



Supplement of

Pb and Fe flow through the mire-lake complex of Skogaryd catchment – a system under anthropogenic influence

Jonas Thomsen et al.

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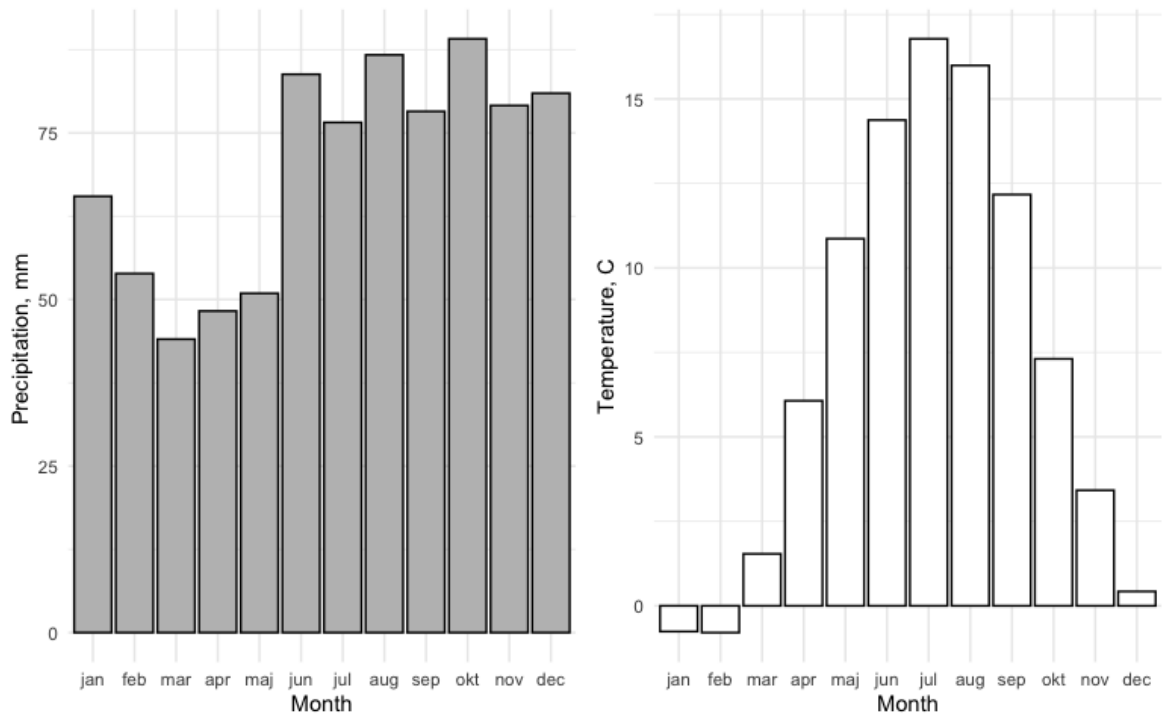


Fig S1. Monthly total precipitation and average temperature for the climate normal period 1991-2020 at the meteorological station in Vänersborg ~10km east of Mycklemossen (Swedish Meteorological and Hydrological Institute).

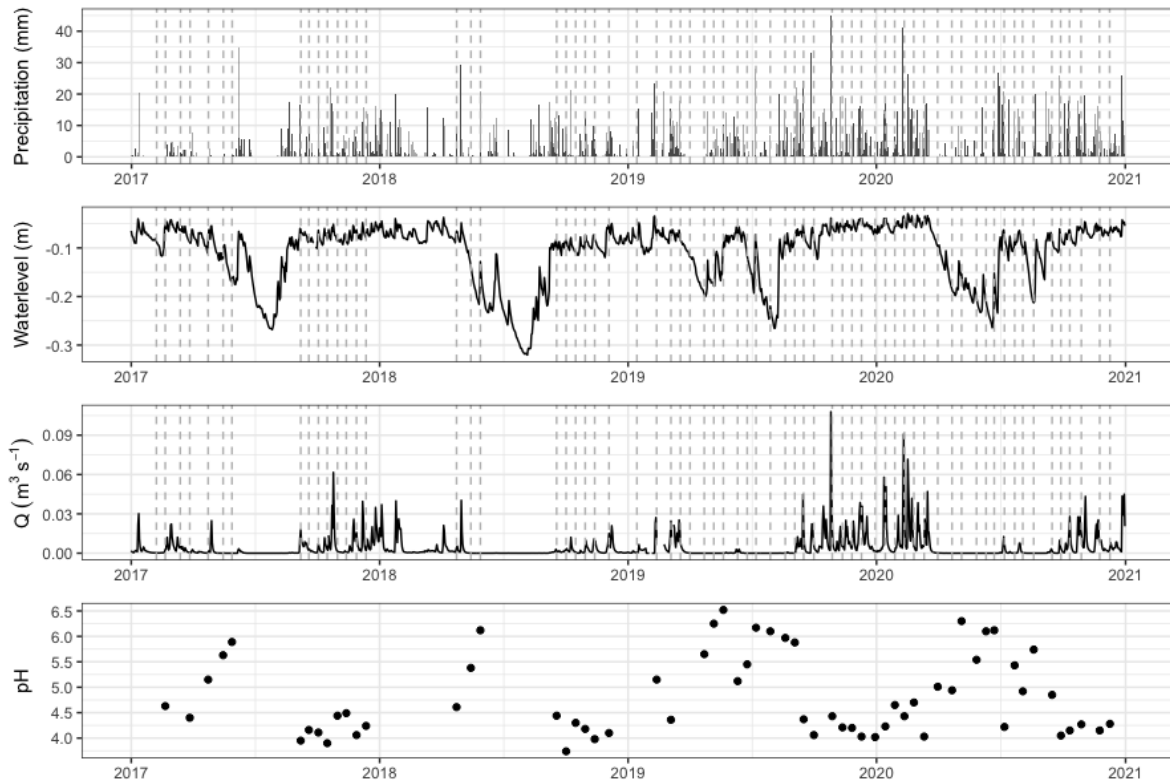


Fig S2. Daily total precipitation and average water table in Mycklemossen and average discharge and pH in stream water from Mycklemosse. Vertical dotted lines mark grab sampling events in stream water (S1) from Mycklemossen where pH (and DOC, metal concentrations) was measured.

