*Applied Geochemistry*

**Zinc isotope fractionation during the sorption of Zn to minerals and organic matter in sediment cores affected by anthropogenic pollution**

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Table S1. δ66Zn values with 2σ and Zn concentrations of the chemical fractions, the sum of fractions and the bulk sediments from the Osaka Bay and Lika Biwa core. The low Zn concentrations of the exchangeable fractions (< 3 %) are not shown. Bulk sediment data have been previously reported (Nitzsche et al., 2022, 2021).

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Year | Znacid #1 | Znacid #2 | Znred | Znox | Znres | Znsum | Znbulk | δ66Znacid #1 | δ66Znacid #2 | δ66Znred | δ66Znox | δ66Znres | δ66Znsum | δ66Znbulk |
| (AD) | (µg g-1) | (‰) |
| Osaka Bay |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| 1995 | 29 | 22 | 43 | 20 | 83 | 197 | 200 | +0.73±0.01 | +0.41±0.02 | +0.04±0.03 | -0.03±0.04 | +0.15±0.02 | +0.22±0.02 | +0.22±0.04 |
| 1991\* | 31 | 21 | 43 | 21 | 90 | 206 | 223 |   |  |  |  |  |  | +0.22±0.03 |
| 1988\* | 35 | 21 | 43 | 15 | 90 | 205 | 208 |   |  |  |  |  |  | +0.22±0.02 |
| 1984 | 38 | 22 | 43 | 22 | 84 | 208 | 211 | +0.72±0.03 | +0.34±0.03 | +0.02±0.03 | -0.14±0.04 | +0.18±0.01 | +0.23±0.02 | +0.19±0.02 |
| 1980\* | 33 | 24 | 46 | 19 | 84 | 207 | 204 |   |   |   |   |   |  | +0.22±0.02 |
| 1976\* | 42 | 25 | 50 | 22 | 79 | 218 | 220 |   |   |   |   |   |  | +0.20±0.03 |
| 1972 | 52 | 30 | 55 | 24 | 75 | 236 | 251 | +0.64±0.04 | +0.37±0.02 | +0.05±0.04 | -0.06±0.02 | +0.18±0.00 | +0.25±0.02 | +0.22±0.07 |
| 1968\* | 53 | 29 | 55 | 23 | 83 | 244 | 245 |  |  |  |  |  |  | +0.23±0.03 |
| 1964\* | 59 | 36 | 65 | 25 | 84 | 270 | 270 |  |  |  |  |  |  | +0.22±0.02 |
| 1960 | 58 | 36 | 70 | 25 | 83 | 272 | 289 | +0.69±0.01 | +0.40±0.01 | +0.10±0.04 | -0.04±0.01 | +0.14±0.01 | +0.27±0.02 | +0.23±0.03 |
| 1956\* | 52 | 29 | 55 | 23 | 85 | 245 | 256 |  |  |  |  |  |  | +0.22±0.03 |
| 1952\* | 36 | 24 | 49 | 21 | 84 | 214 | 211 |  |  |  |  |  |  | +0.24±0.04 |
| 1949 | 31 | 22 | 45 | 16 | 80 | 194 | 211 | +0.76±0.01 | +0.41±0.02 | +0.08±0.01 | +0.03±0.02 | +0.26±0.00 | +0.28±0.02 | +0.24±0.03 |
| 1945\* | 29 | 21 | 42 | 19 | 83 | 193 | 194 |  |  |  |  |  |  | +0.26±0.02 |
| 1941\* | 32 | 23 | 45 | 20 | 78 | 197 | 204 |  |  |  |  |  |  | +0.24±0.01 |
| 1937 | 17 | 13 | 28 | 15 | 75 | 147 | 148 | +0.82±0.00 | +0.44±0.03 | +0.10±0.03 | +0.06±0.02 | +0.23±0.01 | +0.27±0.02 | +0.26±0.02 |
| 1933\* | 12 | 11 | 25 | 14 | 75 | 138 | 142 |  |  |  |  |  |  |  |
| 1924\* | 13 | 11 | 25 | 14 | 71 | 133 | 141 |  |  |  |  |  |  | +0.25±0.02 |
| 1918\* | 10 | 8 | 22 | 13 | 76 | 130 | 130 |  |  |  |  |  |  | +0.25±0.02 |
| 1911\* | 11 | 9 | 22 | 14 | 75 | 130 | 135 |  |  |  |  |  |  | +0.26±0.02 |
| 1904 | 8 | 7 | 19 | 13 | 77 | 123 | 125 | +0.81±0.02 | +0.37±0.02 | +0.080.02 | +0.10±0.02 | +0.25±0.03 | +0.25±0.02 | +0.26±0.01 |
| 1898\* | 6 | 5 | 14 | 11 | 72 | 108 | 114 |  |  |  |  |  |  | +0.26±0.01 |
| 1891\* | 8 | 6 | 18 | 13 | 72 | 116 | 118 |  |  |  |  |  |  |  |
