

**Estimating the bioaccessibility of atmospheric trace elements within the  
Athabasca Bituminous Sands region using the acid soluble ash fraction  
of *Sphagnum* moss**

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**Supporting Information**

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## Presentation of Geochemical Data

This study generated a considerable volume of geochemical data: mosses were collected in triplicate during 3 seasons from a total of 38 peatlands, and both bulk samples and the acid soluble ash (ASA) fraction were analyzed for 5 major and 36 trace elements (TEs). For the convenience of the reader, the Supporting Information includes: i) quality assurance/control tables; ii) a set of maps for the samples collected in 2015, showing the spatial variation in TE concentrations for the bulk moss and ASA fractions; iii) bar graphs summarizing the ranges of concentrations (total and ASA) for industrial and reference sites for 2015, 2019, and 2020, and average TE concentrations for 2020; iv) statistical analyses of the data (i.e. correlograms of all elements and linear regressions of TE concentrations versus Y); v) box plots to evaluate temporal trends in TE concentrations; and vi) enrichment factors.

Enrichment factors were calculated as the ratio of the concentrations of a given TE to that of yttrium (Y), a conservative lithophile element, normalized to the corresponding ratio in the Upper Continental Crust (UCC).<sup>1</sup>

$$EF = \frac{[TE/Y]_{sample}}{[TE/Y]_{UCC}}$$

**Table S1.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRMs) recoveries for **total** concentrations analyzed using ICP-MS.

|    | NIST 1643f Freshwater |       |                 |                                  |          |                                   |          |
|----|-----------------------|-------|-----------------|----------------------------------|----------|-----------------------------------|----------|
|    | LOD                   | LOQ   | Certified value | Measured value*10<br>(mean, n=7) | Recovery | Measured value*100<br>(mean, n=7) | Recovery |
|    | µg/L                  | µg/L  | µg/L            | µg/L                             | %        | µg/L                              | %        |
| Ag | 0.000                 | 0.001 | 0.9703          | 0.905                            | 93       | 0.936                             | 96       |
| As | 0.002                 | 0.008 | 57.42           | 54.2                             | 94       | 54.2                              | 94       |
| Ba | 0.001                 | 0.004 | 518.2           | 492                              | 95       | 513                               | 99       |
| Be | 0.000                 | 0.002 | 13.67           | 12.4                             | 90       | 12.9                              | 94       |
| Cd | 0.000                 | 0.001 | 5.89            | 5.61                             | 95       | 5.75                              | 98       |
| Ce | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Co | 0.000                 | 0.000 | 25.3            | 23.2                             | 92       | 23.6                              | 93       |
| Cr | 0.000                 | 0.006 | 18.5            | 16.5                             | 89       | 18.2                              | 98       |
| Cs | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Cu | 0.001                 | 0.010 | 21.66           | 19.2                             | 89       | 21.0                              | 97       |
| Dy | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Er | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Eu | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Fe | 0.014                 | 0.066 | 93.44           | 88.8                             | 95       | 94.6                              | 101      |
| Ga | 0.000                 | 0.001 |                 |                                  |          |                                   |          |
| Gd | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Ho | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| La | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Li | 0.000                 | 0.002 | 16.59           | 14.7                             | 89       | 15.7                              | 95       |
| Lu | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Mn | 0.001                 | 0.003 | 37.14           | 41.0                             | 110      | 43.0                              | 116      |
| Mo | 0.000                 | 0.001 | 115.3           | 102                              | 89       | 105                               | 92       |
| Nd | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Ni | 0.002                 | 0.025 | 59.8            | 54.5                             | 91       | 55.7                              | 93       |
| Pb | 0.000                 | 0.000 | 18.488          | 16.8                             | 91       | 17.9                              | 97       |
| Pr | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Rb | 0.000                 | 0.001 | 12.64           | 12.0                             | 95       | 12.3                              | 97       |
| Re | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Sb | 0.002                 | 0.009 | 55.45           | 57.6                             | 104      | 58.8                              | 106      |
| Se | 0.024                 | 0.112 | 11.7            | 11.0                             | 94       | 11.3                              | 97       |
| Sm | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Sr | 0.001                 | 0.004 | 314             | 300                              | 96       | 312                               | 99       |
| Tb | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Th | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Tl | 0.000                 | 0.000 | 6.892           | 6.16                             | 89       | 6.52                              | 95       |
| Tm | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| U  | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| V  | 0.000                 | 0.001 | 36.07           | 33.3                             | 92       | 34.6                              | 96       |
| Y  | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Yb | 0.000                 | 0.000 |                 |                                  |          |                                   |          |
| Zn | 0.007                 | 0.056 | 74.4            | 71.2                             | 96       | 71.9                              | 97       |

Note: NIST = National Institute of Standards and Technology; SPS = Spectrapure Standards AS; FFRI = Finnish Forest Research Institute.

**Table S1.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRMs) recoveries for **total** concentrations analyzed using **ICP-MS** (continued).

|    | SPS-SW2 Surface Water |                                   |          |                                   |          |
|----|-----------------------|-----------------------------------|----------|-----------------------------------|----------|
|    | Certified value       | Measured value*100<br>(mean, n=7) | Recovery | Measured value*500<br>(mean, n=7) | Recovery |
|    | µg/L                  | µg/L                              | %        | µg/L                              | %        |
| Ag |                       |                                   |          |                                   |          |
| As | 50                    | 46.3                              | 93       | 47.0                              | 94       |
| Ba | 250                   | 239                               | 96       | 243                               | 97       |
| Be |                       |                                   |          |                                   |          |
| Cd | 2.5                   | 2.44                              | 98       | 2.37                              | 95       |
| Ce | 2.5                   | 2.44                              | 98       | 2.48                              | 99       |
| Co | 10                    | 9.17                              | 92       | 9.11                              | 91       |
| Cr | 10                    | 9.03                              | 90       | 9.12                              | 91       |
| Cs |                       |                                   |          |                                   |          |
| Cu | 100                   | 91.8                              | 92       | 92.9                              | 93       |
| Dy | 2.5                   | 2.44                              | 98       | 2.43                              | 97       |
| Er | 2.5                   | 2.42                              | 97       | 2.43                              | 97       |
| Eu | 2.5                   | 2.43                              | 97       | 2.43                              | 97       |
| Fe | 100                   | 94.0                              | 94       | 92.4                              | 92       |
| Ga |                       |                                   |          |                                   |          |
| Gd | 2.5                   | 2.44                              | 98       | 2.40                              | 96       |
| Ho | 2.5                   | 2.42                              | 97       | 2.47                              | 99       |
| La | 2.5                   | 2.41                              | 96       | 2.39                              | 96       |
| Li |                       |                                   |          |                                   |          |
| Lu | 2.5                   | 2.38                              | 95       | 2.38                              | 95       |
| Mn | 50                    | 56.7                              | 113      | 57.1                              | 114      |
| Mo | 50                    | 44.1                              | 88       | 45.0                              | 90       |
| Nd | 2.5                   | 2.43                              | 97       | 2.44                              | 98       |
| Ni | 50                    | 46.3                              | 93       | 46.6                              | 93       |
| Pb | 25                    | 22.9                              | 92       | 22.9                              | 92       |
| Pr | 2.5                   | 2.43                              | 97       | 2.44                              | 98       |
| Rb | 50                    | 47.3                              | 95       | 45.9                              | 92       |
| Re |                       |                                   |          |                                   |          |
| Sb |                       |                                   |          |                                   |          |
| Se | 10                    | 12.4                              | 124      | 14.0                              | 140      |
| Sm | 2.5                   | 2.55                              | 102      | 2.51                              | 101      |
| Sr | 250                   | 239                               | 96       | 244                               | 98       |
| Tb | 2.5                   | 2.49                              | 99       | 2.47                              | 99       |
| Th | 2.5                   | 2.36                              | 94       | 2.30                              | 92       |
| Tl | 2.5                   | 2.28                              | 91       | 2.30                              | 92       |
| Tm | 2.5                   | 2.44                              | 98       | 2.45                              | 98       |
| U  | 2.5                   | 2.35                              | 94       | 2.52                              | 101      |
| V  | 50                    | 46.4                              | 93       | 46.9                              | 94       |
| Y  | 2.5                   | 2.39                              | 96       | 2.33                              | 93       |
| Yb | 2.5                   | 2.37                              | 95       | 2.37                              | 95       |
| Zn | 100                   | 96.2                              | 96       | 91.6                              | 92       |

Note: FFRI = Finnish Forest Research Institute; NIST = National Institute of Standards and Technology; SPS = Spectrapure Standards AS.



**Table S1.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRMs) recoveries for **total** concentrations analyzed using **ICP-MS** (continued).

|    | NIST 1515 Apple Leaves |                 |                               |           | FFRI-Moss-1343       |                               |           |
|----|------------------------|-----------------|-------------------------------|-----------|----------------------|-------------------------------|-----------|
|    | MDL                    | Certified value | Measured value<br>(mean, n=6) | Recovery  | Recommended<br>value | Measured value<br>(mean, n=6) | Recovery  |
|    | µg/L                   | µg/g            | µg/g                          | %         | µg/g                 | µg/g                          | %         |
| Ag | 0.18                   |                 |                               |           | 0.027                | 0.0229                        | 85        |
| As | 70.78                  |                 |                               |           | 0.105                | 0.768                         | 732       |
| Ba | 4.35                   | <b>48.8</b>     | 49.4                          | 101       | 13.7                 | 14.1                          | 103       |
| Be | 0.05                   |                 |                               |           |                      |                               |           |
| Cd | 0.07                   | <b>0.0132</b>   | 0.0149                        | 113       | 0.106                | 0.119                         | 112       |
| Ce | 0.16                   | 3               | 3.35                          | 112       | 0.25                 | 0.271                         | 108       |
| Co | 0.96                   | 0.09            | 0.102                         | 114       | 0.115                | 0.108                         | 94        |
| Cr | 7.09                   | 0.3             | 0.360                         | 120       | 0.67                 | 0.740                         | 110       |
| Cs | 1.73                   |                 |                               |           | 0.189                | 0.177                         | 94        |
| Cu | 7.10                   | <b>5.69</b>     | 5.77                          | 101       | 3.76                 | 3.45                          | 92        |
| Dy | 0.02                   |                 |                               |           |                      |                               |           |
| Er | 0.00                   |                 |                               |           |                      |                               |           |
| Eu | 0.00                   | 0.2             | 0.264                         | 132       |                      |                               |           |
| Fe | 451.52                 | <b>82.7</b>     | 89.4                          | 108       | 138                  | 166                           | 120       |
| Ga | 0.07                   |                 |                               |           | 0.084                | 0.118                         | 140       |
| Gd | 0.01                   | 3               | 3.16                          | 105       |                      |                               |           |
| Ho | 0.00                   |                 |                               |           |                      |                               |           |
| La | 0.06                   | 20              | 21.1                          | 106       | 0.131                | 0.139                         | 106       |
| Li | 3.38                   |                 |                               |           |                      |                               |           |
| Lu | 0.00                   |                 |                               |           |                      |                               |           |
| Mn | 616.41                 | <b>54.1</b>     | 62.4                          | 115       | 535                  | 566                           | 106       |
| Mo | 0.82                   | <b>0.095</b>    | 0.0958                        | 101       | 0.1                  | 0.0563                        | <b>56</b> |
| Nd | 0.12                   | 17              | 17.0                          | 100       |                      |                               |           |
| Ni | 32.36                  | <b>0.936</b>    | 1.05                          | 112       | 0.95                 | 1.07                          | 112       |
| Pb | 0.53                   | <b>0.47</b>     | 0.407                         | 87        | 3.33                 | 3.54                          |           |
| Pr | 0.01                   |                 |                               |           |                      |                               |           |
| Rb | 0.70                   | <b>10.2</b>     | 9.93                          | 97        | 19.5                 | 18.8                          | 96        |
| Re | 0.00                   |                 |                               |           |                      |                               |           |
| Sb | 0.63                   | 0.013           | 0.0163                        | 125       | 0.052                | 0.0532                        | 102       |
| Se | 17.86                  |                 |                               |           | 0.115                | 0.132                         | 115       |
| Sm | 0.09                   | 3               | 3.06                          | 102       |                      |                               |           |
| Sr | 38.71                  | <b>25.1</b>     | 26.6                          | 106       | 4.64                 | 4.70                          | 101       |
| Tb | 0.00                   | 0.4             | 0.391                         | 98        |                      |                               |           |
| Th | 0.02                   | 0.03            | 0.0280                        | 93        | 0.027                | 0.0341                        |           |
| Tl | 0.03                   |                 |                               |           | 0.085                | 0.0473                        |           |
| Tm | 0.00                   |                 |                               |           |                      |                               |           |
| U  | 0.19                   | 0.006           | 0.00844                       | 141       | 0.085                | 0.0152                        |           |
| V  | 0.38                   | <b>0.254</b>    | 0.236                         | 93        | 1.19                 | 1.18                          | 99        |
| Y  | 0.06                   |                 |                               |           | 0.067                | 0.0782                        | 117       |
| Yb | 0.00                   | 0.3             | 0.194                         | <b>65</b> |                      |                               |           |
| Zn | 10.77                  | <b>12.45</b>    | 15.0                          | 120       | 25.4                 | 28.0                          | 110       |

Note: NIST = National Institute of Standards and Technology; SPS = Spectrapure Standards AS; FFRI = Finnish Forest Research Institute; bold values correspond to certified concentrations; values in red indicate low recoveries, below 80%.