Table S1. Catchment area, discharge and average annual export \pm SD of DOC, Fe and Pb in the period of 2017-2020 from Mycklemossen (S1) and Ersjön (S6).

| | Station 1 | Station 6 | St1 / St6 |
|---|----------------------|-----------------------|-----------|
| Catchment area (km ²) | 0.595 | 1.337 | 0.44 |
| Annual discharge (m ³ yr ⁻¹) | 136 155 \pm 35 168 | 412 878 \pm 181 006 | 0.33 |
| DOC export (kg yr ⁻¹) | 5834 \pm 1674 | 12616 \pm 9293 | 0.46 |
| Fe export (kg yr ⁻¹) | 151 \pm 39 | 325 \pm 232 | 0.46 |
| Pb export (kg yr ⁻¹) | 0.705 \pm 0.193 | 0.681 \pm 0.520 | 1.04 |
| of which from St. 1 | | 84.2 \pm 8.1% | |
| | | = 0.573 \pm 0.441* | |

Table S2. Average element concentration (mg/kg) in peat cores of Mycklemossen mire at selected depth intervals (n=5) in 3 micro topography types (Type): Hollow (HOL), hummock (HUM) and an intermediate form (INT).

| Element | Type | 15-20 cm | | 20-25 cm | | 25-50 cm | | 120-125 cm | | 420-425 cm | |
|---------|------|----------|----------|----------|----------|----------|----------|------------|----------|------------|-----------|
| | | mean | se | mean | se | mean | se | mean | se | mean | se |
| Lead | HOL | 32.21 | ± 8.64 | 22.17 | ± 5.79 | 14.89 | ± 4.65 | 4.75 | ± 2.04 | 1.75 | ± 0.53 |
| | HUM | 28.26 | ± 10.80 | 46.49 | ± 18.89 | 91.80 | ± 33.71 | 7.60 | ± 2.45 | 3.07 | ± 1.76 |
| | INT | 64.25 | ± 17.23 | 60.38 | ± 26.08 | 58.50 | ± 27.79 | 5.19 | ± 1.14 | 3.15 | ± 1.20 |
| Iron | HOL | 606.23 | ± 119.63 | 641.77 | ± 152.07 | 610.21 | ± 132.26 | 1433.99 | ± 247.94 | 7147.57 | ± 1017.80 |
| | HUM | 1237.04 | ± 423.68 | 1532.24 | ± 568.03 | 1757.24 | ± 435.70 | 1413.89 | ± 622.78 | 8481.66 | ± 2261.55 |
| | INT | 1130.62 | ± 399.66 | 856.25 | ± 222.24 | 817.74 | ± 237.26 | 1476.65 | ± 599.08 | 7457.85 | ± 768.39 |

Table S3: ICP-MS parameters for determination of Pb isotopic analysis.

| | |
|--|--|
| Laboratory and Sample Preparation | |
| Laboratory name | Microgeochemistry Laboratory, University of Gothenburg |
| Sample type | surface water samples |
| Sample preparation | acidified to 2% HNO ₃ |
| Measurement session | 240912 |
| Autosampler and Sample Uptake | |
| Model and type | ASX-500 |
| Uptake speed (Nebulizer Pump) [rps] | 0.1 |
| ICP-MS Instrument | |
| Make, Model and type | Agilent 8800QQQ |
| Sample introduction | MicoMist Nebulizer |
| Spraychamber Temp [C] | 2 |
| Plasma | |
| RF power [W] | 1550 |
| RF matching [V] | 1.8 |
| Sample depth [mm] | 8 |
| Nebulizer gas flow [l/min] | 1.07 |
| Lenses | |
| Extract 1 [V] | 4.1 |
| Extract 2 [V] | -200 |
| Omega bias [V] | -95 |
| Omega lens [V] | 7.8 |
| Q1 entrance [V] | 2 |
| Q1 exit [V] | -2 |
| Cell focus [V] | 1 |
| Cell entrance [V] | -81 |
| Cell exit [V] | -85 |
| Deflect [V] | 5.8 |
| Plate bias [V] | -71 |
| Q1 | |
| Q1 bias [V] | -2 |
| Q1 prefilter bias [V] | -32 |
| Q1 postfilter bias [V] | -22 |
| Cell | |
| reaction gas | N ₂ O |

| | |
|---------------------------|------|
| reaction gas flow | 22% |
| OctP bias [V] | -5.1 |
| OctP RF [V] | 200 |
| Energy discrimination [V] | -5.2 |

| | |
|---------|-------|
| Q2 | |
| QP Bias | -10.3 |

Table S4: ICP-MS acquisition parameters for Pb isotopic analysis

| | |
|------------------|----------|
| Acq Mode | Spectrum |
| Q2 Peak Pattern | 3 points |
| Replicates | 10 |
| Sweeps/Replicate | 250 |

| | |
|--------------------|------|
| Stabilization time | 15 s |
| Wait Time Offset | 2 ms |

| Element | Q1 (m/z) | Q2 (m/z) | Dwell time (s) |
|----------------|--------------------|--------------------|--------------------------|
| Hg | 202 | 202 | 1.5 |
| Pb+Hg | 204 | 204 | 20 |
| Pb | 206 | 206 | 3.0 |
| Pb | 207 | 207 | 3.0 |
| Pb | 208 | 208 | 1.5 |

PeriPump Settings**Pre Run**

| | | | |
|-------------|----|---|------------------|
| Uptake time | 40 | s | sample (0.5 rps) |
| Stabilize | 30 | s | sample (0.5 rps) |

Post Run

| | | | |
|-------------|----|---|---------------------------------|
| Rise Port | 10 | s | 0.5% HNO ₃ (0.1 rps) |
| Rise Vial 1 | 30 | s | 2 % HNO ₃ (0.1 rps) |
| Rise Vial 1 | 30 | s | 2 % HNO ₃ (0.5 rps) |
| Rise Vial 1 | 40 | s | 2 % HNO ₃ (0.5 rps) |

Table S5. Reference material for ICP-MS

| | | | 207Pb | 2s2 | 208Pb | 2s2 | 206P | 2s2 | 207Pb | 2s2 | 208Pb | 2s2 |
|--------------------------------------|--|-----------------------------|--------------|------------|--------------|------------|--------------|------------|--------------|------------|--------------|------------|
| | | | /206P | | /206P | | b/204 | | /204P | | /204P | |
| | | | b | | b | | Pb | | b | | b | |
| Primary reference material | | | | | | | | | | | | |
| NIST SRM 981 | <i>Catanzaro et al (1968)</i> | | 0.9146 4 | 0.00033 | 2.1681 | 0.0008 | 16.93 7 | 0.011 | 15.49 | 0.01 | 36.72 | 0.03 |
| Secondary reference solutions | | | | | | | | | | | | |
| NIST SRM 981 | approximately 5.5 µg/l Pb | weighted mean MSWD n= | 0.9142 | 0.0001 | 2.1667 | 0.0016 | 16.93 8 | 0.013 | 15.48 4 | 0.018 | 36.626 | 0.066 |
| | | | 0.13 | | 1.3 | | 0.41 | | 0.33 | | 3.5 | |
| | | | 5/5 | | 5/5 | | 5/5 | | 5/5 | | 5/5 | |
| NIST SRM 981 | approximately 1.1 µg/l Pb | weighted mean MSWD n= | 0.9179 | 0.0017 | 2.1723 | 0.0036 | 16.90 7 | 0.029 | 15.53 1 | 0.033 | 36.676 | 0.076 |
| | | | 0.49 | | 0.87 | | 0.57 | | 1.14 | | 1.7 | |
| | | | 5/5 | | 5/5 | | 5/5 | | 5/5 | | 5/5 | |
| AQUA-1 | <i>Yeghicheyan et al. (2021)</i> this study | weighted mean MSWD n= | 0.8990 | 0.301 | 2.144 | 0.109 | 17.17 1 | 0.008 | 15.53 5 | 0.041 | 37.03 | 0.237 |
| | | | 0.955 | 0.001 | 2.148 | 0.003 | 17.15 8 | 0.032 | 15.53 3 | 0.028 | 36.799 | 0.066 |
| | | | 0.4 | | 0.19 | | 0.99 | | 0.51 | | 0.48 | |
| | | | 5/5 | | 5/5 | | 5/5 | | 5/5 | | 5/5 | |

