**CO₂ flux emissions from the Earth’s most actively degassing volcanoes, 2005–2015**

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The global carbon dioxide (CO₂) flux from subaerial volcanoes remains poorly quantified, limiting our understanding of the deep carbon cycle during geologic time and in modern Earth. Past attempts to extrapolate the global volcanic CO₂ flux have been biased by observations being available for a relatively small number of accessible volcanoes. Here, we propose that the strong, but yet unmeasured, CO₂ emissions from several remote degassing volcanoes worldwide can be predicted using regional/global relationships between the CO₂/Sₘ ratio of volcanic gases and whole-rock trace element compositions (e.g., Ba/La). From these globally linked gas/rock compositions, we predict the CO₂/Sₘ gas ratio of 34 top-degassing remote volcanoes with no available gas measurements. By scaling to volcanic SO₂ fluxes from a global catalogue, we estimate a cumulative “unmeasured” CO₂ output of 11.4 ± 1.1 Mt/yr (or 0.26 ± 0.02 · 10¹² mol/yr). In combination with the measured CO₂ output of 27.4 ± 3.6 Mt/yr (or 0.62 ± 0.08 · 10¹² mol/yr), our results constrain the time-averaged (2005–2015) cumulative CO₂ flux from the Earth’s 91 most actively degassing subaerial volcanoes at 38.7 ± 2.9 Mt/yr (or 0.88 ± 0.06 · 10¹² mol/yr).

Volcanism is the primary mechanism through which carbon (C) stored in the deep Earth is transferred to surface environments to feed C exchanges in the atmosphere-ocean-biosphere system. Over geological time, volcanic CO₂ emissions have been a key control on atmospheric-oceanic CO₂ levels, ultimately regulating evolution of climate and life on our planet. The global volcanic CO₂ flux in modern Earth remains inadequately known and, ironically, is less constrained for subaerial volcanoes than for the less-accessible mid-ocean ridges, for which the ³He flux or the CO₂/Ba ratio proxies have successfully been applied. Direct volcanic CO₂ observations at subaerial volcanoes are technologically challenging from both ground and space due to the large atmospheric CO₂ burden, and thus remain limited in number. The volcanic CO₂ flux can be quantified indirectly by combining simultaneous acquisitions of UV-sensed sulphur dioxide (SO₂) fluxes and gas compositions (CO₂/SO₂ ratios), but gas observational networks are still in a developing stage, resulting in sparse and incomplete gas catalogues. CO₂ flux data have so far been obtained for only <60 of the several hundred currently degassing Holocene volcanoes. CO₂ flux records are continuous enough only for a few (<=10) volcanoes where permanent instrumentation is operating, while sparse results (one or a few campaign-style measurements at most) are available for the remaining ~50. In addition, scarce or even no information exists for several top-ranking degassing volcanoes in remote regions of the world (e.g., Vanuatu, Papua New Guinea, the Solomon arc, and the Sunda-Banda arc in Indonesia). Attempts to extrapolate available measurements to all the subaerial degassing volcanoes have been problematic and require use of questionable statistical approaches. Estimates of the global volcanic CO₂ flux thus vary widely, from 66 to 540 Mt/yr.

Ideally, refining the volcanic CO₂ inventory would require a comprehensive record comprising simultaneous composition/emission measurements for all the currently active strong volcanic gas emitters globally. The top-degassing volcanic targets during 2005–2015 (Table 1) have recently been identified from satellite-based observations of the SO₂ flux using the Ozone Mapping Instrument (OMI). Carn et al. identified 91 volcanoes, listed in Table 1, releasing SO₂ at rates above the OMI detection limit of 16 tons/day. Gas CO₂/S₂ ratios (where S₂ is Total Sulfur, corresponding to SO₂ in these strongly degassing magmatic-volatile emitting volcanoes) are available for 57 out of these 91 volcanic sources, from which SO₂ fluxes can straightforwardly be converted.

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| A | B | Volcano       | Country | Lat    | Long   | Measured SO₂ flux (tons/day) | SD | Measured CO₂/NO₂ (molar) | Predicted CO₂/NO₂ (molar) | SD | L₀ = F₀ × H₀ | M₀ = F₀ × K₀ | N₀ | O |
|---|---|---------------|---------|--------|--------|----------------------------|----|--------------------------|--------------------------|----|--------------|-------------|----|---|
| Group |   |               |         |        |        |                             |     |                          |                          |    |              |              |    |    |
| 1   | Ambrym | Vanuatu      | –16.25  | 168.12 | 7356   | 3168                          | 1.5 | —                        | —                         | 0.4 | 7586         | —            | 3843 |   |
| 1   | Asama  | Japan        | 36.40   | 138.53 | 449    | 430                            | 0.8 | —                        | —                         | 0.2 | 247          | —            | 247  |   |
| 1   | Asu    | Japan        | 32.88   | 131.11 | 628    | 492                            | 1.8 | —                        | —                         | 0.5 | 777          | —            | 650  |   |
| 1   | Augustine | USA    | 59.35   | –153.45| 73     | 140                            | 1.5 | —                        | 0.7                       | 0.7 | 75           | —            | 148  |   |
| 1   | Avachinsky | Russia | 53.25   | 158.83 | 707    | 619                            | 1.2* | —                        | —                         | 0.3 | 584          | —            | 531  |   |
| 1   | Chikurachchi + Ebeko* | Russia | 50.33   | 155.46 | 496    | 469                            | 0.9* | —                        | —                         | 0.3 | 320          | —            | 317  |   |
| 1   | Cleveland USA - AK  | USA    | 52.83   | –169.77| 152    | 142                            | 1.0* | —                        | —                         | 0.3 | 105          | —            | 102  |   |
| 1   | Copahue | Argentina   | –37.86  | –71.16 | 341    | 425                            | 0.9 | —                        | —                         | 0.3 | 211          | —            | 272  |   |
| 1   | Dukono | Indonesia   | 1.68    | 127.88 | 1726   | 611                            | 0.4 | —                        | 0.1                       | 0.45 | 273          | —            | 206  |   |
| 1   | Etna    | Italy        | 37.73   | 15.00  | 2032   | 517                            | 8.5 | —                        | 2.2                       | 2.2 | 9083         | —            | 3844 |   |
| 1   | Erta     | Ethiopia     | 6.58    | 41.06  | 96     | 44                            | 1.7 | —                        | —                         | 0.5 | 104          | —            | 165  |   |

Continued
### Measured volcanoes

| Group | Volcano               | Country          | Lat  | Long  | Measured SO2 flux (tons/day) | SD | Measured CO2/ SO2 (molar) | Predicted CO2/ SO2 (molar) | I  | SD | Predicted CO2 flux (tons/day) | SD | Notes/Data Sources       |
|-------|-----------------------|------------------|------|-------|------------------------------|----|---------------------------|----------------------------|----|----|----------------------------|----|---------------------------|
| 3     | Popocatepetl          | Mexico           | 19.02 | −98.62 | 1658                          | 893 | 8.2                       | —                           | 7.0 | 9345 | —                           | —  | —                         |
| 3     | Stromboli             | Italy            | 38.79 | 15.21  | 181                           | 82  | 7.2                       | —                           | 2.8 | 894  | —                           | —  | 535                       |
| 4     | Ale-Daladilla + Erta Ale | Ethiopia       | 13.60 | 40.67  | 64                           | 24  | 2.3                       | —                           | 0.9 | 99   | —                           | —  | 56                       |
| 4     | Etna                  | Antarctica       | −77.53 | 167.17 | 52                           | 31  | 27.6*                     | —                           | 4.9 | 983  | —                           | —  | 612                       |
| 4     | Kiluaea               | USA              | 19.42 | −155.29 | 5019                         | 2275 | 0.9*                      | —                           | 0.2 | 2933 | —                           | —  | 1578                      |
| 4     | Nyiragongo + Nyamuragira | DR Congo       | −1.41 | 29.20  | 3533                         | 2408 | 6.5*                      | —                           | 1.2 | 15790 | —                           | —  | 11149                     |
| 4     | Piton de la Fournaise | Reunion, France  | −21.23 | 55.71  | 134                          | 162 | 0.3*                      | —                           | 0.1 | 28   | —                           | —  | 34                        |
| 5     | Maapi                 | Sumatra          | −0.39 | 100.46 | 34                           | 34  | 20.5*                     | —                           | 1.1 | 480  | —                           | —  | 485                       |