**Table S2.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRMs) recoveries for concentrations in **acid soluble ash (ASA)** analyzed using **ICP-MS**.

|           | NIST 1643f Freshwater |       |       |                 |                                  |          |                                   |          |
|-----------|-----------------------|-------|-------|-----------------|----------------------------------|----------|-----------------------------------|----------|
|           | LOD                   | LOQ   | MDL   | Certified value | Measured value*10<br>(mean, n=7) | Recovery | Measured value*100<br>(mean, n=7) | Recovery |
|           | µg/L                  | µg/L  | µg/L  | µg/L            | µg/L                             | %        | µg/L                              | %        |
| <b>Ag</b> | 0.000                 | 0.000 | 0.001 | 0.9703          | 0.958                            | 99       | 1.01                              | 104      |
| <b>As</b> | 0.003                 | 0.011 | <LOQ  | 57.42           | 57.2                             | 100      | 58.5                              | 102      |
| <b>Ba</b> | 0.000                 | 0.003 | 0.059 | 518.2           | 500                              | 97       | 524                               | 101      |
| <b>Be</b> | 0.000                 | 0.001 | <LOD  | 13.67           | 13.5                             | 99       | 13.7                              | 100      |
| <b>Cd</b> | 0.000                 | 0.000 | <LOQ  | 5.89            | 5.79                             | 98       | 6.10                              | 104      |
| <b>Ce</b> | 0.000                 | 0.000 | 0.006 |                 |                                  |          |                                   |          |
| <b>Co</b> | 0.000                 | 0.000 | 0.001 | 25.3            | 25.7                             | 102      | 26.5                              | 105      |
| <b>Cr</b> | 0.001                 | 0.007 | 0.033 | 18.5            | 18.2                             | 98       | 19.3                              | 104      |
| <b>Cs</b> | 0.000                 | 0.000 | <LOQ  |                 |                                  |          |                                   |          |
| <b>Cu</b> | 0.003                 | 0.020 | 0.456 | 21.66           | 22.6                             | 104      | 25.1                              | 116      |
| <b>Dy</b> | 0.000                 | 0.000 | 0.000 |                 |                                  |          |                                   |          |
| <b>Er</b> | 0.000                 | 0.000 | <LOQ  |                 |                                  |          |                                   |          |
| <b>Eu</b> | 0.000                 | 0.000 | 0.000 |                 |                                  |          |                                   |          |
| <b>Fe</b> | 0.004                 | 0.033 | 0.452 | 93.44           | 97.8                             | 105      | 102                               | 109      |
| <b>Ga</b> | 0.000                 | 0.001 | 0.005 |                 |                                  |          |                                   |          |
| <b>Gd</b> | 0.000                 | 0.000 | 0.019 |                 |                                  |          |                                   |          |
| <b>Ho</b> | 0.000                 | 0.000 | 0.001 |                 |                                  |          |                                   |          |
| <b>La</b> | 0.000                 | 0.000 | 0.001 |                 |                                  |          |                                   |          |
| <b>Li</b> | 0.000                 | 0.002 | 0.007 | 16.59           | 16.5                             | 99       | 16.9                              | 102      |
| <b>Lu</b> | 0.000                 | 0.000 | <LOQ  |                 |                                  |          |                                   |          |
| <b>Mn</b> | 0.001                 | 0.005 | 0.158 | 37.14           | 35.6                             | 96       | 39.4                              | 106      |
| <b>Mo</b> | 0.000                 | 0.001 | 0.006 | 115.3           | 118                              | 103      | 122                               | 106      |
| <b>Nd</b> | 0.000                 | 0.000 | 0.028 |                 |                                  |          |                                   |          |
| <b>Ni</b> | 0.002                 | 0.014 | 0.102 | 59.8            | 60.6                             | 101      | 62.6                              | 105      |
| <b>Pb</b> | 0.000                 | 0.001 | 0.004 | 18.488          | 17.8                             | 96       | 19.4                              | 105      |
| <b>Pr</b> | 0.000                 | 0.000 | 0.007 |                 |                                  |          |                                   |          |
| <b>Rb</b> | 0.000                 | 0.002 | 0.005 | 12.64           | 12.5                             | 99       | 12.6                              | 100      |
| <b>Re</b> | 0.000                 | 0.000 | <LOD  |                 |                                  |          |                                   |          |
| <b>Sb</b> | 0.000                 | 0.001 | 0.002 | 55.45           | 53.6                             | 97       | 56.2                              | 101      |
| <b>Se</b> | 0.007                 | 0.043 | <LOQ  | 11.7            | 12.1                             | 103      | 12.7                              | 108      |
| <b>Sm</b> | 0.000                 | 0.000 | <LOQ  |                 |                                  |          |                                   |          |
| <b>Sr</b> | 0.001                 | 0.004 | 0.094 | 314             | 314                              | 100      | 326                               | 104      |
| <b>Tb</b> | 0.000                 | 0.000 | 0.000 |                 |                                  |          |                                   |          |
| <b>Th</b> | 0.000                 | 0.000 | <LOQ  |                 |                                  |          |                                   |          |
| <b>Tl</b> | 0.000                 | 0.000 | <LOD  | 6.892           | 6.57                             | 95       | 7.05                              | 102      |
| <b>Tm</b> | 0.000                 | 0.000 | 0.000 |                 |                                  |          |                                   |          |
| <b>U</b>  | 0.000                 | 0.000 | 0.001 |                 |                                  |          |                                   |          |
| <b>V</b>  | 0.000                 | 0.001 | 0.002 | 36.07           | 35.2                             | 98       | 37.0                              | 103      |
| <b>Y</b>  | 0.000                 | 0.000 | 0.001 |                 |                                  |          |                                   |          |
| <b>Yb</b> | 0.000                 | 0.000 | 0.000 |                 |                                  |          |                                   |          |
| <b>Zn</b> | 0.011                 | 0.102 | 2.234 | 74.4            | 79.1                             | 106      | 88.6                              | 119      |

Note: NIST = National Institute of Standards and Technology; SPS = Spectrapure Standards AS.

**Table S2.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRMs) recoveries for concentrations in acid soluble ash (ASA) analyzed using **ICP-MS** (Continued).

|           | SPS-SW2 Surface Water |                                   |          |                                   |          |
|-----------|-----------------------|-----------------------------------|----------|-----------------------------------|----------|
|           | Certified value       | Measured value*100<br>(mean, n=7) | Recovery | Measured value*500<br>(mean, n=7) | Recovery |
|           | µg/L                  | µg/L                              | %        | µg/L                              | %        |
| <b>Ag</b> |                       |                                   |          |                                   |          |
| <b>As</b> | 50                    | 49.8                              | 100      | 52.1                              | 104      |
| <b>Ba</b> | 250                   | 241                               | 97       | 245                               | 98       |
| <b>Be</b> |                       |                                   |          |                                   |          |
| <b>Cd</b> | 2.5                   | 2.49                              | 99       | 2.52                              | 101      |
| <b>Ce</b> | 2.5                   | 2.65                              | 106      | 2.67                              | 107      |
| <b>Co</b> | 10                    | 10.2                              | 102      | 10.1                              | 101      |
| <b>Cr</b> | 10                    | 9.93                              | 99       | 10.4                              | 104      |
| <b>Cs</b> |                       |                                   |          |                                   |          |
| <b>Cu</b> | 100                   | 107                               | 108      | 129                               | 130      |
| <b>Dy</b> | 2.5                   | 2.57                              | 103      | 2.54                              | 102      |
| <b>Er</b> | 2.5                   | 2.59                              | 104      | 2.62                              | 105      |
| <b>Eu</b> | 2.5                   | 2.59                              | 104      | 2.58                              | 103      |
| <b>Fe</b> | 100                   | 104                               | 104      | 107                               | 107      |
| <b>Ga</b> |                       |                                   |          |                                   |          |
| <b>Gd</b> | 2.5                   | 2.59                              | 104      | 2.56                              | 102      |
| <b>Ho</b> | 2.5                   | 2.54                              | 101      | 2.51                              | 100      |
| <b>La</b> | 2.5                   | 2.58                              | 103      | 2.57                              | 103      |
| <b>Li</b> |                       |                                   |          |                                   |          |
| <b>Lu</b> | 2.5                   | 2.51                              | 100      | 2.45                              | 98       |
| <b>Mn</b> | 50                    | 49.6                              | 99       | 56.4                              | 113      |
| <b>Mo</b> | 50                    | 50.4                              | 101      | 52.1                              | 104      |
| <b>Nd</b> | 2.5                   | 2.60                              | 104      | 2.56                              | 102      |
| <b>Ni</b> | 50                    | 51.5                              | 103      | 51.5                              | 103      |
| <b>Pb</b> | 25                    | 24.3                              | 97       | 24.1                              | 97       |
| <b>Pr</b> | 2.5                   | 2.60                              | 104      | 2.60                              | 104      |
| <b>Rb</b> | 50                    | 47.8                              | 96       | 46.8                              | 94       |
| <b>Re</b> |                       |                                   |          |                                   |          |
| <b>Sb</b> |                       |                                   |          |                                   |          |
| <b>Se</b> | 10                    | 15.3                              | 152      | 18.8                              | 188      |
| <b>Sm</b> | 2.5                   | 2.70                              | 108      | 2.65                              | 106      |
| <b>Sr</b> | 250                   | 246                               | 99       | 252                               | 101      |
| <b>Tb</b> | 2.5                   | 2.57                              | 103      | 2.54                              | 102      |
| <b>Th</b> | 2.5                   | 2.59                              | 104      | 2.51                              | 100      |
| <b>Tl</b> | 2.5                   | 2.42                              | 97       | 2.40                              | 96       |
| <b>Tm</b> | 2.5                   | 2.55                              | 102      | 2.55                              | 102      |
| <b>U</b>  | 2.5                   | 2.46                              | 98       | 2.58                              | 103      |
| <b>V</b>  | 50                    | 48.8                              | 98       | 48.9                              | 98       |
| <b>Y</b>  | 2.5                   | 2.70                              | 108      | 2.66                              | 107      |
| <b>Yb</b> | 2.5                   | 2.53                              | 101      | 2.53                              | 101      |
| <b>Zn</b> | 100                   | 120                               | 121      | 133                               | 134      |

Note: NIST = National Institute of Standards and Technology; SPS = Spectrapure Standards AS.

**Table S3.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRMs) recoveries for **total** concentrations analyzed using **ICP-OES**.

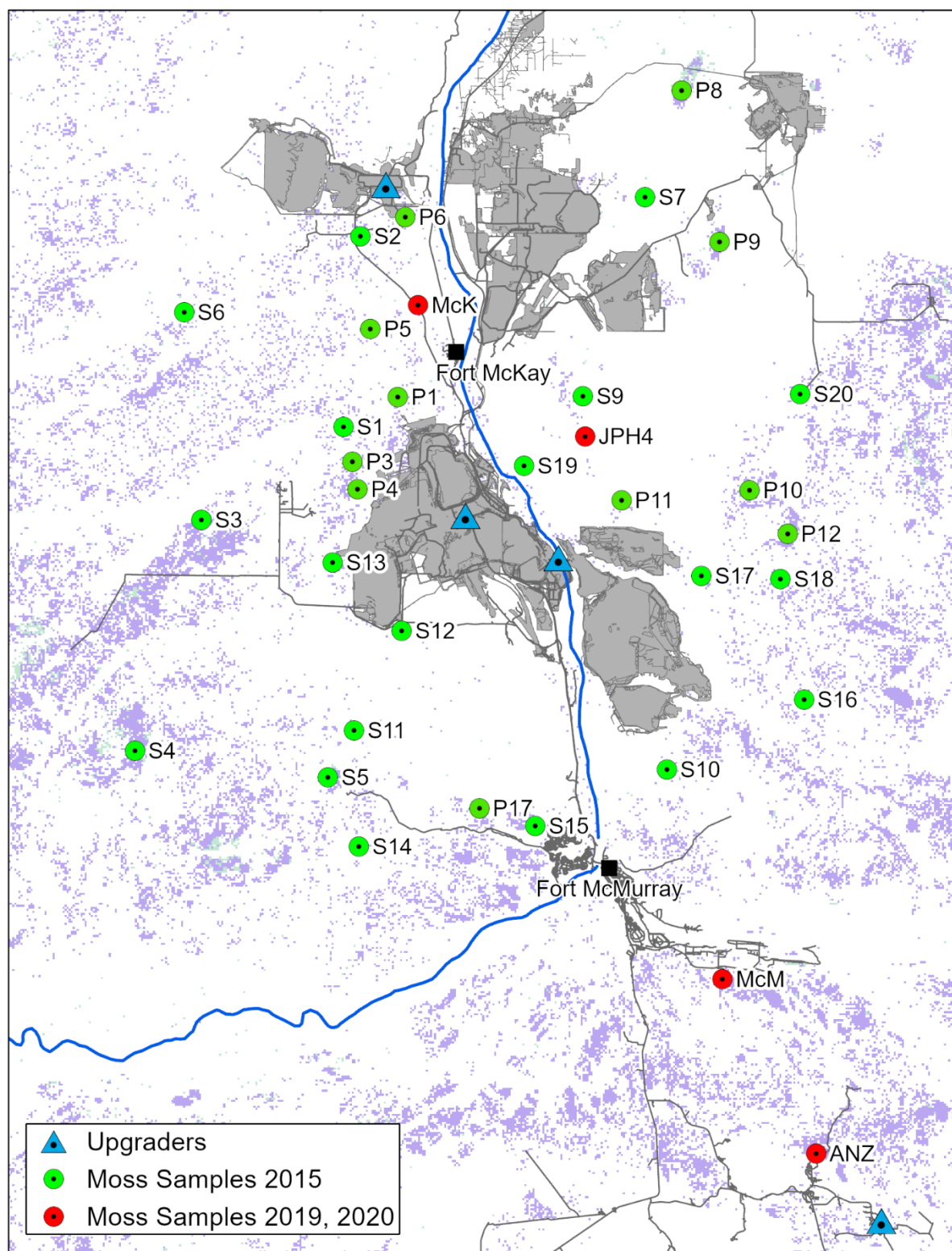
|           | NIST 1515 Apple Leaves |       |       |                 |                            |          | FFRI-Moss-1343    |                            |          |
|-----------|------------------------|-------|-------|-----------------|----------------------------|----------|-------------------|----------------------------|----------|
|           | LOD                    | LOQ   | MDL   | Certified value | Measured value (mean, n=6) | Recovery | Recommended value | Measured value (mean, n=6) | Recovery |
|           | mg/L                   | mg/L  | mg/L  | mg/kg           | mg/kg                      | %        | mg/kg             | mg/kg                      | %        |
| <b>Al</b> | 0.004                  | 0.013 | <LOD  | 284.5           | 232                        | 81       | 169               | 224                        | 132      |
| <b>Ca</b> | 0.004                  | 0.012 | <LOD  | 15250           | 12700                      | 83       | 1920              | 1830                       | 95       |
| <b>Fe</b> | 0.001                  | 0.005 | <LOD  | 82.7            | 68.8                       | 83       | 138               | 150                        | 109      |
| <b>K</b>  | 0.083                  | 0.264 | <LOD  | 16080           | 13700                      | 85       | 4510              | 4700                       | 104      |
| <b>Mg</b> | 0.005                  | 0.018 | <LOD  | 2710            | 2340                       | 86       | 755               | 719                        | 95       |
| <b>Mn</b> | 0.001                  | 0.001 | 0.444 | 54.1            | 50.7                       | 94       | 535               | 466                        | 87       |
| <b>P</b>  | 0.012                  | 0.035 | <LOD  | 1593            | 1360                       | 86       | 1050              | 911                        | 87       |
| <b>S</b>  | 0.009                  | 0.012 | <LOD  |                 | 1630                       |          | 830               | 789                        | 95       |

Note: NIST = National Institute of Standards and Technology; NRCC = National Research Council of Canada.

