SO_2 emissions from Láscar occasionally peak at $\sim 2300\,t\,d^{-1}$ [*Andres et al.*, 1991; *Mather et al.*, 2004], our results agree well with previous results obtained during periods of low degassing [*Andres et al.*, 1991; *Matthews et al.*, 1997; *Henney et al.*, 2012].

3.2. Lastarria

There has been continuous fumarole activity at the summit (upper fields) and northwest flank (bottom field) of Lastarria since the earliest available records. The bottom field is located at ~5000 m asl and is the largest (~0.023 km^2) emission area on the volcano. On 27 November 2012, we used the MultiGAS to characterize the chemical structure and heterogeneity of the bottom field. Our analysis reveals that the Lastarria bottom field has a homogeneous composition (see Figure 1c). The mean CO_2/SO_2 molar ratio was 1.6 (Figure 1d; range, 1.1–2.3), and the characteristic H_2O/CO_2 and H_2/H_2O ratios were 27.8 ± 2.8 and $6 \pm 2 \times 10^{-5}$, respectively. Our filter pack-based halogen/SO_2 molar ratios were $HCl/SO_2 \sim 0.8$, $HF/SO_2 \sim 0.022$, and $HBr/SO_2 = 5 \times 10^{-4}$. The plume was measured downwind of Lastarria for several hours on 27–29 November 2012 with the scanning Mini-DOAS stations placed about 8 km from the plume source. The wind speed at the plume height obtained using the Global Data Assimilation System model fitted well with our own anemometer data. The daily fluxes on these 3 days were 1917 ± 607, 473 ± 188, and $433 \pm 314\,t\,d^{-1}$, respectively, with an overall value for the 3 days of $884 \pm 779\,t\,d^{-1}$.

3.3. Putana, San Pedro, and Ollagüe

On 5 December 2012, we performed UV camera measurements at ~4.7 km from Putana crater (Figure 1g), which yielded an SO_2 output from this volcano of $40 \pm 11\,t\,d^{-1}$ (Table 1). The plume was again measured for several hours on 5, 6, and 9 December 2013 using two scanning Mini-DOAS stations. The daily fluxes on

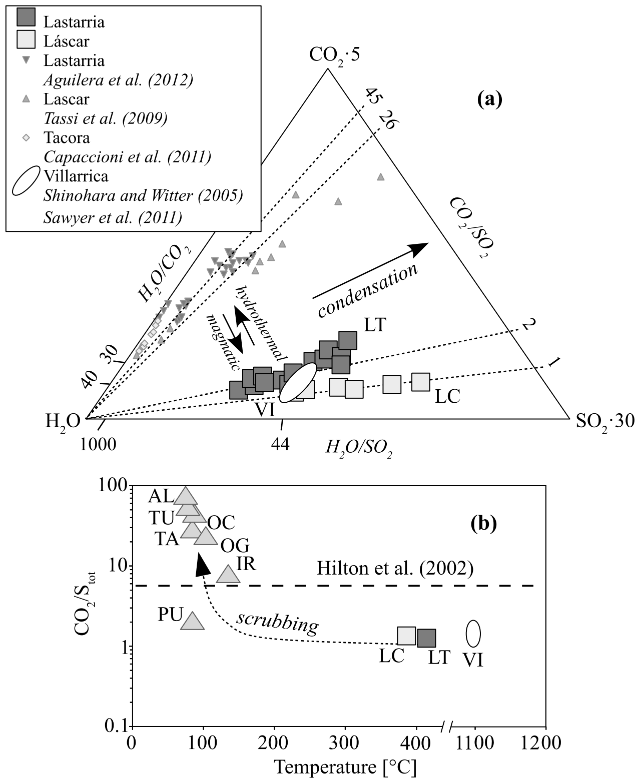

Figure 2. (a) Triangular H_2O-CO_2-SO_2 plot showing the water variability in Lastarria (LT) and Láscar (LC). Compositional data obtained in previous studies (Lastarria, Láscar, and Tacora) show a more hydrothermal nature of the sampled fumarolic gases. Gas compositions converge toward the composition of Villarrica high-temperature gas, which is less affected by water condensation. (b) Temperature dependence of CO_2/S_{tot} molar ratios in gas samples from Alitar (AL), Tupungatito (TU), Olca (OC), Ollagüe (OG), Tacora (TA), Irruputuncu (IR), Putana (PU), Láscar (LC), and Lastarria (LT) in the CVZ within northern Chile, and from Villarrica (VI) in the SVZ. Additional data are from *Aguilera et al.* [2012], *Capaccioni et al.* [2011], *Benavente et al.* [2013], *Tassi et al.* [2011], and *Shinohara and Witter* [2005].