*Unmeasured* volcanoes: those for which no CO2 gas data exist.

| Group | Volcano               | Country          | Lat  | Long  | Measured SO2 flux (tons/day) | SD | Measured CO2/ SO2 (molar) | Predicted CO2/ SO2 (molar) | I  | SD | Predicted CO2 flux (tons/day) | SD | Notes/Data Sources       |
|-------|-----------------------|------------------|------|-------|------------------------------|----|---------------------------|----------------------------|----|----|----------------------------|----|---------------------------|
| 1     | Anatahan              | Northern Mariana Islands | 16.35 | 145.67 | 1335                         | 1867 | 1.2                       | —                           | 0.5 | —  | 1102                       | 1607 | —                         |
| 2     | Aoba                  | Vanuatu          | −15.40 | 167.83 | 2870                         | 1229 | 2.5                       | —                           | 0.7 | —  | 4933                       | 2524 | —                         |
| 2     | Bagana                | Papua New Guinea | −6.09 | 155.23 | 3779                         | 886  | 2.4                       | —                           | 0.7 | —  | 6245                       | 2335 | —                         |
| 2     | Barren Island         | India            | 12.28 | 93.86  | 243                           | 341  | 2.2*                      | —                           | 1.3 | —  | 372                        | 566  | —                         |
| 2     | Batu Tara + Lewotolo  | Indonesia        | −8.27 | 123.51 | 86                           | 63   | 2.6*                      | —                           | 1.3 | —  | 525                        | 2335 | —                         |
| 1     | Buhasa                | Philippines      | 12.77 | 124.05 | 206                          | 199  | 1.2                       | —                           | 0.5 | —  | 170                        | 179  | —                         |
| 1     | Chinginagak          | USA- AK          | 57.14 | −156.99 | 138                          | 127  | 1.2                       | —                           | 0.5 | —  | 114                        | 113  | —                         |
| 2     | Ebulobo               | Indonesia/ Nusa  | −8.82 | 121.18 | 86                           | 63   | 2.6*                      | —                           | 1.3 | —  | 153                        | 137  | —                         |
| 1     | Fuego + Pacaya        | Guatemala        | 14.47 | −90.88 | 252                          | 46   | 1.6*                      | —                           | 0.8 | —  | 269                        | 139  | —                         |
| 2     | Guna                  | Vanuatu          | −14.27 | 167.50 | 434                          | 382  | 2.5*                      | —                           | 0.7 | —  | 749                        | 668  | —                         |
| 4     | Jebel-at-Tair         | Yemen            | 15.55 | 41.83  | 103                          | 295  | 6.2*                      | —                           | 1.8 | —  | 445                        | 1527 | —                         |
| 1     | Kauloon               | Philippines      | 10.41 | 123.13 | 70                           | 182  | 1.2                       | —                           | 0.5 | —  | 57                         | 152  | —                         |
| 3     | Karangantang          | Indonesia/ Sulawesi | 2.78  | 125.40 | 313                          | 85   | 5.0*                      | —                           | 1.3 | —  | 1049                       | 403  | —                         |
| 1     | Karymsky              | Russia           | 54.05 | 159.45 | 912                          | 250  | 1.2                       | —                           | 0.5 | —  | 752                        | 373  | —                         |
| 2     | Kerinci               | Indonesia/ Sumatra | −1.70 | 101.26 | 294                          | 99   | 2.6*                      | —                           | 0.8 | —  | 525                        | 233  | —                         |
| 1     | Ketoi                 | Russia           | 47.34 | 152.48 | 139                          | 151  | 1.2                       | —                           | 0.5 | —  | 114                        | 133  | —                         |
| 1     | Kizimen               | Russia           | 55.12 | 160.36 | 711                          | 1544 | 1.2                       | —                           | 0.5 | —  | 387                        | 1297 | —                         |
| 1     | Korovin               | USA- AK          | 52.38 | −174.15 | 198                          | 160  | 1.2                       | —                           | 0.5 | —  | 163                        | 148  | —                         |
| 2     | Langila               | Papua New Guinea | −5.53 | 148.42 | 629                          | 527  | 2.3*                      | —                           | 0.7 | —  | 994                        | 886  | —                         |
| 2     | Manam                 | Papua New Guinea | −4.08 | 145.04 | 1484                         | 753  | 2.7*                      | —                           | 0.7 | —  | 2755                       | 1570 | —                         |

*From the Sunda-Banda gas-rock association; Table S1c, Fig. 2a*
Table 1. Gas composition and fluxes for the 91 strongest SO2 volcanic gas sources in 2005–2015 worldwide (from Carn et al., 2017). The quoted SO2 fluxes (column F) are 2005–2015 averages (and standard deviation, SD) taken from the compilation of ref.30. The “measured volcanoes” list includes those volcanoes for which SO2 flux and gas composition molar CO2/SO2 ratios have both been measured. Each volcano is assigned to a given Group (1–4) (column A) based on the original categorization of ref.36 (non-arc volcanoes are assigned to Group 4). Unless indicated (see references in column O), the measured CO2/SO2 ratios (column H) are from ref.36. For these strongly degassing volcanoes, we assume total S (ST; quoted in 36) equals to SO2; SO2 satellite detection for all these volcanoes implies high-emission temperatures and limited or no interaction with hydrothermal system (and thus trivial reduced S species, such as H2S). Marapi volcano in Sumatra is an exception because of its hydrothermal signature (high CO2/SO2, high H2S) and is not assigned to any specific group (N.D. = not determined). In cases where combined emissions from two volcanoes are listed in the original dataset30 (see volcanoes labelled with superscripts & and $ in column B), due to insufficient spatial OMI resolution, a weighted average was calculated from the available volcanic gas information for the 2 where possible. Otherwise, equal