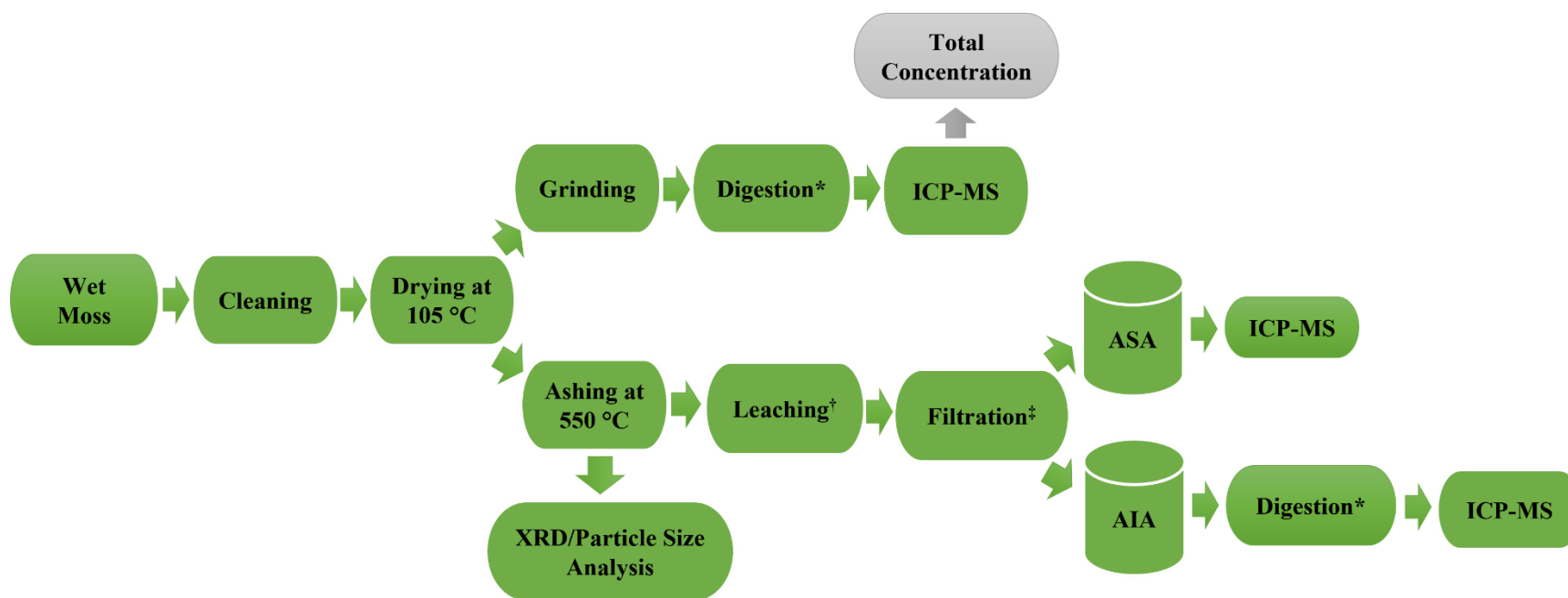
**Table S4.** Limit of detection (LOD), limit of quantification (LOQ), method detection limit (MDL), and standard reference materials (SRM) recoveries for concentrations in **acid soluble ash** (ASA) analyzed using **ICP-OES**.

|    | NIST 1643f Freshwater |       |       |                 |                                  |          | NRCC SLRS-6 River Water |                                  |          |
|----|-----------------------|-------|-------|-----------------|----------------------------------|----------|-------------------------|----------------------------------|----------|
|    | LOD                   | LOQ   | MDL   | Certified value | Measured value*50<br>(mean, n=2) | Recovery | Certified value         | Measured value*50<br>(mean, n=2) | Recovery |
|    | mg/L                  | mg/L  | mg/L  | mg/L            | mg/L                             | %        | mg/L                    | mg/L                             | %        |
| Al | 0.004                 | 0.013 | <LOD  | 0.13            | 0.145                            | 109      | 0.03                    | <LOQ                             |          |
| Ca | 0.004                 | 0.012 | <LOD  | 29.14           | 28.5                             | 98       | 8.76                    | 8.33                             | 95       |
| Fe | 0.001                 | 0.005 | <LOD  | 0.09            | 0.0975                           | 105      | 0.08                    | 0.0850                           | 101      |
| K  | 0.083                 | 0.264 | <LOD  | 1.91            | 1.84                             | 96       | 0.65                    | <LOQ                             |          |
| Mg | 0.005                 | 0.018 | <LOD  | 7.38            | 7.63                             | 103      | 2.13                    | 2.19                             | 103      |
| Mn | 0.001                 | 0.001 | <LOD  | 0.04            | 0.0400                           | 109      | 0.00                    | <LOD                             |          |
| P  | 0.012                 | 0.035 | <LOD  |                 |                                  |          |                         |                                  |          |
| S  | 0.009                 | 0.012 | 0.187 |                 |                                  |          |                         | 1.76                             |          |

Note: NIST = National Institute of Standards and Technology; NRCC = National Research Council of Canada.



**Fig. S1.** Moss sampling sites within the Athabasca Bituminous Sands Regions in fall 2015, 2019, and 2020.



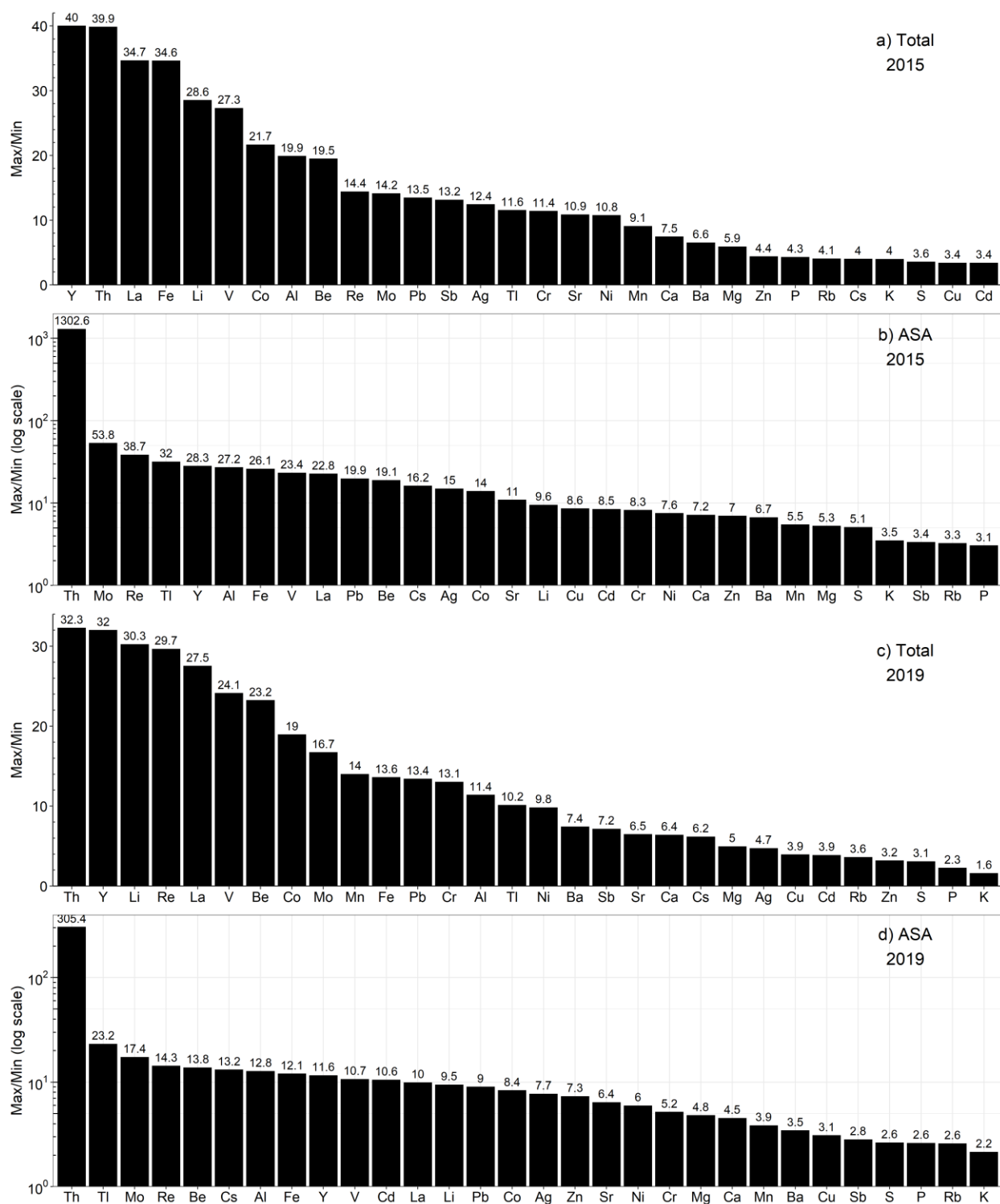
**Fig. S2.** Schematic flow of sample treatments.

\*Digestion: Reaction with concentrated  $\text{HNO}_3$  (sub-boiled twice) and  $\text{HBF}_4$  for 80 min in a microwave under high temperature and pressure.

†Leaching: Reaction with 2%  $\text{HNO}_3$  for 15 min under room temperature and pressure.

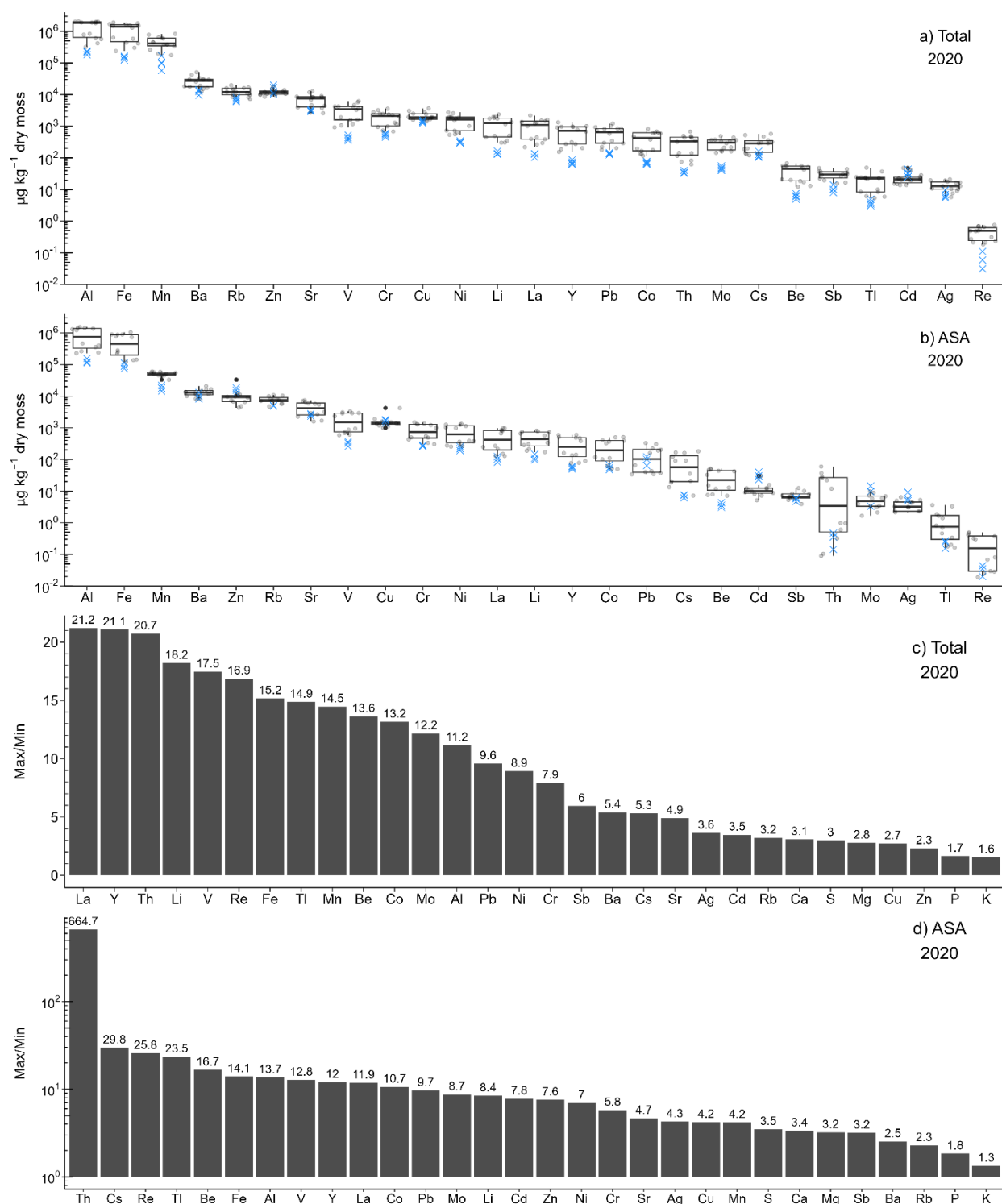
‡Filtration: Polypropylene syringes and 0.45  $\mu\text{m}$  polytetrafluoroethylene filter membranes, with polypropylene filter holders and silicone gaskets for sealing. All were acid-cleaned before use.