Table S6. ^{14}C dating of peat cores sampled at location T3. Peat samples were collected in 20-25, 120-125 and 420-25 cm depth from hummock, intermediate and hollow micro topographies. Values are means of three technical replicates \pm SD. Bomb peak (BP) age is 1950.

| Topography | Depth (cm) | Age (yrs BP-age) | Date of origin |
|---------------------|-------------------|-------------------------|-----------------------|
| Hummock | 20-25 | NA* | NA* |
| | 120-125 | 875 \pm 28 | 1075 |
| | 425-450 | 3 526 \pm 30 | 1576 (BC) |
| Intermediate | 20-25 | 59 \pm 28 | 1891 |
| | 120-125 | 1 839 \pm 29 | 111 |
| | 425-450 | 3 927 \pm 31 | 1977 (BC) |
| Hollow | 20-25 | 256 \pm 28 | 1694 |
| | 120-125 | 1 105 \pm 29 | 845 |
| | 420-425 | 3 199 \pm 30 | 1249 (BC) |

* Sample contained too much modern C for accurate dating of sample age.

Table S7. Output from mixed effects models testing the effects of sampling depth and topography type and their interactions on peat bulk density, pore water pH, SOM%, C content, N content, C to N ratio, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. There was no effect of depth and type on $\delta^{18}\text{O}$ and the results are not shown. $p < 0.05$ are marked in bold.

| | Bulk density ¹ | | | pH | | | SOM ¹ | | | C% ¹ | | |
|------------|---------------------------|----------------|------------------|----|----------------|------------------|------------------|----------------|------------------|-----------------|----------------|------------------|
| | Df | X ² | p | Df | X ² | p | Df | X ² | p | Df | X ² | p |
| Depth | 2 | 18.6 | <0.001 | 1 | .3 | <0.001 | 2 | 27.9 | <0.001 | 1 | 23.6 | <0.001 |
| Type | 2 | 1.0 | 0.59 | 2 | 4.3 | 0.12 | 2 | 6.7 | 0.04 | 2 | 0.7 | 0.70 |
| Depth*Type | 4 | 0.8 | 0.93 | 2 | 6.5 | 0.04 | 4 | 5.3 | 0.26 | 2 | 0.9 | 0.65 |

| | N% ¹ | | | C/N ¹ | | | $\delta^{13}\text{C}$ ¹ | | | $\delta^{15}\text{N}$ ¹ | | |
|------------|-----------------|----------------|------------------|------------------|----------------|------------------|------------------------------------|----------------|------------------|------------------------------------|----------------|------------------|
| | Df | X ² | p | Df | X ² | p | Df | X ² | p | Df | X ² | p |
| Depth | 2 | 27.8 | <0.001 | 2 | 4 | <0.001 | 2 | 47.6 | <0.001 | 2 | 20.4 | <0.001 |
| Type | 2 | 2.8 | 0.25 | 2 | 2.0 | 0.38 | 2 | 1.5 | 0.47 | 2 | 2.0 | 0.36 |
| Depth*Type | 4 | 10.2 | 0.04 | 4 | 0 | 0.04 | 4 | 7.7 | 0.10 | 4 | 1.2 | 0.89 |

¹Depth was treated as a quadratic polynomial term

Table S8. Output from mixed effects models testing the effects of sampling depth and topography type and their interactions on peat lead (Pb) and iron (Fe) concentrations. $p < 0.05$ are marked in bold.

| | Df | Pb | | Fe | |
|------------|----|----------------|------------------|----------------|------------------|
| | | X ² | p | X ² | p |
| Depth | 2 | 31.5 | <0.001 | 288.9 | <0.001 |
| Type | 2 | 7.4 | 0.02 | 3.4 | 0.18 |
| Depth*Type | 4 | 4.1 | 0.39 | 1.1 | 0.90 |

Table S9. Lead isotopic composition in water samples from Mycklemossen, Ersjön and forested catchment.