| Group | Volcano       | Country          | Lat  | Long  | Measured SO2 flux (tons/day) | SD | Measured CO2/SO2 (molar) | Predicted CO2/SO2 (molar) | SD | Measured CO2 flux (tons/day) | Predicted CO2 flux | SD | Notes/Data Sources |
|-------|---------------|------------------|------|-------|-----------------------------|----|------------------------|--------------------------|----|--------------------------|------------------|----|---------------------|
| 1     | Michael       | South Sandwich Ild. (UK) | −57.80 | −26.49 | 263 | 63 | 1.2 | 0.5 | — | 217 | 104 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 1     | Montagu       | South Sandwich Ild. (UK) | −58.42 | −26.33 | 142 | 179 | 1.2 | 0.5 | — | 117 | 155 | *From the SA gas-rock association; Table S1b, Fig. 2d |
| 2     | Pulauweh      | Indonesia/ Nusa   | −8.32 | 121.71 | 60  | 65  | 2.6* | 1.3 | — | 108 | 130 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 2     | Reventador    | Ecuador           | −0.08 | −77.66 | 206 | 187 | 2.2* | 0.8 | — | 304 | 298 | *From the SA gas-rock association; Table S1b, Fig. 2d |
| 3     | Rinjani       | Indonesia/ Lombok | −8.42 | 116.47 | 74  | 131 | 4.3* | 1.3 | — | 219 | 392 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 3     | Sangeang Api  | Indonesia/ Nusa   | −8.21 | 119.07 | 71  | 150 | 4.9* | 1.3 | — | 239 | 508 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 1     | Santiaguito   | Guatemala         | 14.76 | −91.55 | 247 | 119 | 1.6* | 0.8 | — | 271 | 182 | *From the CAVA gas-rock association; Table S1a, Fig. 2a |
| 1     | Sarychev      | Russia            | 48.08 | 153.21 | 260 | 324 | 1.2 | 0.5 | — | 214 | 282 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 2     | Sinabung      | Indonesia/ Sumatra | 3.17  | 98.39  | 327 | 595 | 2.4* | 1.3 | — | 550 | 1043 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 2     | Slamet        | Indonesia/ Java   | −7.24 | 109.21 | 206 | 132 | 2.2* | 1.3 | — | 311 | 272 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 2     | Tavurur       | Papua New Guinea  | −4.24 | 152.21 | 1729| 2535| 2.6* | 0.7 | — | 3091 | 4607 | *From the Group 2 global gas-rock association; Table S1d, Fig. 3a |
| 2     | Timakula      | Solomon back-arc  | −10.38| 165.80 | 256 | 276 | 2.1* | 0.7 | — | 370 | 417 | *From the Group 2 global gas-rock association; Table S1d, Fig. 3a |
| 1     | Tofua         | Tonga Islands     | −19.75| −175.07| 284 | 89  | 1.2 | 0.5 | — | 235 | 122 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 2     | Tungurahua    | Ecuador           | −1.47 | −78.44 | 342 | 235 | 2.5* | 0.8 | — | 588 | 445 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |
| 2     | Tavurur       | Papua New Guinea  | −5.05 | 151.33 | 630 | 581 | 2.4* | 0.7 | — | 1040 | 1005 | *From the Group 2 global gas-rock association; Table S1d, Fig. 3a |
| 1     | Veniaminof    | USA- AK           | 36.17 | −159.38| 250 | 214 | 1.2 | 0.5 | — | 211 | 197 | *From the Sunda-Banda gas-rock association; Table S1c, Fig. 2g |

| Group Total (Mt/yr, 10^9 kg/yr) | Measured SO2 flux | SD | Measured CO2 flux | Predicted CO2 flux | Total CO2 flux |
|--------------------------------|-------------------|----|-------------------|-------------------|---------------|
| GRAND TOTAL                    | 23                | 15 | 27.4 ± 3.6        | 11.4 ± 1.4        | 38.7 ± 2.9    |
| GRAND TOTAL (10^12 mol/yr)     | 0.36              | 0.23 | 0.62 ± 0.08      | 0.26 ± 0.02       | 0.88 ± 0.06    |
gas contribution was assumed for the two volcanoes. The measured CO₂ flux (column L) is calculated from the product of F by H (the quoted standard deviations in column N are based on propagation of the respective errors). The “unmeasured volcanoes” list includes volcanoes for which gas CO₂/Sr data are unavailable. Thirteen of such “unmeasured” (for gas) volcanoes are sited in arc segments with no subducted carbonate-rich lithologies at the respective trenches, and are therefore assigned to Group 1 (e.g., they are assigned the mean CO₂/Sr of such “unmeasured” volcanoes are sited in arc segments with no subducted carbonate-rich lithologies based SO₂ flux measurements11,15. We estimate the cumulative CO₂ flux from the 57 volcanic sources with “measured” gas compositions by applying a Monte Carlo method (see Table 1) to the dataset. The obtained cumulative CO₂ flux for each volcano is left to vary randomly within its mean ± SD value, and the resulting CO₂ fluxes are summed up. The procedure is repeated 100 times, yielding 100 random-generated CO₂ fluxes. The GRAND TOTAL values quoted in the tables are ranges (mean ± 1 SD) of 70% of the three populations of random-generated sums (e.g., the 15% outliers on each end of the populations are omitted). With this procedure, the global volcanic CO₂ flux is assessed at 38.7 ± 2.9 Mt/yr, ~11.4 ± 1.4 Mt/yr of which is estimated for the 34 “unmeasured” volcanoes (those with no measured gas data available).

Results

CO₂ fluxes for the Earth’s best-studied volcanoes. Roughly ~62% of the 91 strongest volcanic SO₂ sources globally30 have been characterised for both SO₂ flux and (episodically) for volcanic gas compositions (Table 1). CO₂ fluxes are thus obtained (see “Methods”) by pairing the OMI-based time-averaged 2005–2015 SO₂ fluxes30 with the characteristic (mean) CO₂/SO₂ ratios in the corresponding high-temperature magmatic gases (data from ref.30 unless otherwise noted). The so-derived CO₂ fluxes (Table 1) range from 28 to 15,800 tons/day, and are in reasonable agreement (typically within a factor ≤40%) with the CO₂ fluxes estimated using ground-based SO₂ flux measurements31,32. We estimate the cumulative CO₂ flux from the 57 volcanic sources with measured gas compositions by applying a Monte Carlo method (see Table 1) to the dataset. The obtained cumulative “measured” flux is 27.4 ± 3.6 Mt/yr (or 0.6 ± 0.08×10⁻¹² mol/yr).

Matching gas and whole-rock trace element compositions. Thirty-four top-ranking volcanic SO₂ sources do not have gas compositional records (Table 1). We hereafter refer to such volcanoes without CO₂/Sr information as “unmeasured” volcanoes. We thus explore a methodology to predict the characteristic volcanic gas CO₂/Sr ratio of each of these 34 “unmeasured” volcanoes using their averaged trace-element volcanic rock composition (Table S1). gas CO₂/Sr ratios in arc volcanoes exhibit systematic global relationships with slab fluid trace-element proxies (e.g., Ba/La or Sr/Nd ratios) in the corresponding whole-rocks, which are interpreted as resulting from melting of subducted sediments in the slab (variably enriched in CO₂; ref.41). These relationships, once set at the scale of individual arc segments (Figs 1 and 2) or volcano Groups (Fig. 3), can now be used to infer the representative volcanic gas CO₂/Sr ratio signature of the 34 “unmeasured” volcanoes (Tables 1 and S1). The procedure is illustrated in Fig. 1 and Table 2 for Pacaya volcano as an example (see “Methods”). The initial step involves establishing a CO₂/Sr vs. Ba/La relationship using data for volcanoes for which both gas and
trace element data are available (for the specific Pacaya example, we use gas/whole-rock information for Central American volcanoes, see Table S1a). As in previous work, the representative CO$_2$/S$_T$ ratios used in the figure are obtained by averaging available results for high-temperature gas samples, in the attempt to reduce the effect of secondary processes (e.g., magmatic S scrubbing during gas-water-rock reactions) that become substantial at hydrothermal (temperature < 400°C) conditions. Secondly, regression analysis is used to fit the gas vs. trace-element trend via either a linear or logarithmic best-fit regression function. Tests made excluding (panel A) or including (panel B) the compositional point of the Depleted Mid-ocean ridge Mantle (DMM; refs 123,124) in the data-fitting found that the second option systematically led to the best-data fits (see Table 2). Finally, the preferred regression model function (RM3 in the Pacaya example; see panel B and Table 2) is used to calculate a “predicted” gas CO$_2$/S$_O$ from available Ba/La data for Pacaya whole-rocks (uncertainty is estimated from confidence interval at one standard deviation on the regression). Our inferred gas CO$_2$/S$_T$ ratio (1.4 ± 0.75; Table 2) is well within the magmatic gas range (CO$_2$/S$_O$ ratio of 1.1 ± 1.0) measured during recent plume observations. A similar CO$_2$/S$_T$ ratio (see Table 2) is predicted using the CAVA gas vs. Sr/Nd ratio association (panel C). In this plot, the yellow and green dashed lines are the linear best-fit regression lines for Group 1 and 2 sub-populations, respectively.

