DATA REPOSITORY ITEM DR2008226

A highly detailed summary of sample preparation and analytical methods can be found in Barnes and Sharp (2006) and Sharp et al. (2007). Here we present the basic preparation and analytical procedures. Amendments to previous procedures due to differences in sample material are also discussed below.

*Chloride concentration*

Well-lithified sediments and serpentinite clasts were sonicated in 18MΩ deionized water to remove seawater and surficial contamination. Subsequent to rinsing to remove contamination, samples were dried, powdered to <100 mesh (<0.149 mm) and leached in 18MΩ deionized water for 24 hours to extract water-soluble chloride. Poorly-lithified serpentine mud and sediments samples were rinsed to remove contamination, crushed in the presence of 18MΩ deionized water, and then leached. Samples were filtered and the extracted solutions were analyzed for $Cl^-$ concentration on the Dionex DX-100 Ion Chromatograph (minimum detection limit for $Cl^-$ = 0.22 ppm). The leached powders were dried, pressed into a pellet, and analyzed for the $Cl^-$ concentration of the structurally bound chloride component on the Rigaku RIX 2100 wavelength dispersive X-ray spectrometer (minimum detection limit for $Cl^-$ = 10 ppm). Note that one sediment sample (185-1149A-7H-1, Table 1) was washed 15 times for 15 minutes each time, instead of the standard 4 times, in an ultra-sonic bath to check if all water-soluble chloride was removed and the structurally bound chloride measurement did not reflect a water-soluble chloride component. No difference in the $\delta^{37}Cl$ value was obtained using this extreme rinsing procedure indicating that our normal leaching and filtering procedure effectively removes all water-soluble chloride.

*Stable chlorine isotope analyses*

$Cl^-$ was released from powdered rock/ash samples by pyrohydrolysis and collected in an aqueous solution (Magenheim et al., 1994; Barnes and Sharp, 2006). Once in an aqueous form $Cl^-$ is converted to AgCl and reacted with $CH_3I$ to produce $CH_3Cl$ (Eggenkamp, 1994). $CH_3Cl$ is purified on a gas chromatographic column and introduced into a gas source mass spectrometer (Finnigan Delta XL Plus) (Barnes and Sharp, 2006). Volcanic gases trapped in Giggenbach bottles were prepared following the method of Eggenkamp (1994). In order to ensure removal of the high concentration of sulfur prior to addition of $AgNO_3$ in volcanic materials and some sediment samples, samples were reacted with 10 mL 50% nitric acid at low heat for 2 hours under a watch glass filled with 18MΩ deionized water to drive off excess sulfur as $SO_2$. Samples with high concentrations of $Cl^-$ were analyzed using the dual inlet method and have an error of ±0.12‰; samples with low concentrations of $Cl^-$ were analyzed using the continuous flow method and have an error of ±0.37‰ based on the long-term average of laboratory samples.