| 1884\* | 7 | 4 | 14 | 12 | 73 | 110 | 110 |  |  |  |  |  |  | +0.27±0.02 |
| 1878\* | 7 | 6 | 15 | 12 | 72 | 111 | 118 |  |  |  |  |  |  | +0.26±0.03 |
| 1871\* | 4 | 4 | 15 | 11 | 73 | 107 | 111 |  |  |  |  |  |  | +0.27±0.02 |
| 1864 | 4 | 3 | 13 | 10 | 65 | 95 | 107 |   |   | +0.04±0.04 | +0.15±0.01 | +0.25±0.04 |  | +0.29±0.02 |
| 1824 | 4 | 4 | 13 | 10 | 63 | 95 | 105 | +0.87±0.03 | +0.23±0.02 | +0.06±0.01 |   | +0.26±0.02 |  | +0.32±0.03 |
| 1763 | 4 | 3 | 13 | 11 | 64 | 95 | 107 | +0.85±0.02 |   | +0.04±0.03 |   | +0.29±0.03 |  | +0.27±0.03 |
| 1707 | 4 | 3 | 12 | 11 | 63 | 93 | 104 | +0.87±0.03 |   | -0.05±0.01 | +0.12±0.02 | +0.27±0.00 |  | +0.31±0.02 |
|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| Lake Biwa |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| 1989 | 21 |  | 33 | 12 | 107 | 176 | 169 |   |  |   |   |   |   |  |
| 1980 | 20 |  | 27 | 15 | 102 | 164 | 168 | +0.85±0.02 |  | +0.21±0.04 | +0.25±0.02 | +0.10±0.03 | +0.22±0.02 | +0.24±0.03 |
| 1974 | 18 |  | 25 | 18 | 105 | 166 | 188 | +0.83±0.03 |  | +0.19±0.02 | +0.24±0.03 | +0.12±0.02 | +0.22±0.02 | +0.24±0.02 |
| 1969\* | 16 |  | 21 | 18 | 114 | 169 | 169 | +0.92±0.03 |  | +0.22±0.02 | +0.28±0.02 | +0.17±0.02 | +0.26±0.02 | +0.23±0.02 |
| 1956\* | 13 |  | 16 | 20 | 112 | 162 | 196 |   |  |   |   |   |   | +0.24±0.03 |
| 1952 | 11 |  | 15 | 16 | 105 | 147 | 168 |   |  |   |   |   |   | +0.25±0.01 |
| 1947 | 11 |  | 14 | 18 | 118 | 162 | 165 | +1.00±0.00 |  | +0.35±0.03 |   | +0.18±0.01 |   | +0.24±0.02 |
| 1943\* | 11 |  | 14 | 18 | 117 | 160 | 163 |   |  |   |   |   |   | +0.27±0.01 |
| 1939 | 11 |  | 19 | 18 | 106 | 153 | 158 | +0.97±0.04 |  | +0.25±0.02 | +0.25±0.03 | +0.17±0.03 | +0.24±0.02 | +0.26±0.01 |
| 1935\* | 8 |  | 14 | 15 | 93 | 131 | 135 |   |  |   |   |   |   | +0.27±0.04 |
| 1931 | 7 |  | 11 | 12 | 97 | 127 | 133 |   |  |   |   | +0.19±0.03 |   | +0.26±0.04 |
| 1914\* |  |  |  |  |  |  | 130 |   |  |   |   |   |   | +0.29±0.05 |
| 1901 | 4 |  | 7 | 8 | 93 | 113 | 121 |   |  |   |   | +0.26±0.01 |   | +0.28±0.03 |
| 1886\* | 4 |  | 7 | 10 | 95 | 116 | 116 |   |  |   |   |   |   | +0.29±0.03 |
| 1870\* | 5 |  | 7 | 9 | 92 | 113 | 120 |   |  |   |   |   |   | +0.29±0.04 |
| 1857 | 4 |  | 7 | 8 | 87 | 108 | 120 |   |  |   |  | +0.19±0.02 |   | +0.31±0.03 |

\*Only bulk sediments analyzed for Zn stable isotope ratios



Figure S1. Scatter plot of the calculated δ66Zn value of the sum of fractions against the measured δ66Zn value of the bulk sediment in the Osaka Bay and Lake Biwa cores.



Figure S2. Temporal variation in δ66Zn values of the mixture of the acid-soluble fraction #2 and the reducible fraction (red circles) in the Osaka Bay core. The bulk sediment is shown as reference (black circles). The δ66Zn values of the mixture of the acid-soluble fraction #2 and the reducible fraction was calculated as δ66Znmix = (δ66Znacid-soluble #2 · *f*acid-soluble #2 +δ66Znreducible · *f*reducible) / (*f*acid-soluble #2 + *f*reducible).



Figure S3. Depth profiles of the molar Sr/Ca (x1000) and Ba/Ca (x1000) ratios of the two acid-soluble fractions in the Osaka Bay core.