| Sample | Comment | mean counts per second (measured) | | | | | 204Hg Correction on mass 204 | | | Uncertainty ³ 2se | 206Pb/207Pb | | 208Pb/206Pb | | 206Pb/204Pb | | 207Pb/204Pb | | 208Pb/204Pb | |
|--|--------------|-----------------------------------|--------------|-------|--------|--------|------------------------------|---------------------------------------|-------|---------------------------------|-------------|-------|-------------|-------|-------------|-------|-------------|-------|-------------|-------|
| | | 202H g | 204Hg+P b | 206Pb | 207Pb | 208Pb | 204Hg ¹ | size 204Pb correction ² | ratio | | 2se | ratio | 2se | ratio | 2se | ratio | 2se | ratio | 2se | ratio |
| Primary reference standard | | | | | | | | | | | | | | | | | | | | |
| 1 | NIST SRM 981 | approximately 11 µg/l Pb | 7 | 29823 | 513125 | 457708 | 1E+06 | 0 | ## | 0.00% | 1.095 | 0.003 | 2.166 | 0.005 | 16.94 | 0.02 | 15.48 | 0.04 | 36.61 | 0.07 |
| 2 | NIST SRM 981 | approximately 11 µg/l Pb | 7 | 28995 | 499027 | 450264 | 1E+06 | 1 | ## | 0.00% | 1.091 | 0.003 | 2.173 | 0.006 | 16.94 | 0.03 | 15.54 | 0.05 | 36.76 | 0.07 |
| 3 | NIST SRM 981 | approximately 11 µg/l Pb | 7 | 28471 | 489524 | 444035 | 1E+06 | 0 | ## | 0.00% | 1.091 | 0.003 | 2.171 | 0.004 | 16.92 | 0.03 | 15.52 | 0.03 | 36.69 | 0.07 |
| 4 | NIST SRM 981 | approximately 11 µg/l Pb | 6 | 28333 | 487591 | 442919 | 1E+06 | 0 | ## | 0.00% | 1.094 | 0.003 | 2.165 | 0.004 | 16.94 | 0.03 | 15.48 | 0.04 | 36.62 | 0.06 |
| 5 | NIST SRM 981 | approximately 11 µg/l Pb | 6 | 28559 | 491832 | 446917 | 1E+06 | 0 | ## | 0.00% | 1.094 | 0.003 | 2.167 | 0.004 | 16.95 | 0.02 | 15.49 | 0.04 | 36.68 | 0.06 |
| 6 | NIST SRM 981 | approximately 11 µg/l Pb | 6 | 28368 | 488236 | 443988 | 1E+06 | 0 | ## | 0.00% | 1.093 | 0.003 | 2.169 | 0.004 | 16.94 | 0.03 | 15.50 | 0.04 | 36.70 | 0.04 |
| 7 | NIST SRM 981 | approximately 11 µg/l Pb | 6 | 28502 | 490553 | 445430 | 1E+06 | 0 | ## | 0.00% | 1.095 | 0.003 | 2.167 | 0.004 | 16.94 | 0.03 | 15.47 | 0.04 | 36.65 | 0.05 |
| 8 | NIST SRM 981 | approximately 11 µg/l Pb | 6 | 28711 | 493801 | 448938 | 1E+06 | 0 | ## | 0.00% | 1.093 | 0.002 | 2.167 | 0.005 | 16.93 | 0.03 | 15.48 | 0.04 | 36.63 | 0.06 |
| 9 | NIST SRM 981 | approximately 11 µg/l Pb | 6 | 28556 | 491010 | 446335 | 1E+06 | 1 | ## | 0.00% | 1.094 | 0.003 | 2.169 | 0.004 | 16.92 | 0.02 | 15.47 | 0.04 | 36.65 | 0.06 |
| Secondary reference standard (NIST SRM 981; but with different concentration as the primary standard) | | | | | | | | | | | | | | | | | | | | |
| 1 | NIST SRM 981 | approximately 5.5 µg/l Pb | 5 | 14774 | 254047 | 227357 | 532062 | 0 | ## | 0.00% | 1.093 | 0.004 | 2.163 | 0.005 | 16.93 | 0.03 | 15.50 | 0.04 | 36.55 | 0.05 |
| 2 | NIST SRM 981 | approximately 5.5 µg/l Pb | 5 | 14263 | 245303 | 222896 | 522263 | 0 | ## | 0.00% | 1.094 | 0.003 | 2.166 | 0.005 | 16.93 | 0.04 | 15.47 | 0.05 | 36.62 | 0.06 |
| 3 | NIST SRM 981 | approximately 5.5 µg/l Pb | 6 | 14331 | 246485 | 224085 | 525651 | 1 | ## | 0.00% | 1.093 | 0.003 | 2.170 | 0.004 | 16.93 | 0.04 | 15.48 | 0.05 | 36.68 | 0.06 |
| 4 | NIST SRM 981 | approximately 5.5 µg/l Pb | 7 | 28414 | 489067 | 444163 | 1E+06 | 0 | ## | 0.00% | 1.095 | 0.003 | 2.166 | 0.003 | 16.94 | 0.02 | 15.48 | 0.03 | 36.64 | 0.05 |
| 5 | NIST SRM 981 | approximately 5.5 µg/l Pb | 5 | 14439 | 248584 | 225940 | 529396 | 0 | ## | 0.00% | 1.094 | 0.003 | 2.167 | 0.003 | 16.95 | 0.02 | 15.49 | 0.04 | 36.67 | 0.06 |
| Secondary reference standard (NIST SRM 981; but with different concentration as the primary standard) | | | | | | | | | | | | | | | | | | | | |
| 1 | NIST SRM 981 | approximately 1 µg/l Pb | 4 | 3010 | 51625 | 46367 | 108476 | 0 | ## | 0.00% | 1.091 | 0.005 | 2.167 | 0.010 | 16.89 | 0.05 | 15.49 | 0.08 | 36.53 | 0.18 |
| 2 | NIST SRM 981 | approximately 1 µg/l Pb | 6 | 2970 | 51118 | 46295 | 108461 | 0 | ## | 0.00% | 1.088 | 0.004 | 2.177 | 0.007 | 16.95 | 0.08 | 15.59 | 0.07 | 36.84 | 0.17 |
| 3 | NIST SRM 981 | approximately 1 µg/l Pb | 5 | 2939 | 50474 | 46000 | 107636 | 0 | ## | 0.00% | 1.091 | 0.004 | 2.170 | 0.007 | 16.91 | 0.08 | 15.51 | 0.08 | 36.65 | 0.17 |
| 4 | NIST SRM 981 | approximately 1 µg/l Pb | 5 | 2947 | 50548 | 46191 | 107881 | 0 | ## | 0.00% | 1.088 | 0.005 | 2.172 | 0.008 | 16.89 | 0.06 | 15.53 | 0.08 | 36.63 | 0.17 |
| 5 | NIST SRM 981 | approximately 1 µg/l Pb | 4 | 2953 | 50748 | 46268 | 108341 | 0 | ## | 0.00% | 1.090 | 0.004 | 2.172 | 0.009 | 16.92 | 0.07 | 15.52 | 0.07 | 36.70 | 0.16 |
| Secondary reference water (AQUA-1) | | | | | | | | | | | | | | | | | | | | |
| 1 | AQUA1 1/1 | | 9 | 3285 | 57300 | 50719 | 119003 | 0 | ## | 0.01% | 1.106 | 0.004 | 2.144 | 0.006 | 17.18 | 0.07 | 15.55 | 0.07 | 36.77 | 0.19 |
| 2 | AQUA1 1/1 | | 10 | 3187 | 55638 | 50022 | 117287 | 0 | ## | 0.01% | 1.106 | 0.005 | 2.145 | 0.006 | 17.20 | 0.08 | 15.55 | 0.05 | 36.83 | 0.12 |
| 3 | AQUA1 1/1 | | 7 | 3193 | 55640 | 50052 | 117661 | 0 | ## | 0.01% | 1.105 | 0.004 | 2.152 | 0.010 | 17.16 | 0.06 | 15.53 | 0.06 | 36.88 | 0.17 |
| 4 | AQUA1 1/1 | | 12 | 3214 | 55888 | 50294 | 118076 | 1 | ## | 0.01% | 1.105 | 0.003 | 2.150 | 0.005 | 17.13 | 0.09 | 15.50 | 0.08 | 36.77 | 0.18 |
| 5 | AQUA1 1/1 | | 9 | 3216 | 55834 | 50339 | 118117 | 1 | ## | 0.01% | 1.103 | 0.004 | 2.153 | 0.009 | 17.10 | 0.08 | 15.51 | 0.06 | 36.75 | 0.12 |
| Secondary reference standard (Skogaryd 01) | | | | | | | | | | | | | | | | | | | | |
| 1 | Skogaryd 01 | | ## | 14074 | 254558 | 220648 | 527785 | 36 | ## | 0.02% | 1.147 | 0.003 | 2.110 | 0.005 | 17.85 | 0.05 | 15.56 | 0.04 | 37.60 | 0.09 |
| 2 | Skogaryd 01 | | ## | 14140 | 255769 | 221631 | 530717 | 34 | ## | 0.02% | 1.147 | 0.003 | 2.111 | 0.003 | 17.85 | 0.02 | 15.56 | 0.05 | 37.63 | 0.04 |
| 3 | Skogaryd 01 | | ## | 14117 | 255684 | 222058 | 531647 | 33 | ## | 0.02% | 1.145 | 0.003 | 2.116 | 0.004 | 17.87 | 0.04 | 15.61 | 0.05 | 37.75 | 0.09 |
| 4 | Skogaryd 01 | | ## | 14103 | 255255 | 221333 | 529803 | 35 | ## | 0.02% | 1.146 | 0.004 | 2.112 | 0.006 | 17.86 | 0.04 | 15.58 | 0.06 | 37.67 | 0.14 |
| Samples from Skogaryd | | | | | | | | | | | | | | | | | | | | |