**CO$_2$/S$_T$ ratios from individual-arc gas vs. trace-element relationships.**

Gas vs. rock (trace element composition) associations are initially analysed at the scale of individual arc segments, in the assumption that, at such regional scales, sources and transport pathways of volatiles and trace elements are relatively uniform. In truth, intra-arc variations in thickness, age, thermal properties and composition of the slab and overlying plate, and in the composition of subducted sediments, are large enough to impact the mechanisms of magma generation, and thus impart regional trends in volatile and trace element signatures of erupted magmas. Nonetheless, it is on these individual-arc trends that we rely below. Three arc segments have enough volcanoes measured for both gases and rocks to allow reliable gas vs. rock associations to be established (Fig. 2).

The Central American Volcanic Arc (CAVA) CO$_2$/S$_T$ vs. Ba/La relationship, obtained from results listed in Table S1a, is illustrated in Figs 1 and 2a. The systematic along-arc trace-element patterns in CAVA volcanic rocks (Fig. 2b,c) originate from changes in geometry, age, thermal regime and extent of serpentinization of the subducting Cocos plate slab. As more recently found, such trace-element variations correlate with those of CO$_2$/S$_T$ ratios in high-temperature magmatic CAVA gases. These correlations (e.g., Figs 1 and 2a) have been explained.
as resulting from the variable addition of C-Ba-Sr-rich fluids issuing from melting of limestone-bearing slab sediments, with the highest slab-fluid influx being observed in Nicaragua, where magmatic gases consistently have C-rich (Group 2) affinity (Fig. 2a). In Costa Rica and El Salvador, magmatic gases are typically C-poorer (Group 1), in line with the lower slab affinity (and more depleted mantle-like signature) of trace-element ratios (Fig. 2). All the CAV A volcanic SO2 emitters of Table 1 have been measured for gas composition (at least for their CO2/ST ratio), except for Guatemalan volcanoes Fuego and Santa Maria. We use the CAV A CO2/ST vs. Ba/La association (of Fig. 2a) to fill this gap of knowledge. Using the RM3 regression model in tandem with mean whole-rock Ba/La ratios (Table S1a and Fig. 2a), we infer CO2/ST ratios of respectively 1.7 ± 0.75 (Fuego) and 1.6 ± 0.75 (Santa Maria).

Table 2. Comparison between measured and predicted (this work) volcanic gas CO2/ST ratios in the Pacaya magmatic gases. At the high-T magmatic gas conditions explored here, total S (ST) corresponds to SO2. The predicted CO2/ST ratios are obtained from the mean Ba/La ratio (or Sr/Nd; see RM5) in Pacaya whole-rocks using the regression functions through the CO2/ST vs. Ba/La (or Sr/Nd; see RM5) association for CAV A volcanoes (dataset listed in Table S1a). Five distinct regression functions are tested, being illustrated (with their corresponding equations and regression coefficients) in Fig. 1. RM1 and RM2 (Fig. 1a) use linear and logarithmic regression models, respectively, and do not include the composition of the Depleted Mid-ocean ridge Mantle in the fit. Regression models RM3 and RM4 (Fig. 1b) are, respectively, linear and logarithmic, and the composition of the Depleted mid-ocean ridge Mantle is included in the fit. Regression models RM5 (Fig. 1c) uses linear regression functions through the CO2/Sr vs. Sr/Nd association for Group 1 CAV A volcanoes. The linear regression model RM3 yields the highest regression coefficient (R2 = 0.7; see Fig. 1b), and is thus adopted here.
Our compilation (Table 1) shows that volcanic gas CO2/ST data are available for the majority of the volcanic SO2 emitters in the Northern (NVZ), Central (CVZ) and Southern (SVZ) Volcanic Zones of the Andes (Southern America). Very limited gas information is available for Ecuadorian volcanoes, however, and here we use the CO2/ST vs. Ba/La association (for South-America: Fig. 2d) to fill this knowledge gap. In the Andes, there is documented evidence in the literature for large along-arc variations in volcanic rock trace-element geochemistry. Our partial whole-rock dataset, based on the subpopulation of Andean volcanoes listed in Table S1b, demonstrates an overall south-to-north increase in trace-element slab-fluid proxies (Ba/La, Sr/Nd and U/Th; Fig. 2e,f), from Copahue volcano in Argentina (SVZ) to Nevado de Ruiz in Colombia (NVZ). Importantly, the along-arc variations in the volcanic gas CO2/ST ratio scale well with the trace-element variation patterns (Fig. 2d), again suggesting common source processes. The trace-element signature of the three most actively degassing volcanoes today in Ecuador, Tungurahua, El Reventador and Cotopaxi (the latter not appearing in the 91 list of top degassing volcanoes), places Ecuadorian magmatism in an intermediate position between Colombian volcanoes in the NVZ (the richest in Ba and Sr, but also CO2; Fig. 2d) and intermediate C-rich Peruvian volcanoes.
Further to the south (in the CVZ). The mean Ba/La ratios, combined with the CO₂/SF ratio (see legend). Trenches are differently colored depending on CO₂ bulk concentration in the trench sediments (data from ref. 43). The map shows that the most strongly CO₂ degassing volcanoes are clustered in the Vanuatu–Papua New Guinea arc segments, in Central America, Southern America (Northern Volcanic Zone), and in the Lesser Antilles, in addition to Italy (Etna), Congo (Nyiragongo + Nyamuragira) and Hawaii (Kilauea). Volcanic CO₂ fluxes are typically lower in higher latitude volcanic regions such as in the Aleutians–Kamchatka–Kurile and in the South–Sandwich Islands, where no carbonate-rich lithologies are subducted at the trenches. The map was generated using the open source QGIS software (available at https://www.qgis.org/it/site/) (Copyright © 2019 AIUPPA. Permission is granted to copy, distribute and/or modify this document under the terms of the GNU Free Documentation License, Version 1.3 or any later version published by the Free Software Foundation; with no Invariant Sections, no Front-Cover Texts, and no Back-Cover Texts. A copy of the license is included in the section entitled "GNU Free Documentation License"). The base map is a relief and bathymetry Raster called «Natural Earth II with Shaded Relief and Water» file #NE2_HR_LC_SR_W.tif (Made with Natural Earth. Free vector and raster map data @ naturalearthdata.com). As for the shaded relief, we use the CleanTOPO2 layer, a modified SRTM30 Plus World Elevation Data also edited by Tom Patterson, US National Park Service. The original source data is from ref. 125.