Note: ASA = Acid Soluble Ash. AIA = Acid Insoluble Ash.

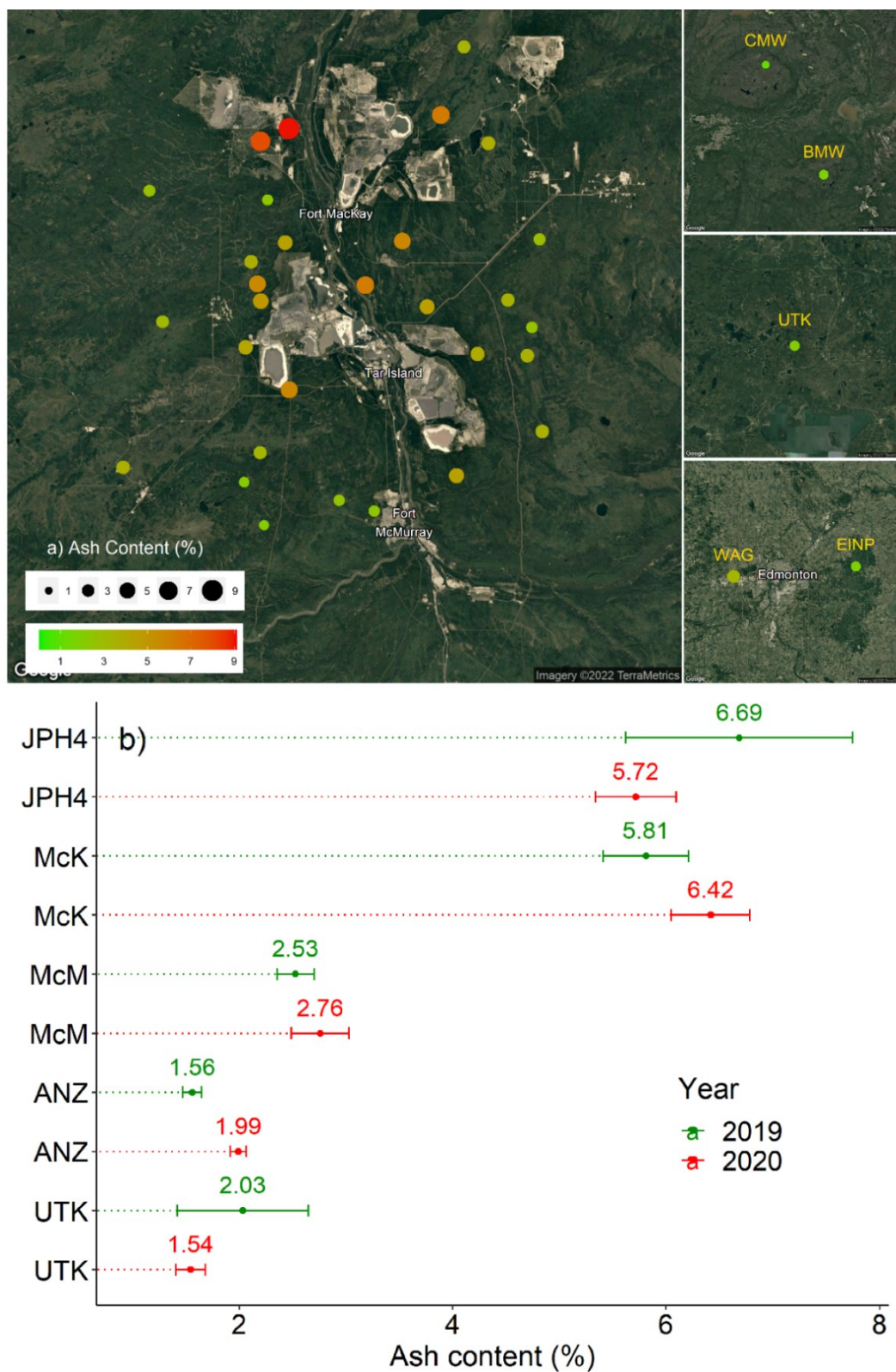


**Fig. S3.** Ratios of maximum (Max) to minimum (Min) element concentrations out of bulk moss and acid soluble ash (ASA) of *Sphagnum* mosses collected in fall 2015 (sites EINP and WAG were excluded) and 2019.



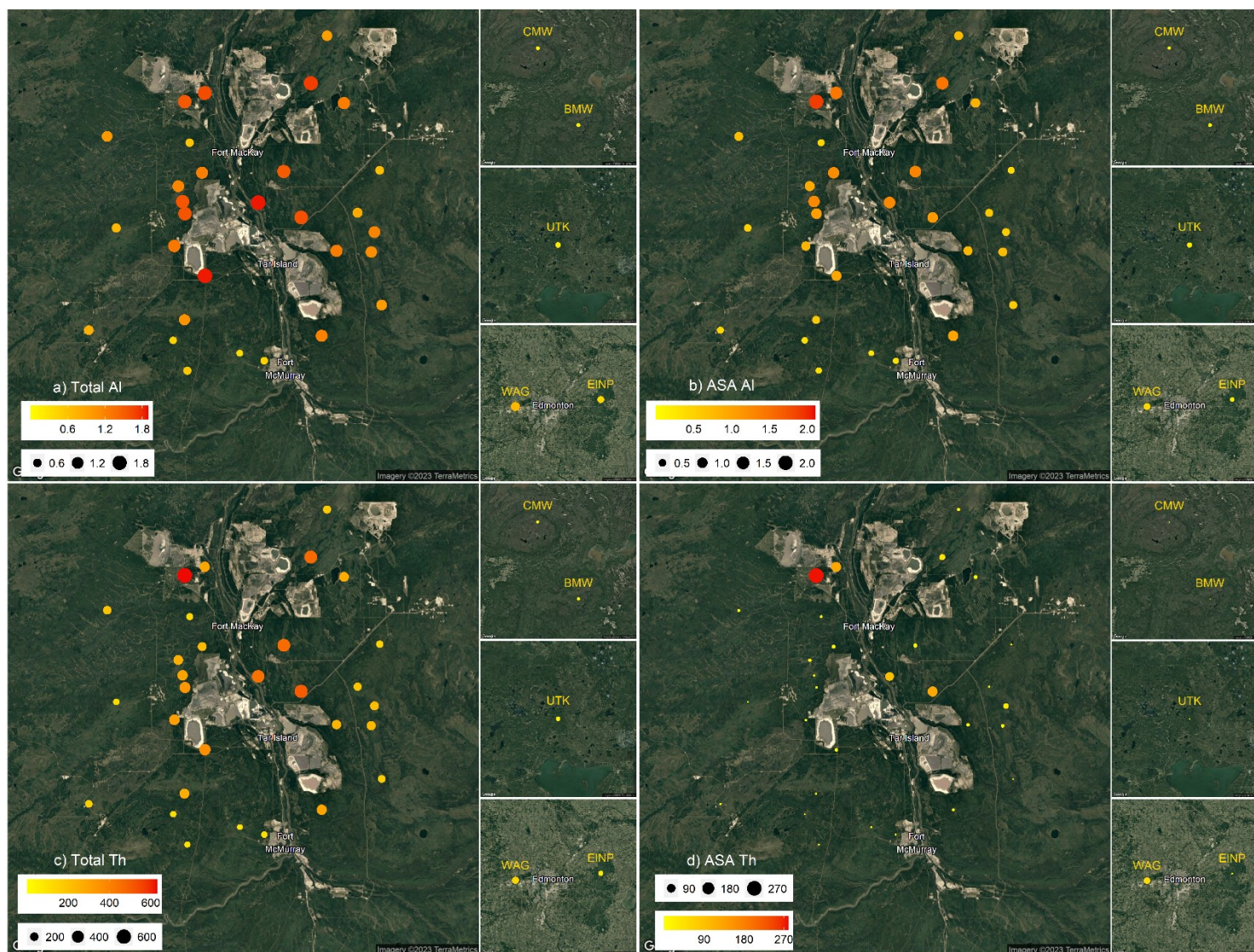


**Fig. S4.** Box plots of element concentrations and element ratios for samples collected in fall 2020. a) and b) Boxplots of elemental concentrations for bulk moss (i.e. total) and acid soluble ash (ASA) at industrial sites. Elemental concentrations in samples from the reference site (UTK) are indicated in blue using the multiplication sign “x”. All values represent concentrations ( $\mu\text{g kg}^{-1}$ ) in dry moss. c) and d) Ratios of maximum (Max) to minimum (Min) elemental concentrations for bulk moss and ASA.



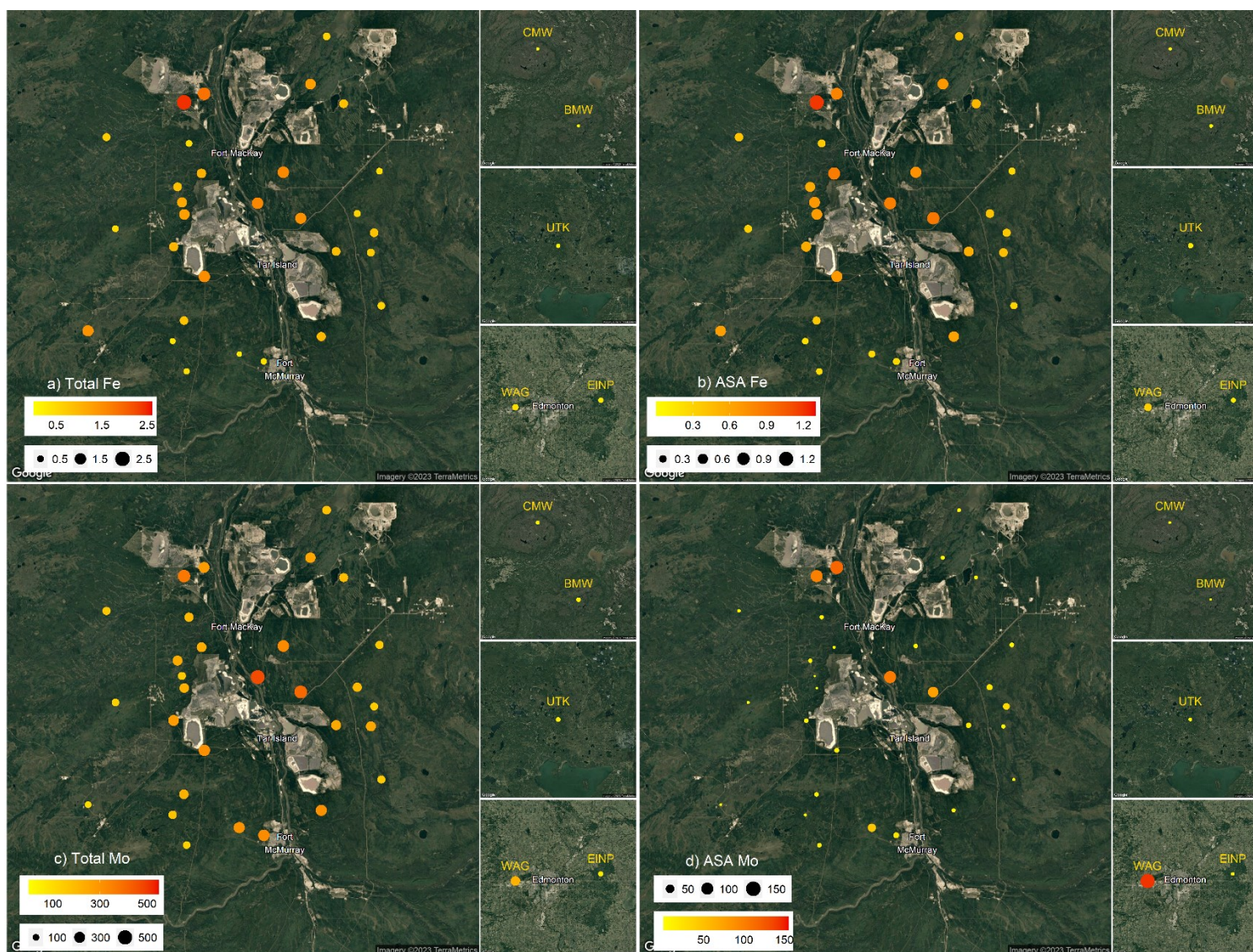
**Fig. S5.** Ash content (%) of *Sphagnum* mosses collected in fall a) 2015 and b) 2019 and 2020. CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park, McK = Fort McKay, McM = Fort McMurray, ANZ = Anzac.