ONLINE SUPPLEMENTAL REFERENCES

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TABLE DR1. CHLORINE CONCENTRATION AND ISOTOPIC DATA FOR SEDIMENTS

| Sample (Leg-Hole-Core(type)-Section, Interval(cm)) | Lithology* | Depth (mbsf) | $Cl_{WSC}$ (wt%) | $Cl_{SBC}$ (ppm) | $\delta^{37}Cl_{WSC}$[†] (‰) | $\delta^{37}Cl_{SBC}$[†] (‰) |
|---|---|---|---|---|---|---|
| Seafloor sediments | | | | | | |
| 129-800A-4R-3, 16-20 | pelagic clay | 23.36 | 1.11 | 115 | -0.29 | *0.26*[§] |
| | | | | | | *0.32* |
| 129-800A-53R-1, 14-21 | clayey radiolarite | 465.04 | 0.36 | 246 | -1.13 | -1.93 |
| | | | | | -1.06 | |
| | | | | | -0.97 | |
| 129-801A-7R-1, 101-106 | pelagic clay with zeolites | 60.01 | 1.26 | 529 | -0.66 | -1.51 |
| 129-801B-1R-5, 18-22 | volcanoclastic sandstone/silty claystone | 200.18 | 0.61 | N.D.[#] | -0.78 | N.D. |
| 129-801B-33R-1, 1-7 | radiolarite/claystone | 442.81 | 0.35 | 365 | -1.26 | -2.51 |
| | | | | | -1.07 | |
| 129-802A-3R-1, 15-20 | tuff and pelagic clay | 14.75 | 0.39 | 872 | -0.31 | -0.86 |
| 129-802A-38R-1, 10-15 | zeolitic pelagic claystone | 339.30 | 0.54 | 151 | -1.01 | *-0.65* |
| 129-802A-42R-2, 11-16 | volcanoclastic turbidite/claystone | 374.29 | 0.35 | 204 | -1.57 | -0.65 |
| 185-1149A-7H-1, 16-20 | ash-bearing siliceous clay | 51.86 | 1.89 | 624 | -0.43 | -0.59 |
| | | | | | | -0.81** |
| 185-1149A-18H-1, 16-20 | clay | 156.36 | 0.87 | 1240 | -0.92 | -1.35 |
| 185-1149B-3R-1, 10-12 | clay | 170.40 | 1.15 | 514 | -0.54 | -0.75 |
| | | | | | | -0.36 |
| 185-1149B-20R-1, 17-22 | radiolarian chert/porcellanite/marlstone | 321.07 | 0.37 | 36 | -0.93 | *-1.11* |
| Sediments overlying Torishima seamount | | | | | | |
| 125-783A-4R-4, 10-12 | glass & nannofossil-rich silty clay | 30.60 | 1.82 | 544 | -0.26 | -1.47 |
| 125-784A-16R-1, 10-12 | glass-rich claystone | 136.10 | 1.75 | 440 | -0.29 | -0.71 |

*Lithology descriptions from Fryer et al., 1990, Lancelot et al., 1990, and Plank et al., 2000.

[†]WSC = water-soluble chloride. SBC = structurally bound chloride.

[§]Italicized data were determined using the continuous flow method (error = ± 0.37‰). Error on all other analyses = ± 0.12‰.

[#]N.D. = not determined. Sample lost during the filtering process.