these 3 days were 55 ± 14, 133 ± 104, and 77 ± 24 t d^{-1}, respectively, with an overall value of 97 ± 78 t d^{-1}. Similarly, the plumes of San Pedro and Ollagüe were measured for several hours using scanning Mini-DOAS instruments on 10 and 12 December 2013 and on 11 and 12 December 2013, respectively; the daily fluxes were 182 ± 188 and 150 ± 140 t d^{-1} at San Pedro (overall, 161 ± 150 t d^{-1}), and 47 ± 18 and 220 ± 181 t d^{-1} at Ollagüe (overall, 150 ± 162 t d^{-1}) (Table 1). Unfortunately no MultiGAS or filter pack data are available for these volcanoes.

4. Discussion

4.1. Magmatic Gas End-Member Composition of Chilean Volcanism

Figures 2, 3 compare our Láscar and Lastarria data set with the results of previous observations at Chilean volcanoes, from Tacora in the north (17°43.2′S) to Villarrica in the south (39°25.2′S). To identify the nature (magmatic versus hydrothermal) of the measured gases, we report in Figure 2a two distinct gas end-members: Villarrica gas and Tacora gas. Gas samples from Villarrica, which originate from a persistent lava lake, represent the best available compositional proxy for the magmatic gas end-member in Chile. In contrast, the CO_2-rich (CO_2/SO_2 ~ 27.9) signature of Tacora gas [*Capaccioni et al.*, 2011] is typical of residual fluids formed after prolonged gas-water-rock interactions [*Symonds et al.*, 2001] and is therefore taken as representative of the hydrothermal compositional end-member.

Our Láscar and Lastarria results demonstrate considerable fluctuations of water contents (at nearly constant CO_2/S molar ratios) at both volcanoes during the measurement periods (Figure 2a). We ascribe this effect to variable extents of water loss—due to condensation—from the plume prior to the sampling associated with MultiGAS measurements. However, we notice that the more-hydrous and thus possibly less-fractionated compositions of the Lastarria and Láscar gas samples converge in Figure 2a toward the composition of Villarrica gas [*Shinohara and Witter*, 2005], and cluster at CO_2/SO_2 and H_2O/CO_2 molar ratios of 1–2 and 30–40, respectively. We conclude from Figure 2a that both the Lastarria and Láscar gas samples have a clear magmatic signature. This is in stark contrast with the more S-depleted (Figure 2a) and Cl-depleted (Figure 3) compositions seen in previous direct-sampling studies performed at Láscar (2002–2006 [*Tassi et al.*, 2009]) and Lastarria (2006–2009 [*Aguilera et al.*, 2012]), which supported the presence of gases with a more hydrothermal nature. This difference may merely reflect dissimilar sampling conditions: while the 2012 MultiGAS observations concentrated on the "bulk" plume, the earlier direct sampling studies (2002–2009) concentrated on a small number of easily accessible, low-flux fumaroles. These fumaroles are potentially more affected by secondary processes (e.g., scrubbing), making their composition potentially unrepresentative of bulk gas emissions. Alternatively, the more magmatic signature of the 2012 gases, relative to pre-2009