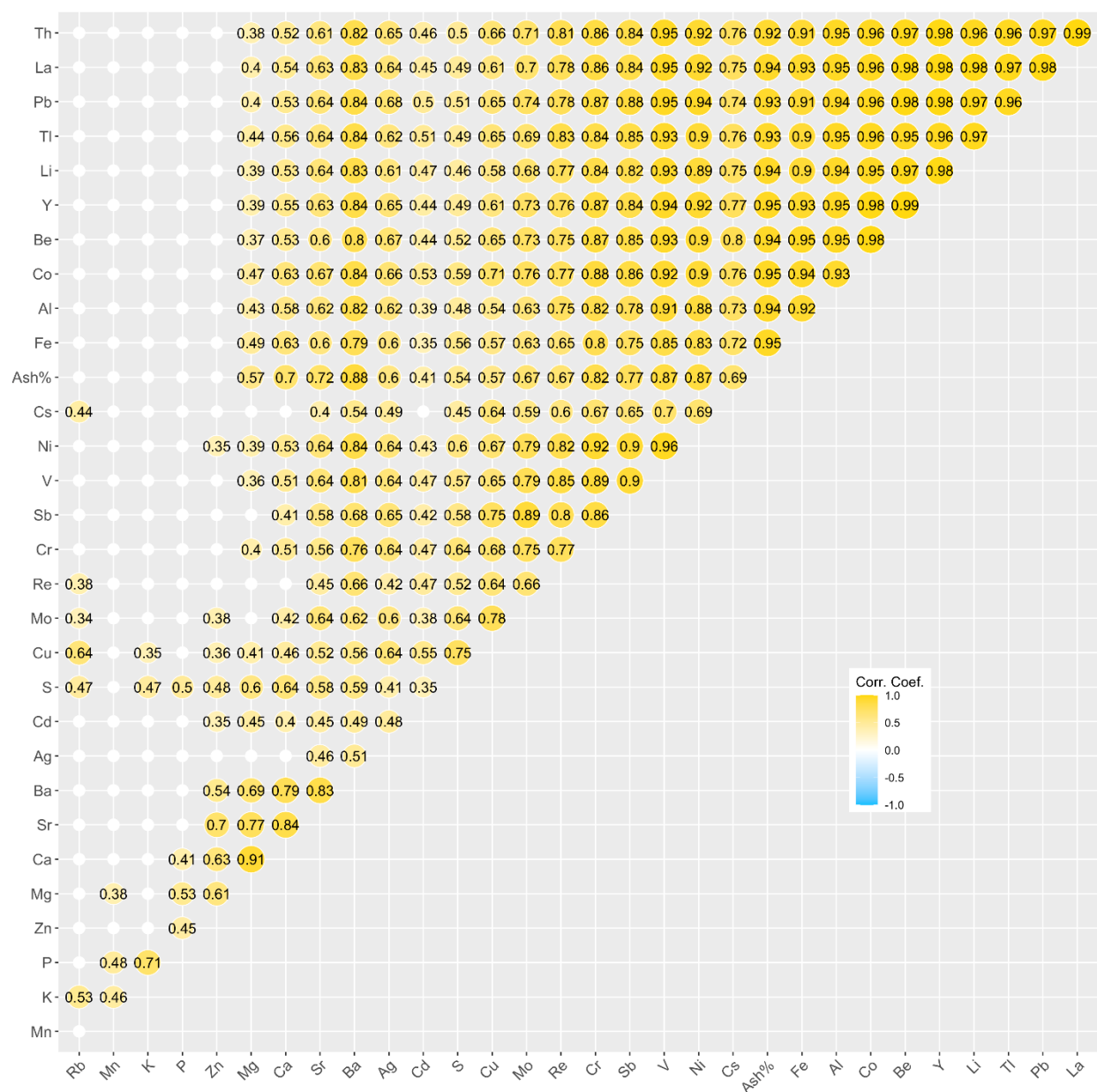


**Fig. S6.** Spatial distribution of a) total and b) acid soluble **Al** ( $\text{g kg}^{-1}$  dry moss), and c) total and d) acid soluble **Th** ( $\mu\text{g kg}^{-1}$  dry moss) concentrations in *Sphagnum* mosses collected in fall 2015. ASA = Acid Soluble Ash, CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.



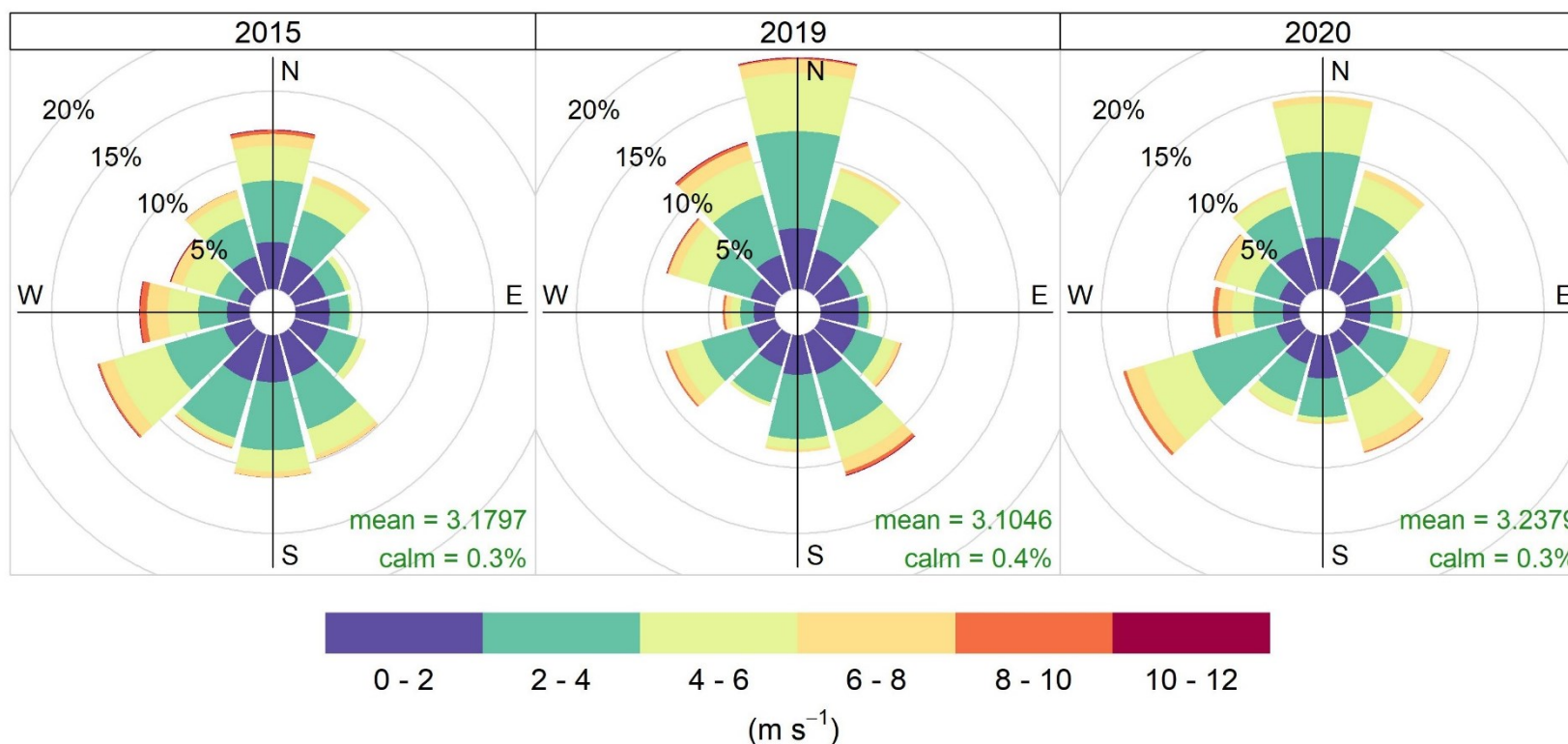


**Fig. S7.** Spatial distribution of a) total and b) acid soluble **Fe** ( $\text{mg kg}^{-1}$  dry moss), and c) total and d) acid soluble **Mo** ( $\mu\text{g kg}^{-1}$  dry moss) concentrations in *Sphagnum* mosses collected in fall 2015. ASA = Acid Soluble Ash, CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.



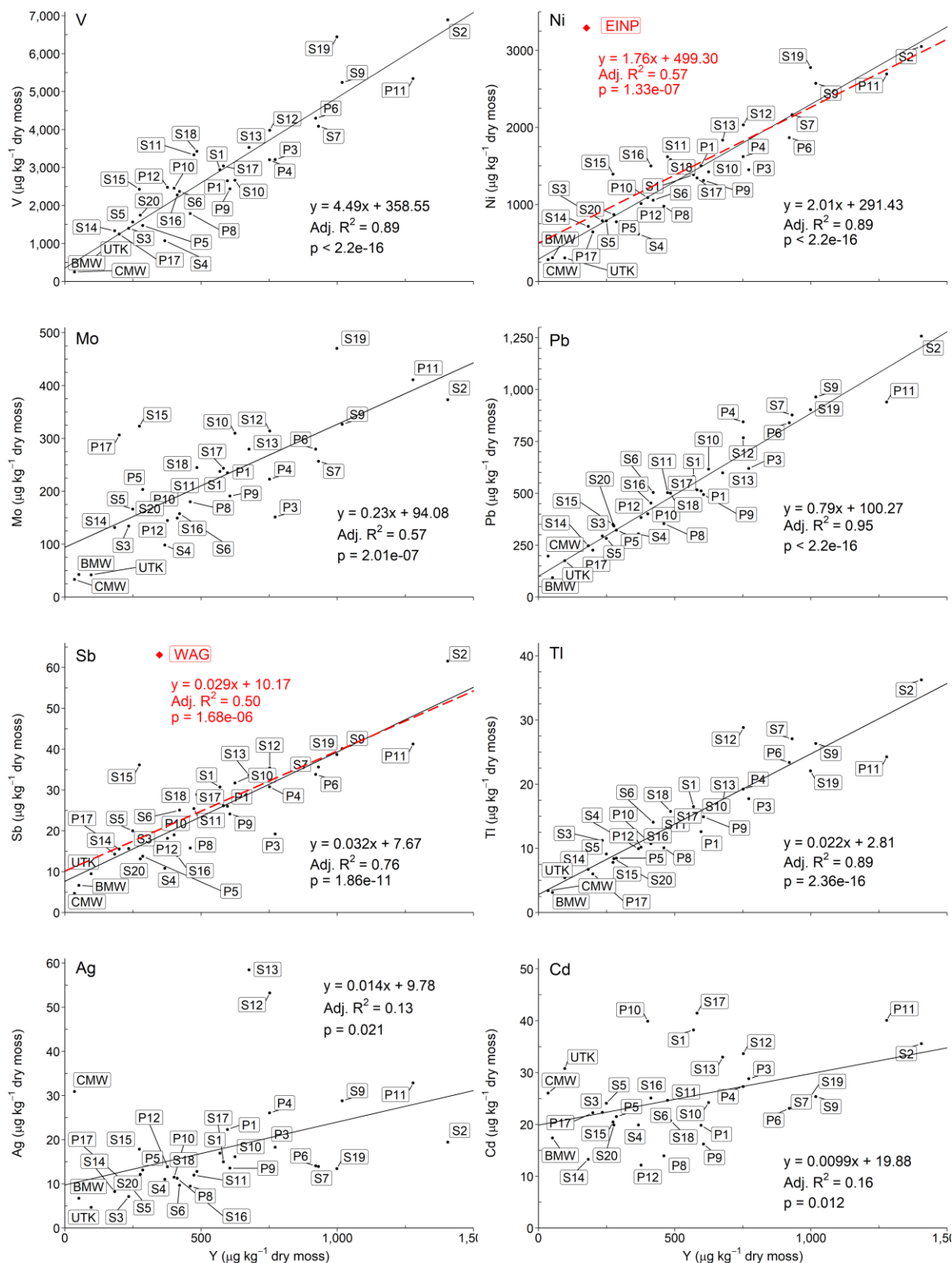
**Fig. S8.** Correlogram of **total** concentrations and ash content of fall 2015 *Sphagnum* mosses based on the Spearman rank-based correlation test. Sites EINP and WAG were excluded. Corr. Coef. = Correlation coefficient. Significance level = 0.05. Only significant correlations are presented.





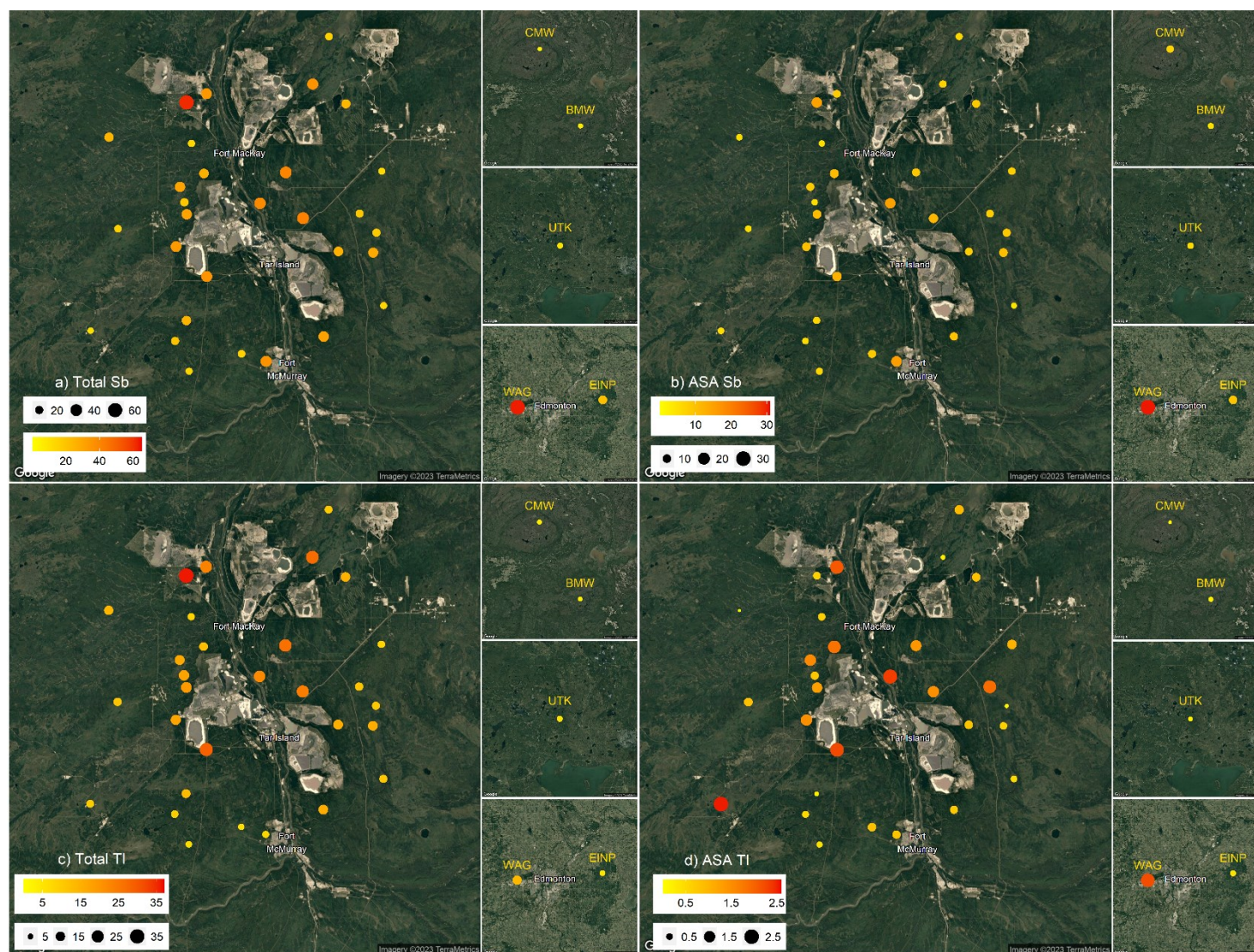
### Frequency of counts by wind direction (%)