Figure 4. Global map illustrating the location of the 91 strongest volcanic CO₂ emitters (data from Table 1). CO₂ flux information for both “measured” (circles with black borders) and “unmeasured” (circles with red borders) volcanoes is shown. Dimension of the symbols is proportional to CO₂ flux, with color fill reflecting the CO₂/SF ratio (see legend). Trenches are differently colored depending on CO₂ bulk concentration in the trench sediments (data from ref. 43). The map shows that the most strongly CO₂ degassing volcanoes are clustered in tropical to sub-tropical regions such as the Vanuatu–Papua New Guinea arc segments, in Central America, Southern America (Northern Volcanic Zone), and in the Lesser Antilles, in addition to Italy (Etna), Congo (Nyiragongo + Nyamuragira) and Hawaii (Kilauea). Volcanic CO₂ fluxes are typically lower in higher latitude volcanic regions such as in the Aleutians–Kamchatka–Kurile and in the South–Sandwich Islands, where no carbonate-rich lithologies are subducted at the trenches. The map was generated using the open source QGIS software (available at https://www.qgis.org/it/site/) (Copyright © 2019 AIUPPA. Permission is granted to copy, distribute and/or modify this document under the terms of the GNU Free Documentation License, Version 1.3 or any later version published by the Free Software Foundation; with no Invariant Sections, no Front-Cover Texts, and no Back-Cover Texts. A copy of the license is included in the section entitled "GNU Free Documentation License"). The base map is a relief and bathymetry Raster called «Natural Earth II with Shaded Relief and Water» file #NE2_HR_LC_SR_W.tif (Made with Natural Earth. Free vector and raster map data @ naturalearthdata.com). As for the shaded relief, we use the CleanTOPO2 layer, a modified SRTM30 Plus World Elevation Data also edited by Tom Patterson, US National Park Service. The original source data is from ref. 125.

The case of Indonesia, which includes the Sunda–Banda and Sangihe–Halmahera arc segments, is particularly problematic (Fig. 2g–k). The large along- and within-arc variations in crustal63 and slab64 structures, combined with heterogeneities in the sedimentary slab input 42 (Fig. 4), make it difficult to characterize regional trends in volatile sources. In the Java sector of the Sunda arc, the respective roles of crust and slab in controlling rock65 and gas66 geochemistry are widely debated, with some authors stressing the importance of upper plate assimilation67,68, and others emphasising a slab control 69–71. The Group 3 signature 36 of Merapi and Bromo (Fig. 2g) supports involvement of crustal carbon in Central Java72. South-to-north along-arc trends in gas 3He/4He (decreasing) and CO₂/3He (increasing) ratios66 suggest a crustal volatile contribution is also likely in Sumatra, where the crust is especially thick and limestones widely exposed63,67. In contrast, crustal assimilation is supposedly minor (if any) in other sectors, including west and east-Java66, Nusa68,73 Banda74 and Halmahera33. In these segments of the Sunda-Banda and Sangihe-Halmahera arcs75, along-arc variations in He-C isotopes66,76,77, and the sparse high-temperature gas information, suggest variable C delivery from the slab, and thus coexistence of Group 1 and 2 volcanism (Fig. 2g). This is not unexpected, in view of the C heterogeneity in subducted sediments, from terrigenous and C-poor (Sumatra-Java) to pelagic and C-richer (Nusa, east Sunda)62 (Fig. 4). The diverse volatile sources that are possibly involved, in addition to the paucity of gas data, create scatter in CO₂/SF vs. Ba/La (Fig. 2g). Only 9 Indonesian volcanoes have been measured for both whole-rock trace element composition and (high-temperature) magmatic gas composition (Table S1c). These CO₂/SF vs. Ba/La data can be fitted by either a linear (RM3) or logarithm (RM4) regression model with identical regression coefficients (R² = 0.52; Fig. 2g). We therefore infer the CO₂/SF ratio signature of the “unmeasured” Indonesian volcanoes (Table 1) by averaging the output of the two regression models (Table S1c). The low regression coefficients (Fig. 2g) imply the inferred CO₂/SF ratios should be treated with caution, as they require validation/refinement with an improved (more than 9 data-points) gas vs. trace element relationship. We caution, in particular, that the predicted CO₂/SF ratios (Table 1) may either over-estimate (for Group 1 volcanoes) or under-estimate (for Group 3 volcanoes) by a factor ~1.3 (the max error in Fig. 2g) the real volcanic gas CO₂/SF ratios of “unmeasured” Indonesian volcanoes.
CO$_2$/S$_2$ ratios from Group-based gas vs. trace element relationships. Several of the “unmeasured” (for gas) volcanoes in Table 1 are sited in arc segments for which insufficient gas/rock information is currently available to establish individual-arc associations (as those analysed in Fig. 2). In order to derive information on their CO$_2$/SO$_2$ ratio gas signature, we use the global relationship between CO$_2$/ST and Ba/La in Groups 1–2 volcanoes (ref.36) (Fig. 3).

The majority of the remaining “unmeasured” (for gas) volcanoes in Table 1 are sited in arc segments for which available deep sea drill holes point to the lack of C-rich lithologies (limestones) in the subducted sediment succession42 (Fig. 4). Trench sediments poor in C have been identified in the segment of the Pacific Ring of Fire (Fig. 4) that stretches from Aleutians-Kuril-Kamchatka to the N/NW to Marianas/Japan/Philippines further south (10 “unmeasured” volcanoes in total – see Table 1). Where high-temperature gas information is available, a CO$_2$-poor (Group 1) signature of volcanic gases36 has typically been observed in such carbonate-poor trenches (Fig. 4), matching well the small sedimentary slab C input42. Sediments are similarly C-poor (e.g., prevalently terrigenous and biosiliceous42) in the Tonga and South Sandwich arcs (3 “unmeasured” volcanoes; Fig. 4). We therefore assign to Group 1 all the “unmeasured” (for gas) arc volcanoes fed by carbonate-poor trenches. Group 1 volcanoes exhibit little change in gas CO$_2$/ST ratios with increasing Ba/La (Fig. 3a). This implies either (i) limited C delivery from the slab in the absence of carbonated sediments (e.g., that fluids/melts delivered by terrigenous sediments, altered oceanic crust and/or serpentinite are not major C sources36), or (ii) that slab C and S are added to the mantle wedge in 1:1 to 4:1 proportions at most (Group 1 volcanoes typically have CO$_2$/ST ratios ~3–4 times higher than the DDM). The lack of dependence on Ba/La (Fig. 3a) means that we can prudently use the measured Group 1 CO$_2$/ST ratio average (1.2 ± 0.5; see Table S1d) for all the “unmeasured” (for gas) Group 1 volcanoes in Table 1.