**Repeat analysis on sample that was washed in 18MΩ deionized water 15 times.

TABLE DR2. CHLORINE CONCENTRATION AND ISOTOPIC DATA FOR SERPENTINE SEAMOUNT MATERIAL

| Sample (Leg-Hole-Core(type)-Section, Interval(cm)) | Lithology* | Depth (mbsf) | $Cl_{WSC}$ (wt%) | $Cl_{SBC}$ (ppm) | $\delta^{37}Cl_{WSC}$ (‰) | $\delta^{37}Cl_{SBC}$ (‰) |
|---|---|---|---|---|---|---|
| Conical seamount | | | | | | |
| Serpentine mud | | | | | | |
| 125-779A-2R-2, 74-76 | aragonite-bearing sandy silt-sized serpentine | 3.34 | 1.16 | 326 | 0.05 | 0.24 |
| 125-780B-1R-2, 130-132 | silty sand-sized serpentine | 2.80 | 1.06 | 319 | 0.13 | 0.15 |
| 125-780C-2R-1, 28-30 | silty sand-sized serpentine | 5.78 | 0.70 | 261 | 0.20 | 0.45 |
| Serpentinite clast | | | | | | |
| 125-779A-3R-CC, 9-13 | serpentinized harzburgite (85%) | 10.69 | 0.09 | 426 | 0.26 | 0.64 |
| | | | | | 0.16 | |
| 125-779A-8R-1, 20-23 | serpentinized harzburgite (100%) | 58.70 | 0.14 | 147 | 1.02 | -0.22 |
| | | | | | 0.15 | |
| | | | | | 0.68 | |
| 125-779A-11R-1, 8-12 | serpentinized harzburgite (90-99%) | 87.38 | 0.15 | 341 | -0.03 | 0.28 |
| | | | | | | 0.49 |
| 125-779A-15R-2, 22-27 | serpentinized dunite (50-80%) | 127.62 | 0.15 | 238 | 0.50 | 0.38 |
| 125-779A-20R-1, 2-5 | serpentinized harzburgite (90-99%) | 169.12 | 0.06 | 253 | -0.10 | 0.12 |
| | | | | | -0.07 | |
| 125-779A-24R-1, 34-38 | serpentinized dunite (100%) | 187.54 | 0.18 | 221 | 0.36 | 0.36 |
| 125-779A-28R-3, 100-104 | serpentinized dunite (100%) | 228.97 | 0.24 | 320 | 0.20 | 0.02 |
| 125-779A-32R-1, 27-30 | serpentinized dunite (100%) | 264.67 | 0.07 | 225 | 0.62 | 0.30 |
| | | | | | 0.44 | |
| 125-779A-35R-1, 75-78 | serpentinized harzburgite (70%) | 294.05 | 0.08 | 315 | 0.37 | 0.47 |
| | | | | | 0.42 | |
| South Chamorro seamount | | | | | | |
| Serpentine mud | | | | | | |
| 195-1200D-2H-1, 85-87 | silty clay serpentine | 7.75 | 0.58 | 511 | 0.09 | 0.57 |
| 195-1200D-8H-1, 110-112 | silty clay serpentine | 26.10 | 0.60 | 262 | 0.07 | 0.13 |
| Serpentinite clast | | | | | | |
| 195-1200A-2R-1, 0-4 | serpentinized harzburgite (99%) | 9.70 | 0.15 | 177 | 0.21 | 1.61 |
| 195-1200A-6R-1, 4-7 | serpentinized harzburgite (75-85%) | 41.44 | 0.13 | 420 | 0.17 | 0.79 |
| 195-1200A-10R-1, 34-36 | serpentinized harzburgite (99%) | 80.24 | 0.07 | 402 | 0.03 | 0.51 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 195-1200A-13R-1, 46-48 | serpentinized harzburgite (98%) | 109.16 | 0.03 | 398 | -0.27<br>-0.37 | 0.59 |
| 195-1200A-13R-2,16-19 | serpentinized harzburgite (80-99%) | 110.11 | 0.15 | 350 | 0.08 | 0.52<br>0.36 |
| 195-1200A-16R-1, 50-53 | serpentinized harzburgite (99%) | 138.10 | 0.15 | 390 | 0.16 | 1.54 |
| 195-1200A-17G-2, 46-48 | serpentinite (99%) | N.R. | 0.09 | 365 | 0.23 | 0.38 |
| | | | | | | |
| <u>Torishima seamount</u> | | | | | | |
| <u>Serpentine mud</u> | | | | | | |
| 125-783A-15R-1, 12-14 | silt-sized serpentine | 129.72 | 0.24 | 409 | -0.04 | N.D. |
| <u>Serpentinite clast</u> | | | | | | |
| 125-783A-18R-1, 99-101 | serpentinized tectonized harzburgite (70-90%) | 159.59 | 0.07 | 745 | -0.13 | -0.03 |
| 125-784A-36R-1, 90-92 | serpentinized harzburgite (80-90%) | 329.90 | 0.44 | 5194<br>5298 | 0.14 | 0.49 |
| 125-784A-45R-1, 89-91 | serpentinized tectonized harzburgite (80-99%) | 416.49 | 0.09 | 611 | 0.66<br>0.60 | 0.29 |

*Note:* Extremely high $\delta^{37}Cl$ values measured for serpentinite samples from the same seamounts (Wei et al., 2008) are most likely erroneous, having been analyzed with a technique that is known to have serious analytical artifacts (Bonifacie et al., 2007; Sharp et al., 2007).

*Lithology descriptions from Leg 195 Initial Reports, Core Descriptions. Percentage serpentinization noted in parentheses.

†N.R. = not reported. Sample from a ghost core (recovered interval of no new penetration because of hole collapse or fill).

§N.D. = not determined.