**Fig. S9.** Wind rose diagrams for 2015, 2019, and 2020 at Mildred Station. Historic hourly weather data from May 1 to August 31 of each year was downloaded from the Government of Canada climate station: Mildred Lake (Latitude: 57°02'28.000" N, Longitude: 111°33'32.000" W, Elevation: 310.00 m, Climate Identifier (ID): 3064528, World Meteorological Organization (WMO) ID: 71255, Transport Canada (TC) ID: WMX). Data was processed in R 4.1.1,<sup>2</sup> and visualized using the *openair* package.<sup>3</sup> Wind rose diagram for 2015 was also used in our previous study by Mullan-Boudreau et al.<sup>4</sup>



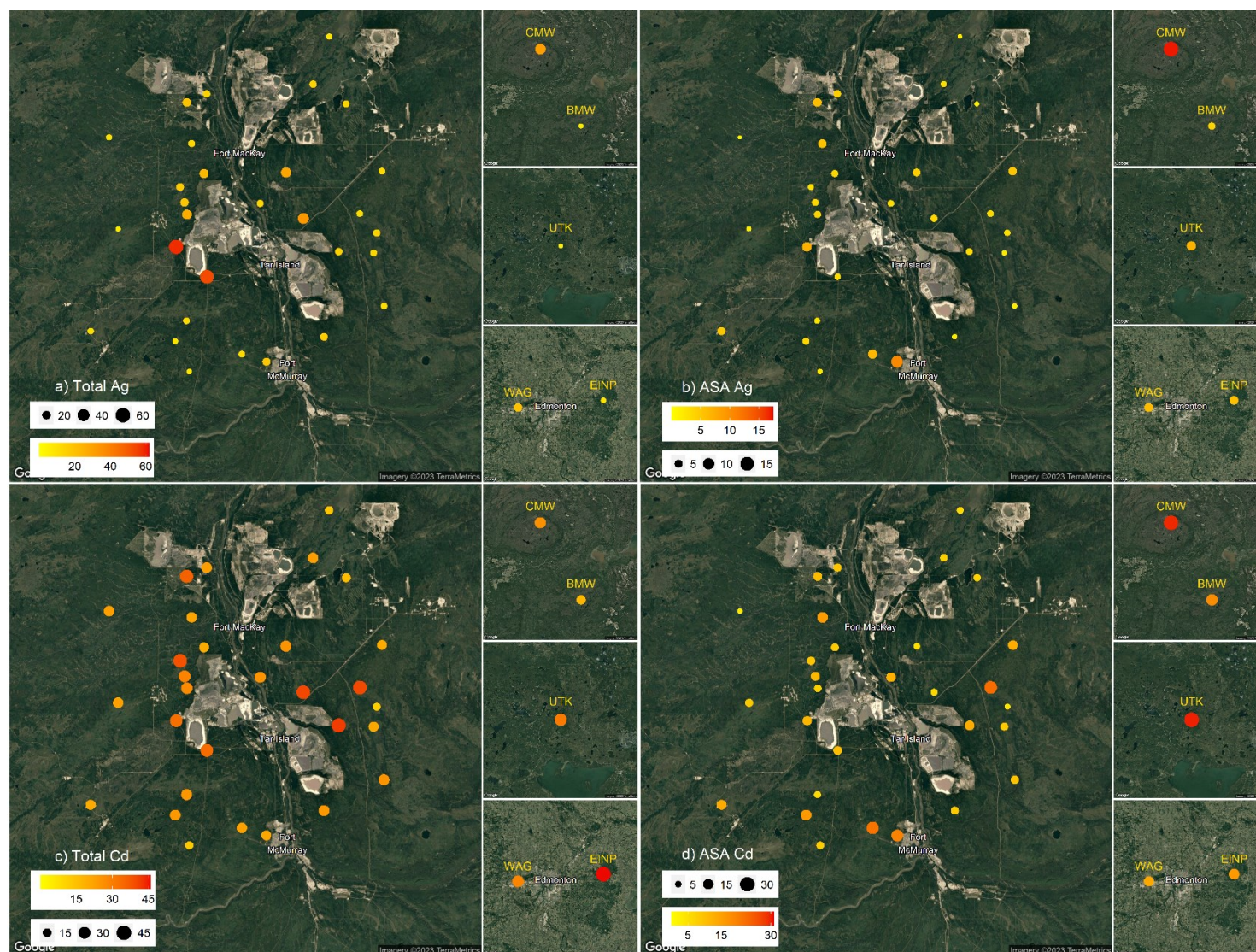
**Fig. S10.** Total V, Ni, Mo, Pb, Sb, Tl, Ag, and Cd concentration versus total Y concentration ( $\mu\text{g kg}^{-1}$  dry moss) in *Sphagnum* mosses collected in fall 2015. CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.





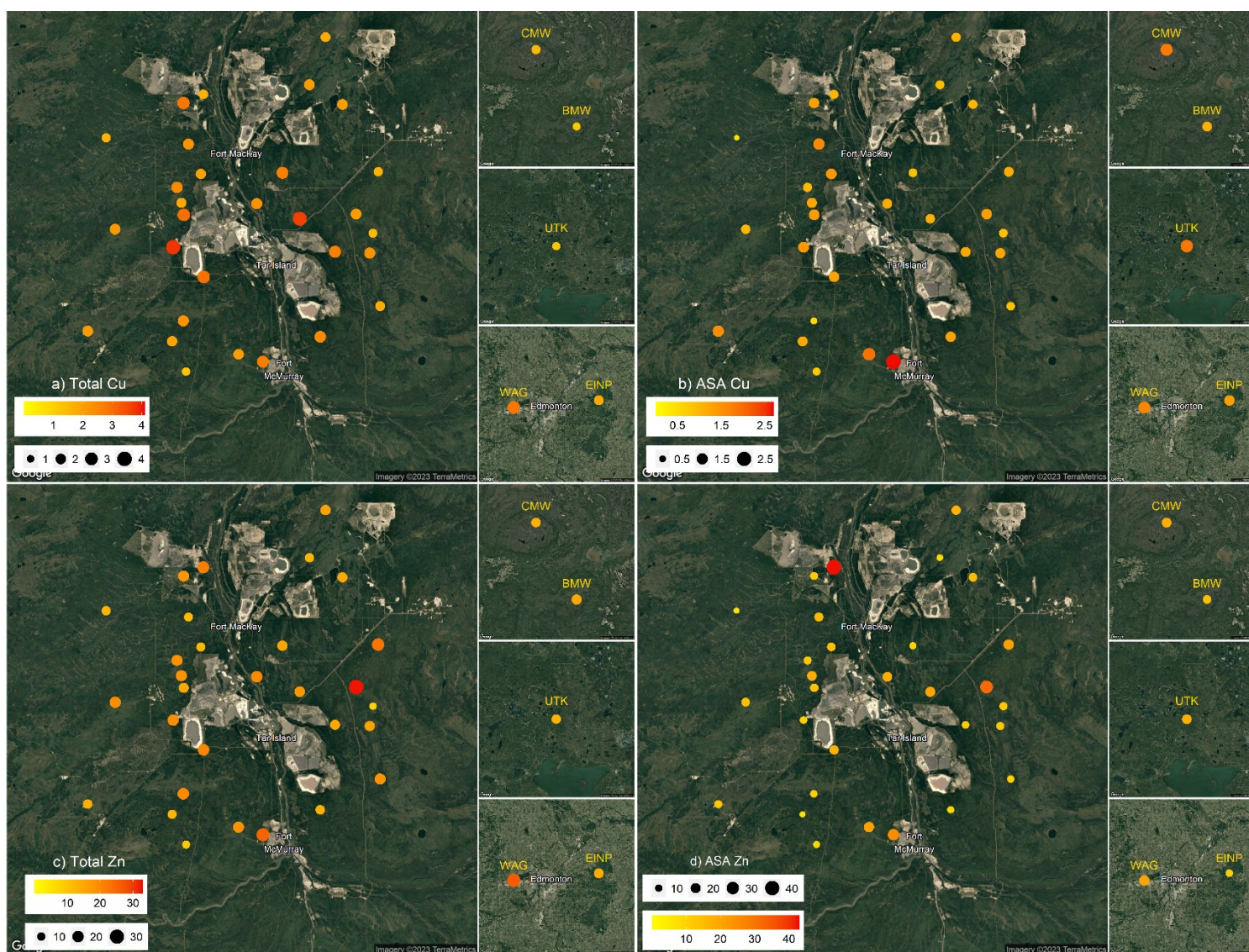
**Fig. S11.** Spatial distribution of a) total and b) acid soluble **Sb**, and c) total and d) acid soluble **TI** concentrations ( $\mu\text{g kg}^{-1}$  dry moss) in *Sphagnum* mosses collected in fall 2015. ASA = Acid Soluble Ash, CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.



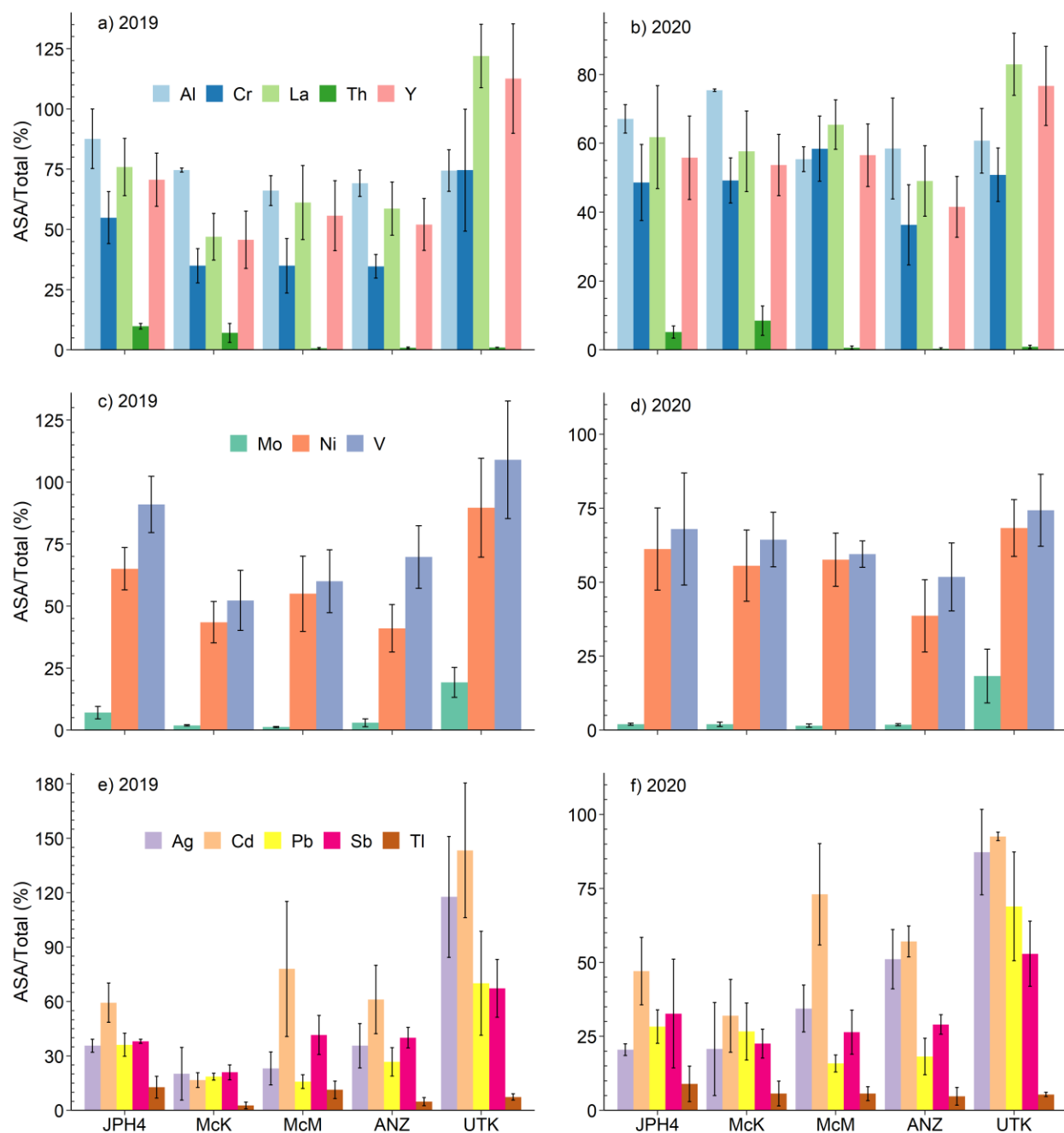


**Fig. S12.** Spatial distribution of a) total and b) acid soluble **Ag**, and c) total and d) acid soluble **Cd** concentrations ( $\mu\text{g kg}^{-1}$  dry moss) in *Sphagnum* mosses collected in fall 2015. ASA = Acid Soluble Ash, CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.