Sample locality 01

| | | | | | | | | | | | | | | | | | | | |
|----|----------------------|----|-------|--------|--------|--------|----|----|-------|---------------|---------------|--------------|----------------|----------------|----------------|---------------|--------------|---------------|--------------|
| 1 | 5-101220201 | ## | 6378 | 114943 | 98527 | 235933 | 61 | ## | 0.04% | 1.146 | 0.003 | 2.113 | 0.006 | 17.89 | 0.04 | 15.63 | 0.03 | 37.73 | 0.05 |
| 2 | 5-101220501 | ## | 6821 | 123014 | 105523 | 252162 | 69 | ## | 0.04% | 1.144 | 0.003 | 2.111 | 0.005 | 17.89 | 0.04 | 15.65 | 0.05 | 37.70 | 0.08 |
| 3 | 5-101220801 | ## | 13042 | 236578 | 202878 | 486051 | 31 | ## | 0.02% | 1.146 | 0.004 | 2.112 | 0.006 | 17.90 | 0.04 | 15.62 | 0.03 | 37.75 | 0.06 |
| 4 | 5-101221001 | ## | 10486 | 189857 | 163662 | 391736 | 45 | ## | 0.02% | 1.144 | 0.003 | 2.116 | 0.005 | 17.89 | 0.05 | 15.65 | 0.05 | 37.80 | 0.06 |
| 5 | 5-101221201 | ## | 5894 | 105892 | 90861 | 217062 | 62 | ## | 0.05% | 1.143 | 0.004 | 2.113 | 0.005 | 17.85 | 0.04 | 15.64 | 0.06 | 37.66 | 0.10 |
| 6 | 5-101221501 | ## | 9361 | 169221 | 145466 | 348180 | 35 | ## | 0.02% | 1.143 | 0.003 | 2.117 | 0.006 | 17.86 | 0.03 | 15.64 | 0.04 | 37.75 | 0.07 |
| 7 | 5-101221701 | ## | 15000 | 271236 | 232187 | 555445 | 79 | ## | 0.02% | 1.146 | 0.003 | 2.110 | 0.005 | 17.89 | 0.05 | 15.62 | 0.05 | 37.67 | 0.09 |
| 8 | 5-101221801 | ## | 12373 | 223890 | 193952 | 463446 | 46 | ## | 0.02% | 1.148 | 0.004 | 2.106 | 0.004 | 17.88 | 0.02 | 15.58 | 0.04 | 37.60 | 0.05 |
| 9 | 5-101230401 | ## | 7015 | 126613 | 109739 | 262641 | 35 | ## | 0.03% | 1.147 | 0.003 | 2.111 | 0.007 | 17.85 | 0.05 | 15.56 | 0.06 | 37.63 | 0.14 |
| 10 | 5-101230501 | ## | 8880 | 160359 | 138927 | 333480 | 42 | ## | 0.03% | 1.147 | 0.004 | 2.116 | 0.005 | 17.85 | 0.04 | 15.55 | 0.06 | 37.72 | 0.11 |
| 11 | 5-101230601 | ## | 14329 | 259464 | 224541 | 538054 | 37 | ## | 0.02% | 1.149 | 0.003 | 2.110 | 0.003 | 17.87 | 0.04 | 15.56 | 0.04 | 37.65 | 0.08 |
| 12 | 5-101230701 | ## | 11732 | 212641 | 183439 | 439248 | 30 | ## | 0.02% | 1.144 | 0.003 | 2.116 | 0.005 | 17.88 | 0.06 | 15.64 | 0.06 | 37.79 | 0.10 |
| 13 | 5-101230901 | ## | 14139 | 256818 | 221596 | 529791 | 32 | ## | 0.01% | 1.144 | 0.003 | 2.112 | 0.004 | 17.92 | 0.03 | 15.66 | 0.04 | 37.79 | 0.10 |
| 14 | 5-101231001 | ## | 17147 | 310782 | 269426 | 643135 | 40 | ## | 0.01% | 1.144 | 0.003 | 2.110 | 0.004 | 17.88 | 0.04 | 15.63 | 0.04 | 37.68 | 0.07 |
| 15 | 5-101231101 | ## | 15047 | 272315 | 235789 | 565032 | 50 | ## | 0.02% | 1.148 | 0.003 | 2.111 | 0.005 | 17.87 | 0.03 | 15.56 | 0.03 | 37.67 | 0.05 |
| 16 | 5-101231201 | ## | 16318 | 296342 | 255758 | 612324 | 41 | ## | 0.01% | 1.145 | 0.003 | 2.115 | 0.005 | 17.92 | 0.04 | 15.66 | 0.04 | 37.83 | 0.06 |
| 17 | 5-101231301 | ## | 15965 | 289932 | 250519 | 599442 | 37 | ## | 0.01% | 1.147 | 0.003 | 2.109 | 0.005 | 17.91 | 0.04 | 15.61 | 0.04 | 37.73 | 0.06 |
| 18 | 5-101231401 | ## | 15467 | 280632 | 242670 | 581666 | 34 | ## | 0.01% | 1.150 | 0.003 | 2.109 | 0.004 | 17.89 | 0.02 | 15.56 | 0.03 | 37.68 | 0.08 |
| 19 | 5-101231501 | ## | 13973 | 253370 | 218448 | 522721 | 35 | ## | 0.02% | 1.144 | 0.004 | 2.115 | 0.005 | 17.89 | 0.03 | 15.64 | 0.05 | 37.76 | 0.07 |
| 20 | 5-101231601 | ## | 11786 | 213544 | 184863 | 442265 | 32 | ## | 0.02% | 1.147 | 0.004 | 2.110 | 0.003 | 17.88 | 0.04 | 15.58 | 0.05 | 37.65 | 0.04 |
| 21 | 5-101231701 | ## | 10118 | 183638 | 158824 | 380740 | 25 | ## | 0.02% | 1.149 | 0.003 | 2.110 | 0.004 | 17.91 | 0.04 | 15.58 | 0.05 | 37.73 | 0.07 |
| 22 | 5-101240101 | 93 | 6311 | 114521 | 98967 | 236816 | 24 | ## | 0.03% | 1.146 | 0.004 | 2.111 | 0.006 | 17.92 | 0.06 | 15.63 | 0.06 | 37.77 | 0.14 |
| 23 | 5-101240201 | 83 | 6533 | 118478 | 102282 | 244885 | 18 | ## | 0.02% | 1.145 | 0.003 | 2.114 | 0.006 | 17.90 | 0.04 | 15.64 | 0.04 | 37.79 | 0.07 |
| 24 | 5-101240301 | ## | 11074 | 200814 | 173562 | 416679 | 21 | ## | 0.02% | 1.148 | 0.004 | 2.115 | 0.006 | 17.89 | 0.04 | 15.58 | 0.03 | 37.77 | 0.07 |
| 25 | 5-101240501 | 99 | 12674 | 229964 | 198819 | 476264 | 21 | ## | 0.01% | 1.145 | 0.003 | 2.115 | 0.004 | 17.89 | 0.04 | 15.62 | 0.04 | 37.78 | 0.09 |
| | min | | | | | | | | | 1.143 | | 2.106 | | 17.850 | | 15.554 | | 37.598 | |
| | max | | | | | | | | | 1.150 | | 2.117 | | 17.921 | | 15.663 | | 37.832 | |
| | weighted mean | | | | | | | | | 1.1460 | 0.0008 | # | 2.11175 | 0.00095 | 17.8872 | 0.0070 | 15.61 | 0.02 | 37.72 |
| | MSWD | | | | | | | | | 1.5 | | 1.4 | | 1.13 | | 2.70 | | 3.40 | |
| | n= | | | | | | | | | 25 | | 25 | | 25 | | 25 | | 25 | |