TABLE DR3. CHLORINE ISOTOPE DATA FOR VOLCANIC MATERIALS

| Sample | Island | Latitude/Longitude | Description | T (ºC) | $\delta^{37}Cl_{bulk}$ (‰) |
|---|---|---|---|---|---|
| <u>Volcanic gas and geothermal well samples</u> | | | | | |
| Sou-1 | Hakone | 35º14.492'N/139º01.925'E | volcanic gas | 96.1 | _-5.41*_ |
| | | | | | *-3.18* |
| Sou-2 | Hakone | 35º14.492'N/139º01.925'E | volcanic gas | 96.1 | *-2.52* |
| Owa-1 | Hakone | 35º14.488'N/139º01.230'E | volcanic gas | 96.5 | *-0.94* |
| Noh-1 | Hakone | 35º13.442'N/139º01.948'E | volcanic gas | 95.8 | *-2.70* |
| Uto-1 | Oshima | 34º45.243'N/139º24.125'E | volcanic gas | 81.0 | *-2.04* |
| Jin-1 | Shikinejima | 34º19.128'N/139º12.882'E | volcanic gas | 66.2 | -0.94 |
| Geo-1 | Hachijojima | 33º04.494'N/139º48.742'E | geothermal well | 170.8 | *-2.25* |
| Aog-2 | Aogoshima | 32º27.136'N/139º45.735'E | volcanic gas | 99.2 | *-1.44* |
| Aog-3 | Aogoshima | 32º27.435'N/139º45.717'E | volcanic gas | 99.4 | *-1.41* |
| MAR05-4 | Uracas | 20º32.748'N/144º53.475'E | volcanic gas | 144.0 | *-0.80* |
| | | | | | *-0.06* |
| MAR05-5 | Uracas | 20º32.748'N/144º53.475'E | volcanic gas | 144.0 | *2.59*† |
| | | | | | *-0.37* |
| | | | | | *-0.20* |
| MAR7 | Agrigan | 18º46.694'N/145º40.282'E | volcanic gas | 98.0 | *-1.93* |
| MAR3 | Pagan | 18º04.265'N/145º40.498'E | volcanic gas | 96.1 | *-1.72* |
| | | | | | |
| <u>Volcanic ash and lava samples</u> | | | | | |
| Osh-02 | Oshima | 34º47.825'N/139º21.652'E | beach sand | | -0.46 |
| Mih-1 | Oshima | 34º43.987'N/139º23.135'E | lava | | *-0.28* |
| Niij-4 | Niijima | 34º25.517'N/139º16.901'E | pyroclastic surge deposit | | -0.77 |
| Hac-09 | Hachijojima | 33º04.756'N/139º51.444'E | scoria and ash | | -0.22 |
| Hac-03 | Hachijojima | 33º06.154'N/139º47.128'E | ash | | -1.05 |
| Aog-6 | Aogoshima | 32º27.992'N/139º45.303'E | scoria and ash | | -0.57 |
| Urac05 | Uracas | 20º32.522'N/144º53.993'E | ash | | -1.46 |
| 04-Maug-03 | Maug | 20º1.487'N/145º13.892'E | beach sand | | *0.01* |
| Asun-20-02 | Asuncion | 19º41.339'N/145º23.440'E | ash | | *-0.47* |
| Agri-19-02 | Agrigan | 18º44.352'N/145º41.193'E | scoria/lapilli | | -0.10 |
| 04-Pag-11 | Pagan | 18º9.193'N/145º46.576'E | olivine-plagioclase clasts | | *-2.56* |

| | | | | |
|---|---|---|---|---|
| | | | | *-1.77* |
| Ala-02 | Alamagan | 17º35.183'N/145º50.285'E | scoria | *-0.23* |
| 04-Gug-11 | Guguan | 17º18.008'N/145º50.308'E | scoria | *-0.76* |
| | | | | -0.16 |
| Sari-15-02 | Sarigan | 16º42.371'N/145º46.026'E | scoria | -1.11 |
| 04-Ana-04-3 | Anatahan | 16º20.358'N/145º38.426'E | scoria | 0.01 |
| Anat05-01 | Anatahan | 16º20.409'N/145º41.748'E | ash | *0.38* |
| | | | | |
| Basalt samples from the Guguan cross-chain | | | | |
| D50-1-1 | | | | 0.47 |
| D48-1-3 | | | | 0.02 |
| D46-1-5 | | | | 0.19 |

*Note:* When two analyses for the same sample were determined only the average value was plotted.

*Italicized data were determined using the continuous flow method.

†Outlier and not included in average calculations (Fig.3).