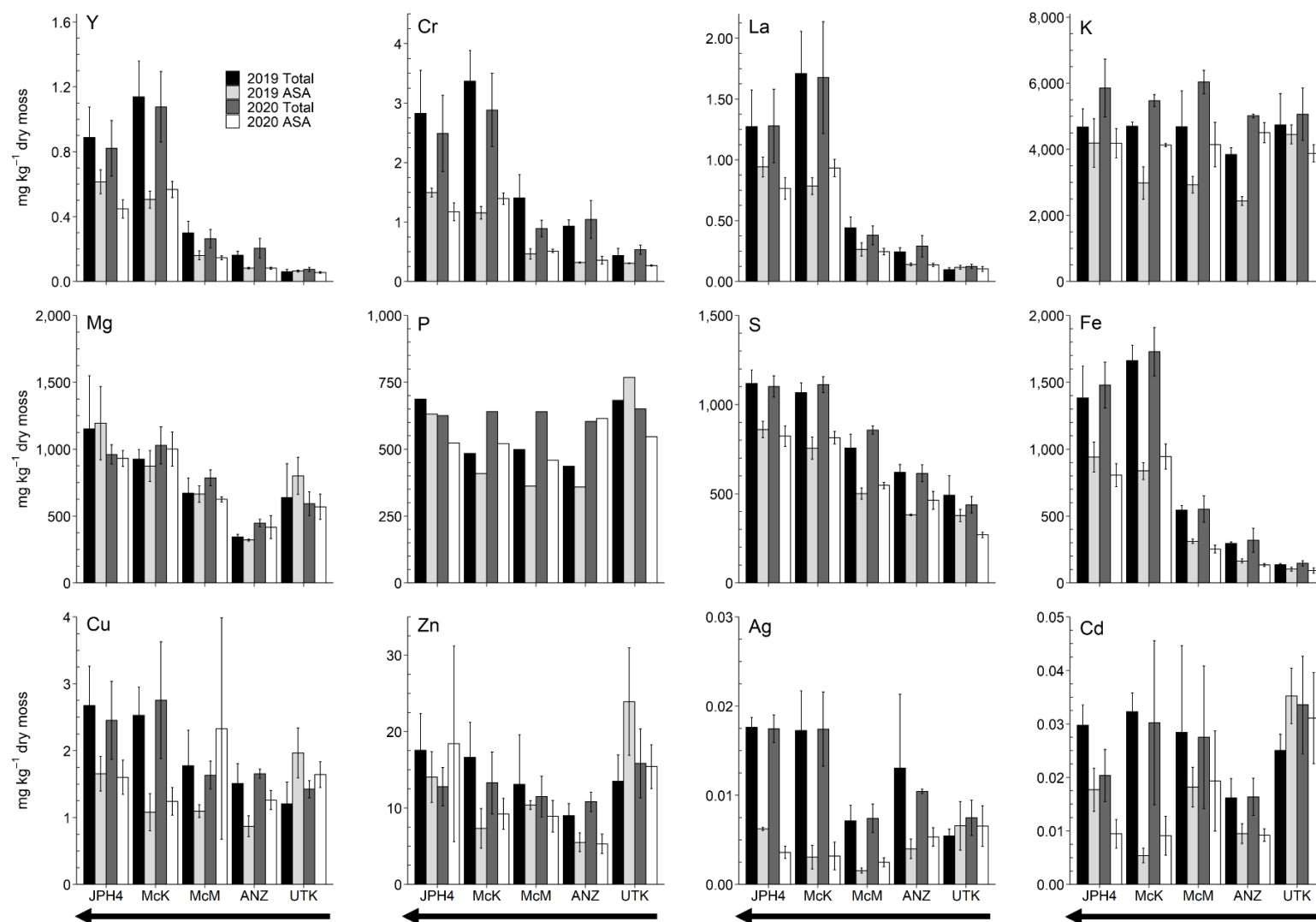




**Fig. S13.** Spatial distribution of a) total and b) acid soluble **Cu**, and c) total and d) acid soluble **Zn** concentrations ( $\text{mg kg}^{-1}$  dry moss) in *Sphagnum* mosses collected in fall 2015. ASA = Acid Soluble Ash, CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.

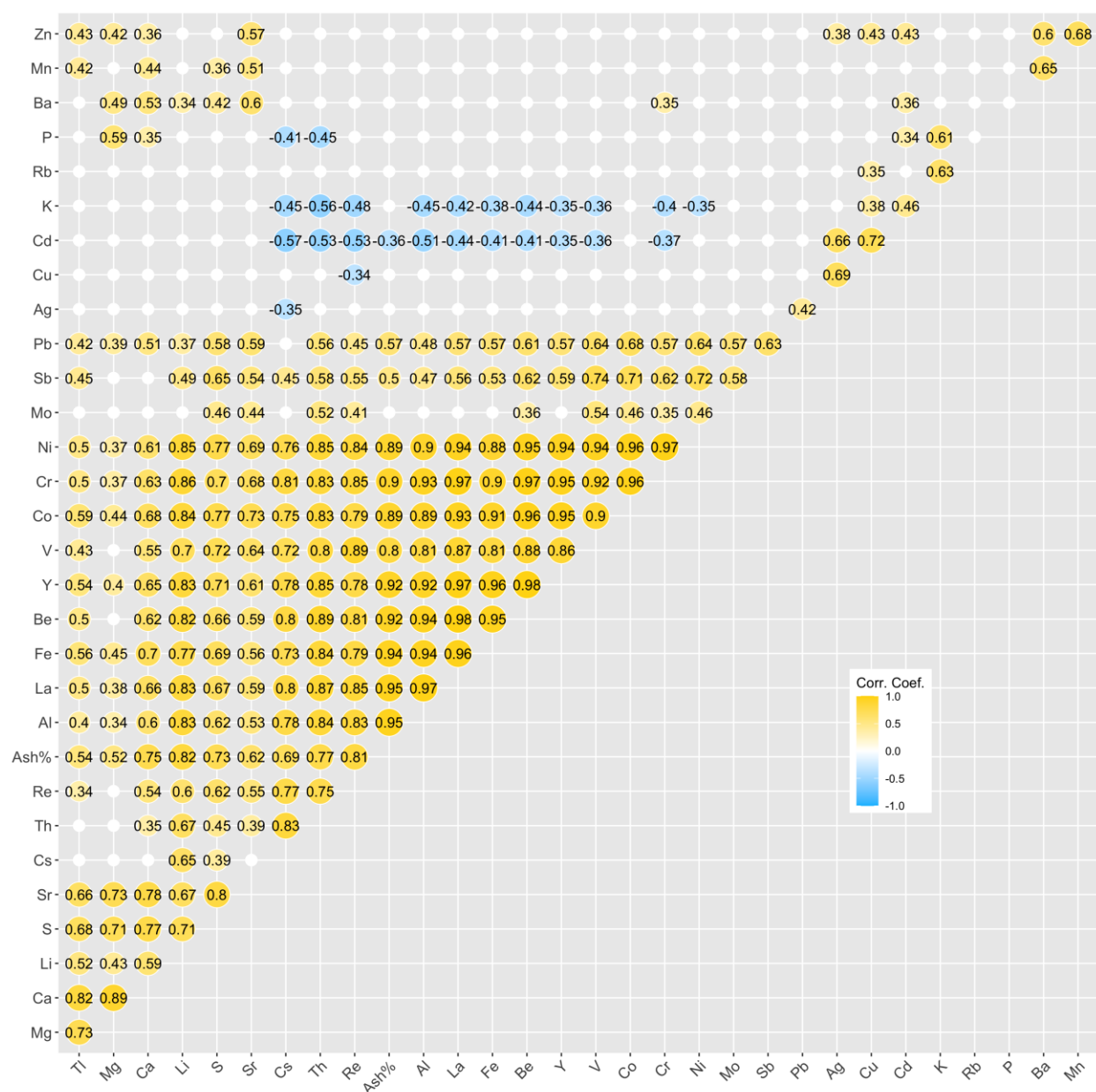


**Fig. S14.** Proportion of acid soluble to total concentration of trace elements in *Sphagnum* mosses collected in fall 2019 (a, c, & e) and 2020 (b, d, & f). ASA = Acid Soluble Ash, McK = Fort McKay, McM = Fort McMurray, ANZ = Anzac, UTK = Utikuma. Error bars represent  $\pm$  standard deviation.

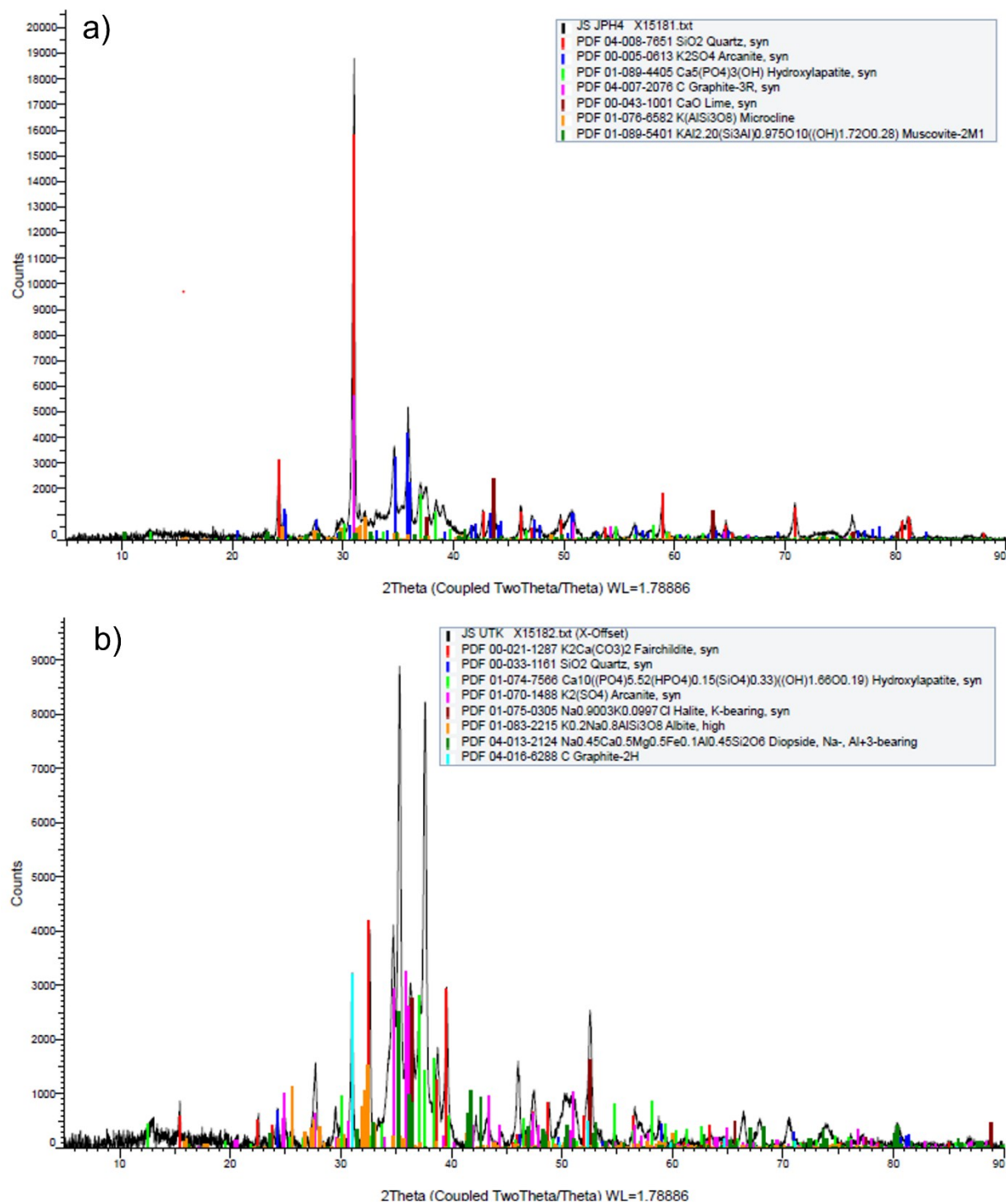


**Fig. S15.** Total and acid soluble concentrations (mg kg<sup>-1</sup> dry moss) of conservative lithophile elements (Y, Cr, La), essential elements (K, Mg, P, S, Fe, Cu, Zn), and potentially toxic elements (Ag, Cd) in *Sphagnum* mosses collected in fall 2019 and 2020. ASA = Acid Soluble Ash, McK = Fort McKay, McM = Fort McMurray, ANZ = Anzac, UTK = Utikuma. The arrow points from the reference site (UTK) toward industry. Error bars represent  $\pm$  standard deviation.





**Fig. S16.** Correlogram of element concentrations in Acid Soluble Ash (ASA) and ash content of fall 2015 *Sphagnum* mosses based on the Spearman rank-based correlation test. Sites EINP and WAG were excluded. Corr. Coef. = Correlation coefficient. Significance level = 0.05. Only significant correlations are presented.