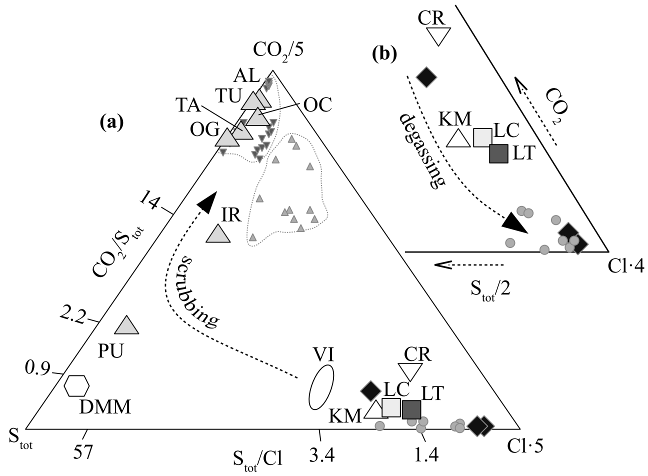

Figure 3. (a) Triangular CO_2-S_{tot}-HCl plot of the molar gas compositions of northern Chile volcanoes and Villarrica. The compositions of the melt inclusions from Villarrica (dark gray diamonds) and residual glasses from Llaima (small gray circles) are shown. Previous data of Láscar [*Tassi et al.*, 2009] and Lastarria [*Aguilera et al.*, 2012] fumaroles are displayed within the dotted gray lines. For comparison, the Costa Rica magmatic gas (CR, white triangle) from A. Aiuppa et al. (The Costa Rica-Nicaragua volcanic segment: along-arc variations in volcanic gas chemistry and an improved CO_2 budget, submitted to *Earth and Planetary Science Letters*, 2014) is also shown. Compositional data of depleted mid-ocean ridge basalt mantle (DMM, white hexagon) are from *Saal et al.* [2002]. (b) Magnification of the lower right part of the triangular plot highlighting a degassing trend from a CO_2-rich end-member to a Cl-rich end-member.

observations, may indicate a real evolution of the volcanic systems: degassing and seismic activities have only recently resumed at Láscar [*Global Volcanism Program*, 1994], and ground uplift has intensified at Lastarria in the past few years [*Froger et al.*, 2007].

The plot of CO_2/S_{tot} versus outlet temperature in Figure 2b offers additional evidence for identifying the magmatic versus hydrothermal origin of the 2012 gases. The CO_2/S_{tot} molar ratios in Chilean gas samples vary by 2 orders of magnitude, from ~80 in low-temperature gases (<150°C) down to ~1 at magmatic temperatures (Figure 2b). This trend, which has recently been established similarly for the Northwest Pacific Arc Region [*Aiuppa et al.*, 2012], is interpreted to reflect the increasing action of secondary processes at low temperature; under these conditions, the reactive S and Cl species are selectively removed by gas scrubbing [*Symonds et al.*, 2001], masking the pristine magmatic gas signature (Figures 2b and 3). In contrast, secondary processes exert only marginal effects at the high-temperature conditions of the 2012 Lastarria and Láscar samples. The magmatic signature of these gases is further demonstrated by comparison with the volatile compositions of the melt inclusions and residual glasses from Villarrica [*Witter et al.*, 2004] and Llaima [*De Maisonneuve et al.*, 2012] (Figure 3). The melt inclusions and residual glasses data record the evolution of volatiles during magma ascent and degassing, from a deep-seated magma with an inferred initial CO_2 content of 2480 ± 1400 ppm (for Villarrica [*Witter et al.*, 2004]) to a more-degassed, Cl-rich magma represented by the residual glasses (Figure 3). The compositions of Lastarria and Láscar gas samples overlap with the compositions of melt inclusions, reinforcing the magmatic nature of the gases, further suggesting that shallow magma degassing is the source of gas emissions at both volcanoes.

In view of the similarity between the SVZ (Villarrica) and CVZ (Lastarria and Láscar) data, we argue that a common magmatic gas end-member—with $H_2O/CO_2 = 30–40$, $CO_2/S_{tot} = 0.9–2.2$, and $S_{tot}/HCl = 1.4–3.4$—can be considered to be characteristic of the entire Chilean arc. This fits very well with volatile compositions measured or inferred for primitive melt inclusions from the SVZ, which typically have H_2O/CO_2 ratios of 30–75 [*Watt et al.*, 2013], and typical S/Cl rations of 1 to 2 (range: 0.1–3) [*Wehrmann et al.*, 2014a]. Our inferred magmatic CO_2/S_{tot} signature for Chile (0.9–2.2) is approximately fourfold smaller than that proposed by *Hilton et al.* [2002] (Figure 2b), which is probably due to their data set being dominated by low-temperature gases. With our new compositional data, the Chilean arc sits at the CO_2-poor margin of volcanic arc gas compositions [*Fischer*, 2008]. Our Chilean gas compositions also match well the accepted range of arc gas S_{tot}/HCl compositions, the majority of which cluster at ~2 [*Aiuppa*, 2009]. The relatively uniform gas composition from the CVZ and the SVZ supports the presence of similar volatile contributions to Chilean magmas from mantle and subduction-derived fluids, and relatively uniform composition of the latter along the entire arc length. Although along-arc variations have been identified in the SVZ in both extent of crustal contamination [*Hidreth and Moorbath*, 1988], isotopic signature of magmas [*Jacques et al.*, 2014], and S-Cl volatile contents in melt inclusions [*Wehrmann et al.*, 2014a], however, the trace-element proxies (e.g., Ba/La) for slab contribution show far more uniform behavior in SVZ [*Wehrmann et al.*, 2014b] and CVZ [*Matthews et al.*, 1994, 1999; *Richards et al.*, 2013] magmas than in other arc segments (e.g., CAVA [*Sadofsky et al.*, 2008]). Detailed geochemical studies show that the SVZ slab component is also uniform across the arc [*Jacques et al.*, 2013]. Thus, we interpret the similarly low Ba/La ratios (15–25) in SVZ and CVZ magmas to imply a uniform, more modest (relative to CAVA) contribution from slab-derived fluids, fitting well the low-carbon signature of Chilean volcanic gases seen in this study. We admit, however, that since only three volcanoes with high-T gases were measured, more observations are needed to corroborate our hypothesis.