Group 2 volcanoes are, by definition46, those having CaCO$_3$-rich sediments in their trenches. These volcanoes typically have more C-rich volcanic gas composition (CO$_2$/S$_2$ ratio >2 but ≤4) and exhibit stronger, steeper correlation between gas CO$_2$/ST and trace element ratios (Fig. 3a). These Group 2 volcanoes are located in high biological productivity zones close to the tropics, where sediments are increasingly biogenic in nature and/or where seafloor is shallow enough (above the calcite compensation depth, CCD) to support carbonate deposition42 (Fig. 4). Of the few remaining “unmeasured” (for gas) volcanoes in Table 1, those in the Papua New Guinea-Solomon-Vanuatu arc segment are thus potential candidates for Group 2. The Papua New Guinea-Solomon arc sectors (Fig. 4) are a particular challenge because no gas samples are available, and no deep sea drill holes have been placed in the seafloor of the Solomon Sea, seaward of their trenches. Likewise, there are few relevant piston cores to provide any seafloor samples. Our inferences are thus based on seafloor depth, assumptions about the regional CCD, and drill sites in other, nearby southwest Pacific marginal seas. At DSDP Site 63, in the East Caroline Basin north of New Britain, carbonate lithologies were encountered throughout the entire section, from the Quaternary to the middle Oligocene basaltic basement38. This site, at 4472 m water depth, has thus been above the CCD over its entire history. Similarly, drilling at DSDP 287 (4653 m water depth), in the Coral Sea south of Papua New Guinea and east of the Solomon Islands, intercepted abundant carbonate lithologies through most of the sedimentary section to its lower Eocene basement39. Given that the water depths of the Solomon Sea are predominantly <4500 m seaward of the New Britain, Solomon and Northern Vanuatu trenches, we expect this seafloor to have been above the CCD for much of its history as well, and thus to be delivering carbonate-rich sediment to these subduction zones. Based on the above, we consider it very likely that
“unmeasured” volcanoes in the Papua New Guinea- Northern Solomon-Vanuatu arcs belong to Group 2. We use therefore the CO₂/S ratio vs. Ba/La global association for Group 2 volcanoes (see Fig. 3a) to predict (based on trace element information) CO₂/S ratios ranging from 2.1 ± 0.7 to 2.7 ± 0.7 for these volcanoes (Tables 1 and S1e). We note that the two “measured” volcanoes in the central and southern Vanuatu arc (Bembow on Ambrym Island, and Yasur on Tanna Island) both exhibit Group 1 gas affinity (CO₂/S of 1.5–1.6), implying that the predicted C-richer gas signature for northern Vanuatu volcanoes requires validation from measurements.

**Discussion**

**Validity of whole rock trace element proxy for CO₂/S.** Our predicted CO₂/S ratios stand on the assumption that gas compositions are linked to trace element compositions of their source magmas at either regional (Fig. 2) or global (Fig. 3) scale. Implicit in establishing such relationships is that gas (CO₂/S) and trace-element (Ba/La) whole-rock tracers are inherited by the same processes at their source, and are similarly conserved during magma ascent, decomposition and degassing/eruption. For Ba/La, a link has been made between signatures of arc rocks and subducted sediments at corresponding tranches, so that this and other trace element ratios are commonly used as slab-fluid proxies for characterizing the mantle source of magmas. Both elements exhibit incompatible behaviour during magma differentiation, so that the source-inherited ratios are essentially conserved during magma evolution, at least for the mafic to intermediate (andesitic) magma compositions considered here (as outlined in the Method).

The behaviour of volatile components CO₂ and S is obviously complex during the generation and evolution of slab fluids and mantle-derived magmas. Not only are slab sources and processes only partially understood for C and S, but these volatile species will be selectively extracted from melt and partitioned into the vapour phase according to their complex solubilities (that depend on T–P–X–redox conditions). Upon magma decompression and differentiation, one may thus argue that degassing-related fractionations, for which abundant model, experimental and observational evidence exists, act as to render the CO₂/S ratios in both degassed melt (preserved in melt inclusions in phenocrysts) and exsolved vapour (discharged as volcanic gases) unrepresentative of the mantle source compositions, and thus unlinked to Ba/La or other trace element proxies.

Where sufficient data exists (e.g., Figs 2a,d and 3a), however, the CO₂/S vs. Ba/La correlations appear systematic and statistically significant, and we consider unlikely that these associations are purely accidental. Our regional/global associations here, thus, implicate that the time-averaged CO₂/S ratios of volcanic gases ultimately reflect the volatile ratios in the parental (un-degassed) melt, and in the mantle source. To reconcile this with the well-established degassing-driven CO₂ vs. S fractionations, we observe that, at least at mafic systems, comparison between measured and modelled (from numerical simulation of magma degassing paths using volatile saturation codes) gas CO₂/S ratios typically imply equilibrium pressures (e.g., pressures of final gas-melt segregation) of 0.1–3 MPa during quiescent degassing activity. Thus, at least during non-eruptive periods, during which the majority of the volcanic gas observations in the literature are taken, observations and models both indicate very shallow (a few hundred meters below the magma-air interface) gas segregation from the convecting feeding magmas. If shallow closed-system degassing conditions prevail, then the magmatic gas phase released as volcanic gas during open-vent activity does represent an integral of volatiles exsolved from melt during most separation depths at various volcanoes. In mafic systems, the SO₂ flux is a proxy for the rates of magma degassing in a volcano’s shallow (<3 km) plumbing system. As such, at least in principle, shallow magma ascent and decomposition should be tracked by increasing SO₂ flux and decreasing CO₂/S ratios in the surface gas output, a relationship that is not observed in our global dataset (Fig. 5). The SO₂ flux-independent, distinct CO₂/S distributions of Group 1, 2 and 3 volcanoes (see Fig. 5) suggest, instead, that source signature, rather than degassing, ultimately controls the longer-term, time-averaged volcanic gas compositions.

The lack of a systematic correlation between volcanic gas CO₂/S ratios and SO₂ fluxes (Fig. 5) further supports the idea that the former are not significantly affected by variable extents of magma degassing and gas-melt separation depths at various volcanoes. In mafic systems, the SO₂ flux is a proxy for the rates of magma degassing in a volcano’s shallow (<3 km) plumbing system. As such, at least in principle, shallow magma ascent and decomposition should be tracked by increasing SO₂ flux and decreasing CO₂/S ratios in the surface gas output, a relationship that is not observed in our global dataset (Fig. 5). The SO₂ flux-independent, distinct CO₂/S distributions of Group 1, 2 and 3 volcanoes (see Fig. 5) suggest, instead, that source signature, rather than degassing, ultimately controls the longer-term, time-averaged volcanic gas compositions. We caution that CO₂/S ratio volcanic gas compositions may become less source-related in intermediate to silicic systems, where the gas output is often buffered by gas-melt equilibration in crustal, vapour-saturated magma reservoirs. It is thus possible that part of the scatter in our gas vs. trace-element associations (Figs 2 and 3) is caused by the intermediate (andesitic) systems included in our dataset. Silicic systems have intentionally been excluded from our compilation.