Sample locality 05

| | | | | | | | | | | | | | | | | | | | |
|---|----------------------|----|------|--------|--------|--------|----|----|-------|---------------|---------------|--------------|----------------|----------------|----------------|---------------|--------------|---------------|--------------|
| 1 | 5-101220805 | ## | 5480 | 99245 | 85173 | 205489 | 58 | ## | 0.05% | 1.158 | 0.004 | 2.107 | 0.006 | 17.98 | 0.03 | 15.52 | 0.04 | 37.83 | 0.08 |
| 2 | 5-101221805 | ## | 8476 | 154306 | 132293 | 318983 | 59 | ## | 0.03% | 1.160 | 0.003 | 2.103 | 0.004 | 18.04 | 0.02 | 15.56 | 0.04 | 37.89 | 0.07 |
| 3 | 5-101230105 | ## | 5072 | 92529 | 79243 | 191143 | 48 | ## | 0.05% | 1.161 | 0.004 | 2.102 | 0.005 | 18.11 | 0.06 | 15.60 | 0.08 | 38.02 | 0.16 |
| 4 | 5-101230205 | ## | 5118 | 93100 | 79953 | 192412 | 49 | ## | 0.05% | 1.158 | 0.004 | 2.103 | 0.005 | 18.06 | 0.04 | 15.60 | 0.06 | 37.91 | 0.10 |
| 5 | 5-101230405 | ## | 5250 | 95501 | 82088 | 197944 | 40 | ## | 0.05% | 1.157 | 0.003 | 2.109 | 0.005 | 18.06 | 0.05 | 15.61 | 0.07 | 38.02 | 0.13 |
| 6 | 5-101230505 | ## | 6582 | 119705 | 102690 | 247831 | 59 | ## | 0.04% | 1.159 | 0.004 | 2.107 | 0.005 | 18.05 | 0.06 | 15.57 | 0.06 | 37.96 | 0.14 |
| 7 | 5-101230605 | ## | 7310 | 132790 | 114404 | 275347 | 37 | ## | 0.03% | 1.154 | 0.003 | 2.110 | 0.004 | 17.97 | 0.05 | 15.57 | 0.06 | 37.85 | 0.07 |
| 8 | 5-101230705 | ## | 7432 | 135695 | 116492 | 280615 | 72 | ## | 0.03% | 1.158 | 0.004 | 2.104 | 0.004 | 18.06 | 0.05 | 15.59 | 0.05 | 37.95 | 0.11 |
| | min | | | | | | | | | 1.154 | | 2.102 | | 17.966 | | 15.523 | | 37.831 | |
| | max | | | | | | | | | 1.161 | | 2.110 | | 18.114 | | 15.609 | | 38.021 | |
| | weighted mean | | | | | | | | | 1.1580 | 0.0013 | # | 2.10560 | 0.00170 | 18.0310 | 0.0330 | 15.57 | 0.02 | 37.90 |
| | MSWD | | | | | | | | | 1.5 | | 1.6 | | 3.9 | | 1.3 | | 1.8 | |
| | n= | | | | | | | | | 8 | | 8 | | 8 | | 8 | | 8 | |