4.2. Volatiles Output From the CVZ

Given the magmatic nature of the gas samples discussed above, we now attempt to constrain the total output of volatiles from the CVZ. The scanning Mini-DOAS and UV camera measurements made at Lastarria, Láscar, Putana, San Pedro, and Ollagüe lead to a cumulative SO_2 output from the five volcanoes of ~1800 t d^{-1} (or 6.6×10^5 t yr^{-1}) in 2012–2013 (Table 1). Three additional volcanoes (Irruputuncu, Olca, and Tacora) in this arc segment are reported to comprise high-temperature fumaroles [*Capaccioni et al.*, 2011; *Tassi et al.*, 2011]. The strongest S emitter of the three volcanoes is probably Irruputuncu, a volcano that typically vents S-rich gas [*Tassi et al.*, 2011]. However, even Irruputuncu has regularly failed to produce any statistically significant signal in OMI (Ozone Monitoring Instrument) satellite data sets [*Carn et al.*, 2013], implying an SO_2 flux of ≤190 t d^{-1} (considering the OMI detection limit quoted by *Fioletov et al.* [2011]). Assuming that each of the three volcanoes emits ~100 t d^{-1} on average, the cumulative SO_2 output would only rise to a maximum of ~2100 t d^{-1}. We therefore consider the cumulative SO_2 output from Lastarria, Láscar, Putana, San Pedro, and Ollagüe as a good proxy for the total SO_2 output from the CVZ (Table 1). We avoid using extrapolation techniques to quantify unmeasured emissions [e.g., *Hilton et al.*, 2002] because the validity of the power law assumption of *Brantley and Koepenick* [1995] has recently been questioned [*Mori et al.*, 2013]. However, we argue that if the recently recorded peak (2300 t d^{-1}) SO_2 emissions from Láscar were to be taken into account [*Andres et al.*, 1991; *Mather et al.*, 2004], the cumulative CVZ SO_2 output would increase to ~3500 t d^{-1} (1.3×10^6 t yr^{-1}). With our assumptions, the total CVZ SO_2 output of 6.6×10^5 to 1.3×10^6 t yr^{-1} would therefore correspond to 19–40% of the extrapolated annual flux from the Andes (~3×10^6 t yr^{-1} [*Andres and Kasgnoc*, 1998; *Hilton et al.*, 2002]), and to 3.3–5.1% of the estimated global SO_2 fluxes from subduction zone volcanoes [*Andres and Kasgnoc*, 1998; *Hilton et al.*, 2002; *Shinohara*, 2013]. For comparison, this inferred CVZ SO_2 output is of the same order as the combined SO_2 output from Villarrica and Llaima (~3.5×10^5 t yr^{-1} [*Mather et al.*, 2004; *Sawyer et al.*, 2011]), which are the two most actively degassing volcanoes in the SVZ (a similar rate of quiescent SO_2 degassing (~7.66×10^5 t yr^{-1}) was recently proposed by *Voelker et al.* [2014] for the SVZ). Considering the unknown but potentially additional large contributions from recently erupting volcanoes (e.g., Puyehue-Cordón Caulle and Copahue), SVZ is probably the strongest degassing source in Chile.