The good match between our predicted and measured CO₂/SO₃ ratios at Pacaya volcano (Fig. 1a) also support, although indirectly, the validity of our gas vs. trace element associations. In addition to Pacaya, recent airborne gas measurements at Tungurahua and Cotopaxi volcanoes in Ecuador have found CO₂/SO₃ ratios (in the 2 to 2.5 range) fully overlapping our predicted range (2.5 ± 0.8; Table 1). These successful tests provide confidence in the robustness of our predicted CO₂/S ratios. We caution that, in order to validate our methodology further and reduce the scatter in gas vs. trace element scatter plots (e.g., Fig. 3g), gas observations should be prioritized in remote, unexplored volcanoes in Papua New Guinea, Sandwich Islands, Solomon Islands, Sumatra, east Sunda-Banda, and north-Vanuatu. In some of these arc segments (e.g., Sumatra, Sunda, crustal C may be involved, in which case our predicted CO₂/S ratios may underestimate the actual magmatic gas ratio (by a factor up to ~1.5–2). We also advise that, since only high-temperature (SO₂-dominated) gas data are used to...
establish our gas vs. trace-element associations (Figs 2 and 3), our predicted CO2/ST ratios are representative of the magmatic gas signature, irrespectively of whether or not hydrothermal processes are acting to alter the actual and total gas volcano emissions. For example, the hydrothermal (H2S-rich) gas emissions from Marapi volcano in Sumatra have measured CO2/ST ratios of 20.5 ± 1.1 (Table 1), well distinct from what we would predict (CO2/ST ratio of ~2.6) using the whole-rock Ba/La (19 ± 3; Table S1c) and the Indonesian gas vs. trace-element relationship (Fig. 2g). As such, discrepancy between measured and predicted CO2/ST ratio at any other hydrothermal volcano may lead to apportioning the fraction of S lost to (or C produced by) the hydrothermal system. While we believe that hydrothermal processing should be the exception rather than the rule for the satellite-sensed volcanoes here, we ultimately anticipate our predicted CO2/ST ratios (Table 1) will require revision and upgrading as new high quality gas data become available for newly measured volcanoes.

One important aspect to consider is that our regional/global associations (Figs 2 and 3) are based on averaging trace element information for rocks erupted during decades to millennia of volcanic activity. As such, the CO2/ST ratios predicted from such associations should be viewed as long-term means over a volcano's lifespan, rather than the instantaneous measurements as obtainable by direct gas observations. These “geologic” gas CO2/ST ratios may thus serve, when combined with measured S content in mafic glass inclusions, to estimate the initial CO2 content in parental, un-degassed melts, and eventually in the sub-arc mantle. Both are similarly poorly constrained due to pre- and post-entrapment loss to vapour of poorly soluble CO2.

A decadal global CO2 flux budget. Our predicted CO2/ST ratios are converted into CO2 fluxes (Table 1) by assuming ST = SO2 and scaling to the OMI-based mean SO2 fluxes for the 2005–2015 period30. We focus on the OMI satellite dataset owing to advantages brought by its global and coincident observations, but yet observe that quantitatively similar results would be obtained using ground-based SO2 flux observations instead15. The predicted CO2 fluxes range from 57 tons/day (Kanlaon volcano in the Philippines) to 6200 tons/day (Bagana volcano in PNG) (Figs 4 and 6). Uncertainty in the derived CO2 fluxes (see Table 1, column N) is based on propagation of the respective errors on SO2 flux (column G) and predicted CO2/ST ratios (column I).

The total cumulative CO2 emissions from the 34 “unmeasured” volcanoes (those with no measured gas information available) would thus be ~11.4 ± 1.1 Mt/yr (~0.26 ± 0.02·1012 mol/yr), thus adding an additional ~34% to the cumulative “measured” mean CO2 emissions in 2005–2015 (27.4 ± 3.6 Mt/yr; Table 1). Finally, our extrapolated (measured + predicted) CO2 flux budget is 38.7 ± 2.9 Mt/yr (or 0.88 ± 0.06·1012 mol/yr). It is important to notice that our approach, in which the CO2/ST ratio signature of each volcano is independently evaluated, leads to far better constrained CO2 budget (7% uncertainty at 1 SD) that would be possible using any “averaged” volcanic

Figure 6. The 28 top-ranking volcanic point sources of SO2 (left) and CO2 (right) during 2005–2015. Data are from Table 1. SO2 fluxes are 2005–2015 means from ref.30. The CO2 fluxes are calculated from SO2 using measured or predicted CO2/ST ratios (see Table 1). Different volcano groups are identified by different colours. The global CO2 budget is dominated by CO2-rich Group 2–3 arc volcanoes. Two rift volcanoes (Nyiragongo and Nyamuragira) and one within-plate (WP) volcano (Kilauea) appears in the top-10 list of CO2 emitting volcanoes110.
CO2/S02 ratio proxy (as has been often attempt in past studies). For example, scaling the mean global SO2 flux (23 ± 15 Mt/yr) to the mean volcanic CO2/S02 ratio (2.7 ± 3.6) (all data from Table 1) would lead to a global CO2 flux of 62 ± 92 Mt/yr (e.g., 148% uncertainty at 1 SD).

Based on our results, we infer that 6 strongly degassing volcanoes with time-averaged (2005–2015 means) CO2 fluxes of ≥ 5000 tons/day dominate the global CO2 budget (Figs 4 and 6). One of these (Bagana, PNG) is an “unmeasured” volcano and would not have been identified as a top CO2 emitter without the proxy approach developed here. It is interesting to observe that while the SO2 global budget is dominated by the Group 1 volcanoes (accounting for 13 out of the 28 strongest volcanic SO2 sources; Fig. 6), the CO2 global budget is predominantly determined by the SO2-enriched arc volcanoes in Group 2 (13 out of 28) and Group 3 (5 out of 28, with 2 - Popocatepetl and Etna - in the top-5 list) (Fig. 6). Two continental rift volcanoes (Nyiragongo and Nyamuragira) and two within-plate volcanoes (Kilauea and Erebus) also appear in our top-28 list of volcanic CO2 emitters (Fig. 6).

Our extrapolated global CO2 flux of 38.7 ± 2.9 Mt/yr is lower than previous global volcanic CO2 flux estimates in the literature, ranging from 66 to 540 Mt/yr (see ref.13 for a review). Several causes can explain this mismatch.

First, and most importantly, our global volcanic CO2 budget here only includes the contribution from the “strongly degassing volcanoes” that emit SO2 in quantities large enough to be detected from space (by OMI in this specific case28). We therefore admittedly do not take into consideration in our estimate the CO2 contribution from mildly degassing “magmatic” volcanoes (those still emitting SO2, but at levels too low to be resolved by satellites) and from “hydrothermal” volcanoes in which CO2 is emitted in combination with H2S (instead of SO2). Although typically exhibiting weaker surface gas manifestations, compared to the OMI-detected volcanoes characterised here, these magmatic-hydrothermal systems do often exhibit C-rich gas compositions38 (reflecting the extent/ mechanism of gas-water-rock reaction with meteoric fluids39), and do emit CO2 at the ~1000 tons/day level in the most extreme cases40, but most typically in the hundreds of tons/day range42. Considering that several hundreds of volcanoes worldwide are currently undergoing mild magmatic-hydrothermal degassing activity, this emission type could be responsible for the emission of several tens of Mt CO2/yr globally13,14. Also, we do not account for the CO2 output from volcanic lakes103, and diffuse/regional soil CO2 emissions around volcanic systems104, for which more data and alternative extrapolation approaches would be required. We therefore stress our results are not intended to represent total CO2 emissions from global subaerial volcanism, but rather the magmatic CO2 budget fraction contributed by the most actively degassing volcanoes on Earth.