Sample locality 12

| | | | | | | | | | | | | | | | | | | | |
|----|----------------------|----|------|-------|-------|--------|----|----|-------|---------------|---------------|--------------|----------------|----------------|----------------|---------------|--------------|---------------|--------------|
| 1 | 5-101230112 | 98 | 1409 | 25792 | 21884 | 52889 | 23 | ## | 0.13% | 1.172 | 0.005 | 2.086 | 0.008 | 18.32 | 0.18 | 15.63 | 0.13 | 38.15 | 0.31 |
| 2 | 5-101230412 | ## | 1002 | 18379 | 15406 | 37398 | 27 | ## | 0.20% | 1.187 | 0.006 | 2.070 | 0.012 | 18.57 | 0.16 | 15.65 | 0.11 | 38.39 | 0.32 |
| 3 | 5-101230512 | ## | 1590 | 29001 | 24624 | 59518 | 30 | ## | 0.13% | 1.171 | 0.007 | 2.088 | 0.008 | 18.30 | 0.12 | 15.62 | 0.10 | 38.15 | 0.21 |
| 4 | 5-101230612 | ## | 1103 | 20168 | 16865 | 41101 | 35 | ## | 0.21% | 1.189 | 0.009 | 2.073 | 0.007 | 18.66 | 0.11 | 15.69 | 0.16 | 38.63 | 0.23 |
| 5 | 5-101231112 | ## | 1928 | 35372 | 29997 | 72434 | 37 | ## | 0.11% | 1.172 | 0.005 | 2.083 | 0.008 | 18.38 | 0.08 | 15.67 | 0.06 | 38.23 | 0.16 |
| 6 | 5-101231212 | ## | 2494 | 45661 | 38722 | 93433 | 39 | ## | 0.09% | 1.172 | 0.005 | 2.082 | 0.004 | 18.30 | 0.08 | 15.61 | 0.05 | 38.05 | 0.14 |
| 7 | 5-101231312 | ## | 2885 | 53103 | 44806 | 108354 | 35 | ## | 0.08% | 1.178 | 0.004 | 2.076 | 0.007 | 18.37 | 0.07 | 15.59 | 0.07 | 38.07 | 0.16 |
| 8 | 5-101231412 | ## | 1876 | 34705 | 28968 | 70336 | 42 | ## | 0.13% | 1.191 | 0.004 | 2.062 | 0.009 | 18.61 | 0.10 | 15.62 | 0.09 | 38.31 | 0.17 |
| 9 | 5-101231512 | ## | 3137 | 57776 | 48861 | 118077 | 33 | ## | 0.07% | 1.176 | 0.004 | 2.079 | 0.007 | 18.34 | 0.06 | 15.59 | 0.07 | 38.07 | 0.14 |
| 10 | 5-101231612 | ## | 1783 | 32714 | 27692 | 66982 | 29 | ## | 0.11% | 1.175 | 0.005 | 2.083 | 0.010 | 18.34 | 0.09 | 15.61 | 0.05 | 38.15 | 0.27 |
| 11 | 5-1012317012 | 92 | 1966 | 36193 | 30758 | 74227 | 22 | ## | 0.09% | 1.170 | 0.005 | 2.087 | 0.007 | 18.32 | 0.05 | 15.66 | 0.07 | 38.17 | 0.13 |
| | min | | | | | | | | | 1.170 | | 2.062 | | 18.300 | | 15.585 | | 38.048 | |
| | max | | | | | | | | | 1.191 | | 2.088 | | 18.660 | | 15.688 | | 38.626 | |
| | weighted mean | | | | | | | | | 1.1778 | 0.0052 | # | 2.08010 | 0.00460 | 18.3760 | 0.0710 | 15.63 | 0.02 | 38.18 |
| | MSWD | | | | | | | | | 9.0 | | 3.8 | | 6.4 | | 0.8 | | 2.7 | |
| | n= | | | | | | | | | 11 | | 11 | | 11 | | 11 | | 11 | |

Sample locality 06

| | | | | | | | | | | | | | | | | | | | |
|----|----------------------|----|------|-------|-------|--------|----|----|-------|------------|---------------|------------|----------------|----------------|----------------|---------------|--------------|-------------|--------------|
| 1 | 5-101220506 | ## | 2389 | 42880 | 36951 | 88697 | 50 | ## | 0.10% | 1.154 | 0.005 | 2.105 | 0.008 | 17.98 | 0.09 | 15.58 | 0.10 | 37.79 | 0.20 |
| 2 | 5-101230106 | ## | 3947 | 71482 | 61586 | 147866 | 40 | ## | 0.06% | 1.154 | 0.004 | 2.105 | 0.006 | 18.01 | 0.05 | 15.61 | 0.04 | 37.85 | 0.08 |
| 3 | 5-101230206 | ## | 3349 | 60678 | 52292 | 125620 | 33 | ## | 0.07% | 1.154 | 0.004 | 2.106 | 0.006 | 18.02 | 0.06 | 15.62 | 0.08 | 37.91 | 0.18 |
| 4 | 5-101230406 | ## | 2570 | 46106 | 39760 | 95425 | 46 | ## | 0.09% | 1.153 | 0.005 | 2.106 | 0.007 | 17.92 | 0.09 | 15.55 | 0.09 | 37.69 | 0.19 |
| 5 | 5-101230506 | ## | 2264 | 40645 | 35069 | 84071 | 34 | ## | 0.10% | 1.152 | 0.004 | 2.104 | 0.005 | 17.95 | 0.07 | 15.58 | 0.08 | 37.73 | 0.16 |
| 6 | 5-101230606 | ## | 1908 | 34194 | 29537 | 70880 | 33 | ## | 0.12% | 1.151 | 0.005 | 2.109 | 0.010 | 17.96 | 0.06 | 15.61 | 0.06 | 37.83 | 0.17 |
| 7 | 5-101230706 | 95 | 1297 | 23257 | 20049 | 48161 | 23 | ## | 0.13% | 1.153 | 0.004 | 2.107 | 0.011 | 17.96 | 0.09 | 15.57 | 0.07 | 37.79 | 0.16 |
| 8 | 5-101230806 | 73 | 1904 | 34444 | 29692 | 71277 | 15 | ## | 0.08% | 1.153 | 0.006 | 2.105 | 0.007 | 17.97 | 0.07 | 15.58 | 0.08 | 37.77 | 0.15 |
| 9 | 5-101231006 | 67 | 1551 | 28065 | 24196 | 58021 | 15 | ## | 0.09% | 1.153 | 0.004 | 2.103 | 0.010 | 17.99 | 0.11 | 15.60 | 0.11 | 37.79 | 0.27 |
| 10 | 5-101231106 | 82 | 1740 | 31494 | 27198 | 65157 | 17 | ## | 0.09% | 1.151 | 0.005 | 2.105 | 0.006 | 18.02 | 0.09 | 15.65 | 0.09 | 37.87 | 0.20 |
| 11 | 5-101231206 | ## | 2929 | 53089 | 45750 | 109988 | 21 | ## | 0.06% | 1.154 | 0.004 | 2.108 | 0.004 | 17.98 | 0.08 | 15.59 | 0.07 | 37.86 | 0.18 |
| 12 | 5-101231306 | ## | 4781 | 86855 | 74850 | 179858 | 23 | ## | 0.04% | 1.154 | 0.005 | 2.107 | 0.008 | 17.98 | 0.06 | 15.58 | 0.07 | 37.82 | 0.17 |
| 13 | 5-101231406 | ## | 4645 | 84224 | 72737 | 174559 | 28 | ## | 0.04% | 1.151 | 0.003 | 2.109 | 0.005 | 17.95 | 0.07 | 15.59 | 0.06 | 37.80 | 0.15 |
| 14 | 5-101231506 | ## | 5003 | 90736 | 78156 | 187389 | 26 | ## | 0.04% | 1.154 | 0.003 | 2.101 | 0.005 | 17.95 | 0.05 | 15.55 | 0.06 | 37.66 | 0.11 |
| 15 | 5-101231606 | ## | 5405 | 98280 | 84738 | 203045 | 26 | ## | 0.04% | 1.153 | 0.004 | 2.102 | 0.005 | 17.98 | 0.04 | 15.59 | 0.05 | 37.75 | 0.09 |
| 16 | 5-101231706 | 96 | 4070 | 73687 | 63745 | 152749 | 17 | ## | 0.04% | 1.149 | 0.004 | 2.109 | 0.005 | 17.92 | 0.07 | 15.59 | 0.07 | 37.73 | 0.12 |
| 17 | 5-101240106 | 80 | 2704 | 49098 | 42349 | 101688 | 17 | ## | 0.06% | 1.153 | 0.005 | 2.107 | 0.006 | 18.00 | 0.05 | 15.61 | 0.05 | 37.87 | 0.12 |
| 18 | 5-101240206 | 75 | 2792 | 50688 | 43674 | 104760 | 17 | ## | 0.05% | 1.154 | 0.004 | 2.103 | 0.008 | 17.98 | 0.09 | 15.58 | 0.08 | 37.76 | 0.15 |
| 19 | 5-101240306 | 78 | 2580 | 46841 | 40438 | 97038 | 18 | ## | 0.06% | 1.152 | 0.004 | 2.108 | 0.006 | 18.00 | 0.06 | 15.63 | 0.08 | 37.88 | 0.13 |
| 20 | 5-101240406 | 60 | 2045 | 37009 | 31953 | 76517 | 15 | ## | 0.07% | 1.151 | 0.007 | 2.104 | 0.010 | 17.94 | 0.07 | 15.58 | 0.08 | 37.68 | 0.17 |
| 21 | 5-101240506 | 62 | 2467 | 44871 | 38702 | 92941 | 15 | ## | 0.06% | 1.153 | 0.004 | 2.107 | 0.006 | 18.01 | 0.07 | 15.62 | 0.06 | 37.90 | 0.10 |
| | min | | | | | | | | | 1.149 | | 2.101 | | 17.916 | | 15.545 | | 37.664 | |
| | max | | | | | | | | | 1.154 | | 2.109 | | 18.023 | | 15.650 | | 37.909 | |
| | weighted mean | | | | | | | | | 1.1527 | 0.0009 | # | 2.10690 | 0.00140 | 17.9790 | 0.0140 | 15.60 | 0.02 | 37.80 |
| | MSWD | | | | | | | | | 0.4 | | 0.6 | | 0.9 | | 0.4 | | 1.3 | |
| | n= | | | | | | | | | 21 | | 21 | | 21 | | 21 | | 21 | |