The measured SO_2 fluxes combined with the volcanic gas compositions of Láscar and Lastarria provide the basis for quantifying the outputs of other major volatiles. Despite its apparent long inactivity, Lastarria currently represents the most important gas source in the CVZ, with a total volatiles output (~13,500 t d^{-1}) that is twice that of Láscar (~6517 t d^{-1}) (Table 1). These two volcanoes have a combined CO_2 output of ~1500 t d^{-1}. We additionally estimate, based on a total SO_2 output of ~1800 t d^{-1} and a magmatic CO_2/S_{tot} ratio of 0.9–2.2, that the total CO_2 output from the CVZ is 1743–1988 t d^{-1}. Based on these calculations, northern Chile contributes only a minor fraction (~1%) of the total CO_2 output from subaerial volcanoes worldwide [*Burton et al.*, 2013]. We stress that this CO_2 budget does not take into account the contribution of diffuse soil degassing, which is poorly constrained for Chilean volcanoes, except for Villarica. At this volcano, CO_2 soil degassing was found to be negligible compared to summit crater degassing [*Witter et al.*, 2004].

In an attempt to evaluate how representative our 2012–2013 measurements are of the long-term degassing behavior of the CVZ, we compared the present-day gas fluxes of Table 1 with petrological volatiles output inventories. In the last 12 Myr, the rate of magma extrusion along the ~330 km long Central Andean trench (from 12°30α′S to 22°30′S) has been ~2.2×10^{-6} km^3 km^{-1} yr^{-1} [*Crisp*, 1984]. A comparable rate (10–13×10^{-6} km^3 km^{-1} yr^{-1}) was estimated by *Voelker et al.* [2011] for the SVZ. If we tentatively extend the Central Andean magma extrusion rate to northern Chile and scale it to the total trench length of 1500 km, we then obtain an order of magnitude estimate for the CVZ magma eruption rate of ~3.3×10^{-3} km^3 yr^{-1}. For a magma density of 2800 kg m^{-3} (andesite) and for preeruptive and posteruptive magmatic S contents of ~4100 and ~300 mg kg^{-1}, respectively (as for S in primitive undegassed melt inclusions and residual glasses of Láscar [*Andres et al.*, 1991]), this would correspond to a time-averaged SO_2 output of ~7.6×10^4 t yr^{-1} from CVZ volcanism. The proposed (mass) ratio between intrusive and extrusive volcanism for the Andes is ~6/1 [*Crisp*, 1984], which leads to a total (intrusive + extrusive) SO_2 output of ~5.3×10^5 t yr^{-1} (Table 1). Finally, the magmatic gas CO_2/S_{tot} ratio of ~0.9–2.2 (molar) converts the SO_2 output into a total CO_2 output of 3.3–8.1 × 10^5 t yr^{-1} (Table 1). The resulting annual SO_2 and CO_2 petrological outputs are in close agreement with our present-day (2012–2013) outputs of ~6.4–7.3 × 10^5 and 6.3×10^5 t yr^{-1}, respectively (Table 1). While our calculations here should be taken as order of magnitude estimates (given the overall uncertainties in the input parameters), the similarity of these values suggests (i) that the present-day fluxes are representative of the long-term CVZ degassing behavior and (ii) that Láscar and Lastarria contribute most of the CVZ gas output. The absence of Deep Sea Drilling Project/Ocean Drilling Program reference sites offshore of northern Chile, and the lack of knowledge about the compositions of subducted materials [*Plank and Langmuir*, 1998], precludes quantitative comparison between our measured CO_2 output and the CO_2 input flux at the trench (as recently obtained for other arc segments [*Freundt et al.*, 2014; *Voelker et al.*, 2014]).

5. Conclusions

The Láscar and Lastarria volcanoes in northern Chile emitted in 2012–2013 a typically magmatic gas phase whose compositions of major species (H_2O/CO_2 = 30–40, CO_2/S_{tot} = 0.9–2.2; S_{tot}/HCl = 1.4–3.4) resembled those of high-temperature open-vent emissions from Villarrica in the SVZ. This similarity suggests stable magmatic volatiles contents and origin along several thousands of kilometers of the southern Andean trench. We calculate that the CVZ presently emits more than 10^7 t yr^{-1} H_2O-rich volcanic gases into the atmosphere, essentially in a quiescent (noneruptive) form. Our measured CO_2 (~1500 t d^{-1}) and SO_2 (~1800 t d^{-1}) outputs in 2012–2013 correspond, respectively, to ~1.3% and ~3.3–5.1% of the corresponding global gas outputs from arc volcanoes. These present-day fluxes closely match the petrologically estimated long-term outputs obtained from intrusive + extrusive magma fluxes and preeruptive S contents.

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