Secondly, the mismatch in the estimated CO2 fluxes (this work and previous studies) derives (at least partially) from the distinct gas datasets used. We here specifically base our CO2 budget calculations on a consistent set of coincident (satellite-based) SO2 flux measurement, taken during a relatively short (decadal) period and with same retrieval/processing technique. In contrast, previous estimates have been hampered by the combination of sparse observations, taken over several decades, and with diverse observational/retrieval techniques. Even volcanoes which are persistently active alternate periods of elevated degassing with phases of reduced activity, and so non-coincident observations (taken over periods spanning several decades) may lead to biases. For example, by combining measurements taken between 1954 and 2011, a cumulative CO2 flux of 59.7 Mt/yr (from 33 measured volcanic gas plumes) was obtained11, or 2 times more than our mean 2005–2015 flux. We also explicitly use CO2/S02 information for high-temperature magmatic gases only, in contrast with previous efforts28 in which individual arc CO2 emissions have been quantified also considering low-temperature hydrothermal gas samples in which the C-rich composition is not representative of the strongly degassing “magmatic” arc systems. We also cannot rule out that part of the discrepancy is due to our Ba/La approach, which may only represents the sub-Moho magmatic CO2 flux, and not a potentially large recycled crustal CO2 flux. Finally, our “measured” CO2 dataset is extrapolated to the total number of “unmeasured” strongly degassing volcanoes by predicting, for each of them, the specific CO2/S02 ratio gas signature, rather than relying on the assumption that the global CO2 flux population obeys a specific statistical distribution (e.g., the power law distribution105).

Our results implicate that the arc volcano C flux (~ 8 ± 0.6 Mt C/yr) corresponds to a significant amount (~50%) of the subducted sedimentary carbonate (15 ± 2 Mt/yr; ref.106), but only a relatively small fraction (~21%) of the total C input at arc trenches (40–114 Mt C/yr; refs11,12). Thus, either the C input is balanced by “diffuse” C output forms, such as regional aquifers or soil degassing107 in the arc crust, or a substantial fraction of the subducted C is ultimately not erupted, but rather stored either in the lithospheric mantle10 or in the deep mantle12.

**Methods**

The SO2 flux compilation30 we rely on in this study includes a list of the 91 top-ranking volcanic SO2 degassing sources in 2005–2015 (Table 1). This set of consistent (identical retrieval/processing technique) and simultaneous (global) measurements has improved upon the shortcomings of previous catalogues19, which combined SO2 fluxes obtained with diverse techniques and in disparate temporal intervals (often differing by several decades).

These SO2 flux data are converted into CO2 fluxes by using either measured or predicted molar CO2/S02 ratios. For these strongly degassing volcanoes, S02 is assumed to correspond to SO2 throughout, since SO2 detection by satellites implies limited or no interaction with hydrothermal system (and thus trivial reduced S species, such as H2S).

**Measured volcanoes.** For 57 out of these 91 volcanic SO2 sources, we convert SO2 fluxes into CO2 fluxes, by pairing the former with the characteristic (mean) molar CO2/S02 ratios in the corresponding volcanic gases (Table 1). For arc volcanoes, we use the time-averaged molar CO2/S02 ratios compiled by (ref.98), integrated with novel gas information for eight new targets that have only recently been measured for the first time (see Table 1 for data provenance). Arc volcanoes are ranked in Groups (1 to 3) following the original categorization98. For non-arc volcanoes (here referred as Group 4), we average available volcanic gas information in the literature (see Table 1 for data sources). Note that, for both arc and non-arc, in cases where more than one volcano are listed
in the original dataset (e.g., Nyiragongo + Nyamuragira) due to insufficient spatial OMI resolution, we averaged the available volcanic gas information for the individual volcanoes, weighting each volcano's CO2/SO2 ratio by its ground-based S flux (where available) to obtain a combined CO2/SO2 ratio for the pair (see Table 1).

**Unmeasured volcanoes.** Thirty-four out of the 91 top-ranking volcanic SO2 sources have never been characterised for volcanic gas composition (Table 1). These include four of the top ten-ranking volcanic SO2 emitters (Bagana, Rabaul and Manam in Papua New Guinea, and Aoba in the Vanuatu archipelagos; Fig. 4). To indirectly infer the molar CO2/SO2 ratio gas signature of each of these 34 volcanoes, we used the averaged (mean) trace-element composition of the corresponding volcanic rocks. To this aim, as in earlier work, we extract trace-element information (Ba, La, Sr, Nd, U and Th whole-rock concentrations) either from the Earthchem data portal (http://www.earthchem.org), or from other sources (for volcanoes that do not appear on Earthchem) (see Table S1). Mafic to intermediate (<55% SiO2) rocks are only considered, same as in other work. From these, we calculate, for each volcano, the mean (±1 SD) of the Ba/La whole-rock ratios (Sr/Nd and U/Th ratios were also calculated; see Table S1). These ratios, in combination with the gas vs. whole-rock relationships illustrated in Figs 1–3, are finally used, to predict the characteristic volcanic gas CO2/SO2 ratio signature for each of the 34 “unmeasured” volcanoes.

The procedure is exemplified in Fig. 1 for the Pacaya volcano example. We select Pacaya because the recently obtained gas compositions can serve as a test of the methodology. The initial step involves establishing the relationship between CO2/SO2 gas ratios and whole-rock Ba/La ratios, using data for volcanoes for which both gas and trace element data are available (see Fig. 1; Table S1). The CO2/SO2 vs. Ba/La relationship can be established at the scale of individual arc segments (e.g., Figs 1 and 2), or for volcano Groups (Groups 1 or 2) (Fig. 3). For the Pacaya example, we rely on gas/whole-rock information for the well-characterised Central American Volcanic Arc (CAVA; Fig. 1).

Secondly, we use regression analysis to fit the gas vs. trace-element association via either a (i) linear or (ii) logarithm regression model (Fig. 1). We find that linear regression yields the best data fit in the majority of the cases (see the Pacaya example, Fig. 1a,b), and this regression model is used throughout unless indicated (see Table S1). We also find that data fitting is systemically optimised when the DMM composition is included in the fitting procedure (compare Fig. 1a,b), and this option is maintained throughout. Note, however, the method output (e.g., the outputted CO2/SO2 ratio) is poorly sensitive to this choice (see Table 2). Finally, the adopted regression model function (RM3 in the Pacaya example; Fig. 1 and Table 2) is used to calculate a “predicted” gas CO2/SO2 from available Ba/La information (Fig. 1). The confidence interval or delta, calculated from the regression line and one standard deviation about the regression, is taken as a proxy for the uncertainty in the predicted CO2/SO2 ratios. Uncertainty on the predicted ratios, as derived, incorporates (although indirectly) uncertainty/variability in “measured” gas CO2/SO2 ratios (average uncertainty at 1σ, ~26%) and whole-rock Ba/La ratios (average uncertainty at 1σ, ~16%) (see Table S1). In the specific Pacaya example (Fig. 1 and Table 2), our “predicted” gas CO2/SO2 ratio (1.4 ± 0.75) matches well the recently measured magmatic gas range (CO2/SO2 ratio of 1.1 ± 1.0). Our tests show that remarkably similar CO2/SO2 ratios (see Table 2) are obtained using other trace-element slab fluid tracers, such as the Sr/Nd ratio (Fig. 1c). We opt in the following for the Ba/La regression model because (i) Ba is more frequently available than Nd in the Earthchem dataset for the majority of the volcanoes, and (ii) use of the Sr/Nd ratio requires a priori knowledge of volcanic affinity for a specific Group (Group 1 and 2 typically exhibit diverse distributions in a CO2/SO2 vs. Sr/Nd scatter plot; see Fig. 1c). This latter information is frequently not a priori available (see below). The same procedure is applied to all unmeasured volcanoes (Table S1a), and the “predicted” ratios (assumed to correspond to CO2/SO2) are combined with SO2 flux results to ultimately infer the CO2 fluxes (Table 1).

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Author Contributions
A.A. conceived the idea. All of the authors participated in data collection and interpretation. A.A. drafted the manuscript with the help of all co-authors.

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