Blank analyses over the whole analytical session

| | | | | | | |
|----|-------|---|----|-----|-----|-----|
| 1 | Blank | 4 | 8 | 117 | 106 | 244 |
| 2 | Blank | 5 | 10 | 166 | 150 | 366 |
| 3 | Blank | 5 | 9 | 147 | 132 | 311 |
| 4 | Blank | 3 | 9 | 128 | 115 | 277 |
| 5 | Blank | 5 | 7 | 94 | 86 | 195 |
| 6 | Blank | 5 | 7 | 100 | 88 | 209 |
| 7 | Blank | 4 | 11 | 186 | 160 | 378 |
| 8 | Blank | 8 | 11 | 166 | 141 | 333 |
| 9 | Blank | 6 | 12 | 191 | 165 | 393 |
| 10 | Blank | 6 | 10 | 135 | 119 | 283 |
| 11 | Blank | 5 | 11 | 174 | 155 | 370 |
| 12 | Blank | 6 | 10 | 159 | 145 | 347 |
| 13 | Blank | 4 | 11 | 183 | 164 | 387 |
| 14 | Blank | 4 | 13 | 213 | 196 | 450 |
| 15 | Blank | 5 | 10 | 166 | 150 | 361 |
| 16 | Blank | 6 | 9 | 125 | 114 | 275 |
| 17 | Blank | 4 | 12 | 196 | 183 | 432 |
| 18 | Blank | 5 | 9 | 134 | 120 | 293 |
| 19 | Blank | 5 | 12 | 191 | 174 | 386 |
| 20 | Blank | 4 | 7 | 97 | 86 | 206 |
| 21 | Blank | 5 | 9 | 136 | 115 | 272 |
| 22 | Blank | 4 | 11 | 163 | 147 | 343 |
| 23 | Blank | 5 | 11 | 171 | 155 | 353 |

Supplementary information - Pb isotopic analysis

Pb isotopic analysis for determination of origin of Pb in the environment

Pb isotope analyses were performed at the Microgeochemistry Laboratory at the University of Gothenburg using an Agilent 8800 ICP-MS/MS. The sample introduction system used was an ASX-500 autosampler (Agilent), a peristaltic pump, a concentric glass nebulizer (MicroMist) and a Scott double-pass quartz glass spray chamber. The AS-500 autosampler was placed in an ISO 5 clean room environment and was connected to the ICP-MS in the adjacent Microgeochemistry laboratory with a tubing length of approximately 1 m.

All sample preparations were performed in the ISO 5 clean room environment using Milli-Q water (resistivity > 18.2 MΩ cm) and ultrapure nitric acid (NORMATOM®, VWR chemicals). Samples, reference waters and blanks were prepared with a matrix of 2% HNO₃. A dilute solution of the digested common lead isotopic Standard Reference Material (SRM) 981 (NIST, (Cantanzaro et al., 1968)) was used as primary standard to correct for drift and mass bias. The approximate lead concentration of this primary reference solution was 11 µg/l. AQUA-1 natural drinking water certified reference material (NRC Canada, (Yeghicheyan et al., 2021)) was analysed as secondary reference solution to estimate accuracy and precision. Weighted averages for all lead isotope ratios for five interspersed AQUA-1 analyses overlap in 2 s with the Pb isotopes ratios reported by Yeghicheyan et al. 2021 and their expanded uncertainties. To ascertain identical lead ratios for lead isotopes measured across pulse counting mode, two other dilutions of NIST SRM 981 with concentrations of approximately 1.1 µg/l and 5.5 µg/l were run repeatedly throughout the measurement session. These lead concentrations are similar to the lead concentration of the water samples taken from Skogaryd. Pb isotopes. Additionally, a blank was measured after every standard and sample block and used to subtract the mercury and lead background. ²⁰⁴Hg on monitored counts per second (CPS) on mass 204 was subtracted by using CPS on ²⁰²Hg and the isotopic abundance for both isotopes (CIAAW, 2021)).

The operating conditions of the ICP-MS and acquisition parameters are listed in Table S2, S3 and S4. The ICP-MS was run in MS/MS mode with NO₂ as reaction gas. Tuning of the instrument was performed. The peak shape and position were optimised for masses 206, 207 and 208 for both quadrupoles using diluted NIST SRM 981 after auto-tune of the lenses using a 10 µg/l Li, Y, Ce, Tl, and Co Tuning solution (Agilent).

Uncertainties reported in the data table include the standard error of the 10 replicates added in quadrature with the excess scatter of the primary reference solution NIST SRM 981 of the respective ratios. The uncertainty in the ²⁰⁴Hg correction was propagated where necessary. All uncertainties are given at 2se. Systematic uncertainties are not reported in the data tables but include the ratio uncertainty of NIST SRM 981 and the long-term excess scatter.

References

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