**Fig. S17.** X-ray diffraction (XRD) analysis of a) JPH4 and b) UTK ash samples (both from 2020). UTK = Utikuma.

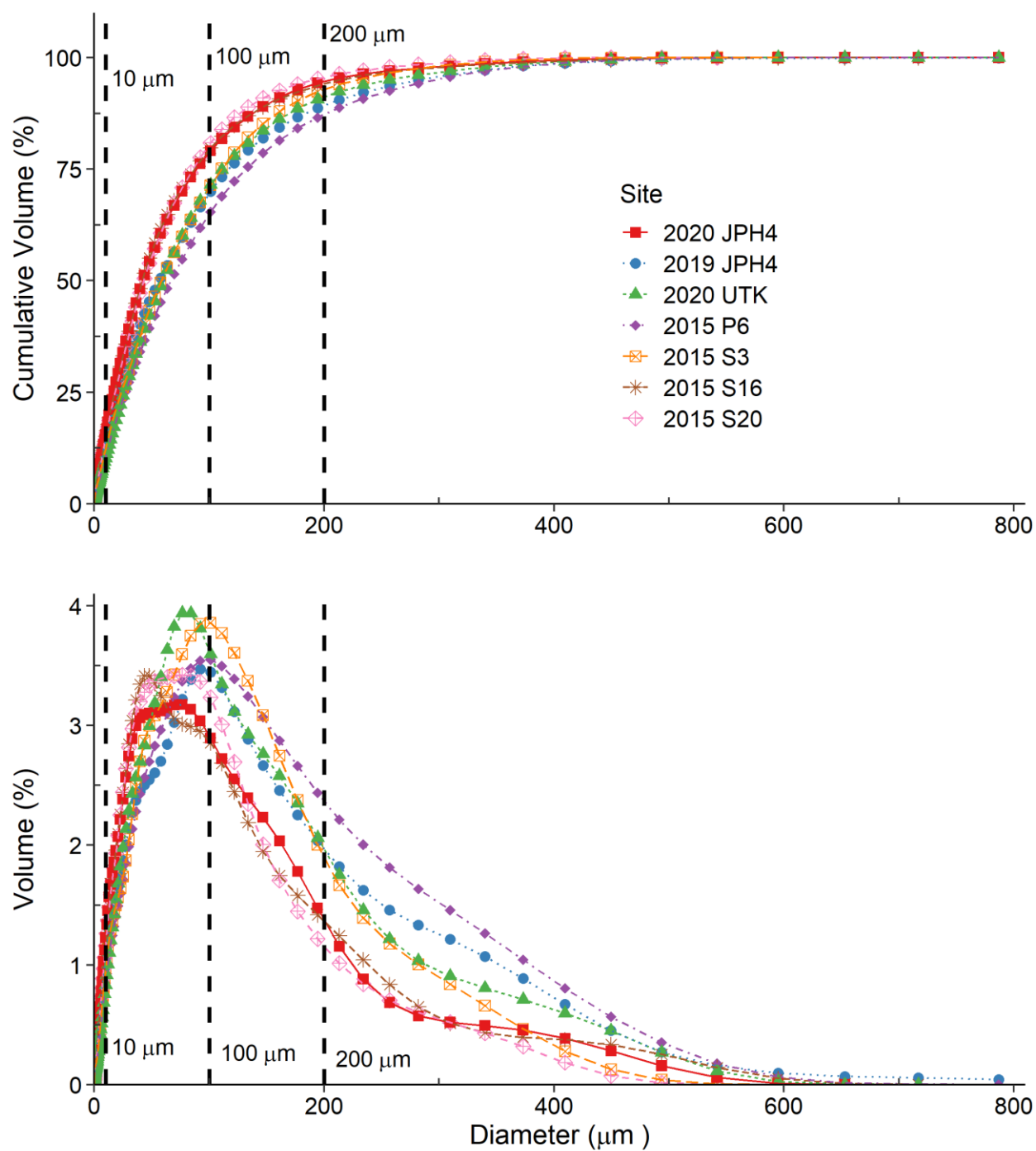
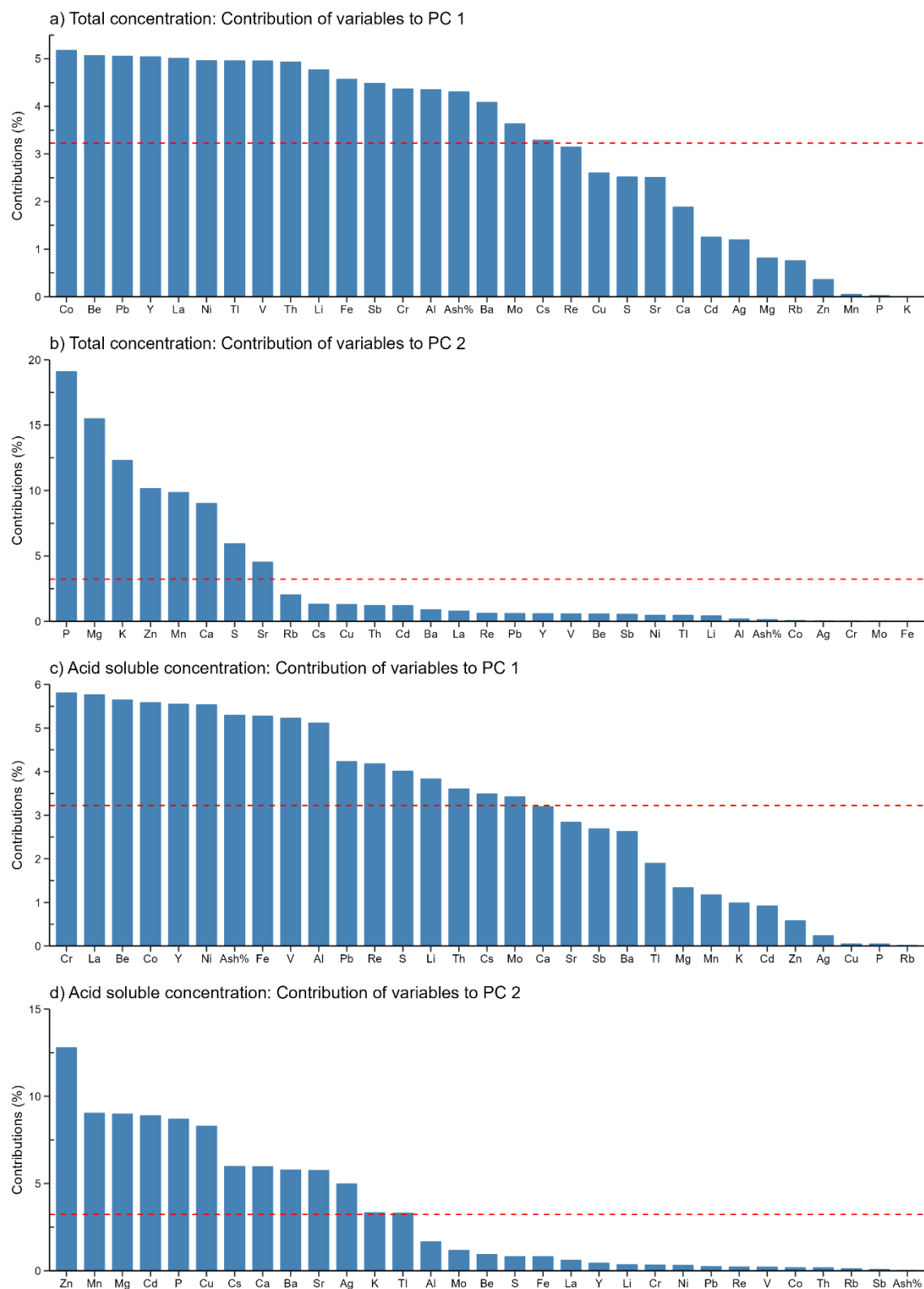
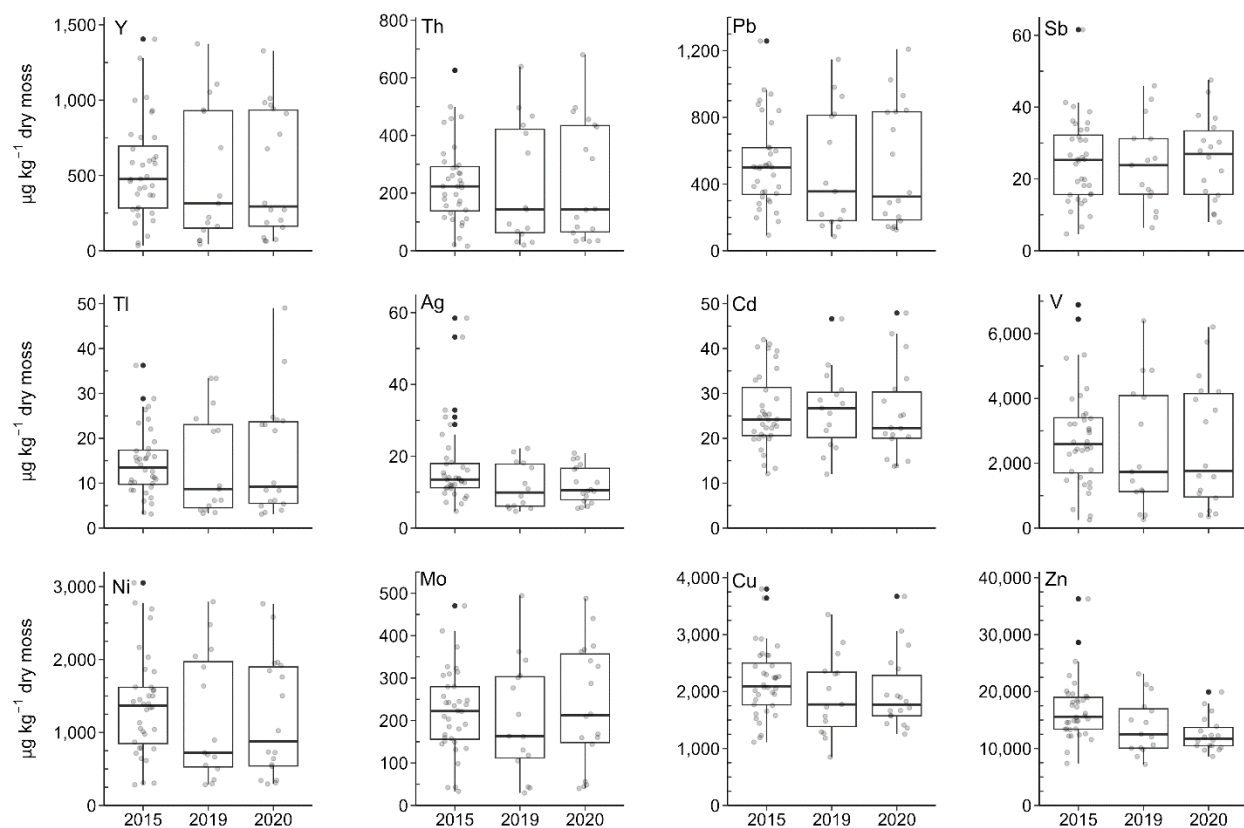


Fig. S18. Particle size distribution of ash samples of *Sphagnum* mosses by laser diffraction. UTK = Utikuma.

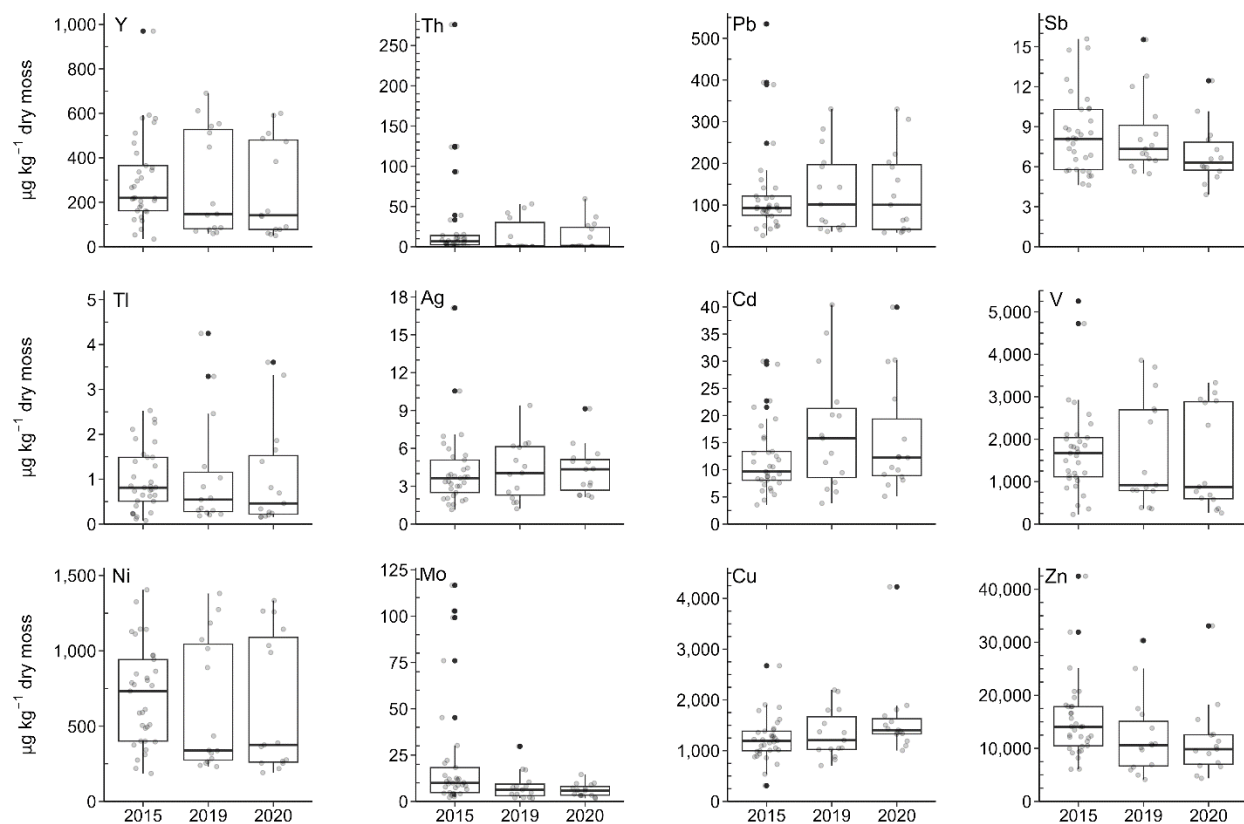


**Fig. S19.** Contributions of variables in accounting for the variability in principal component (PC) 1 and PC 2 from principal component analysis of fall 2015 *Sphagnum* mosses using total concentrations (a, b) and acid soluble concentrations (c, d). The red dashed line indicates the average contribution.

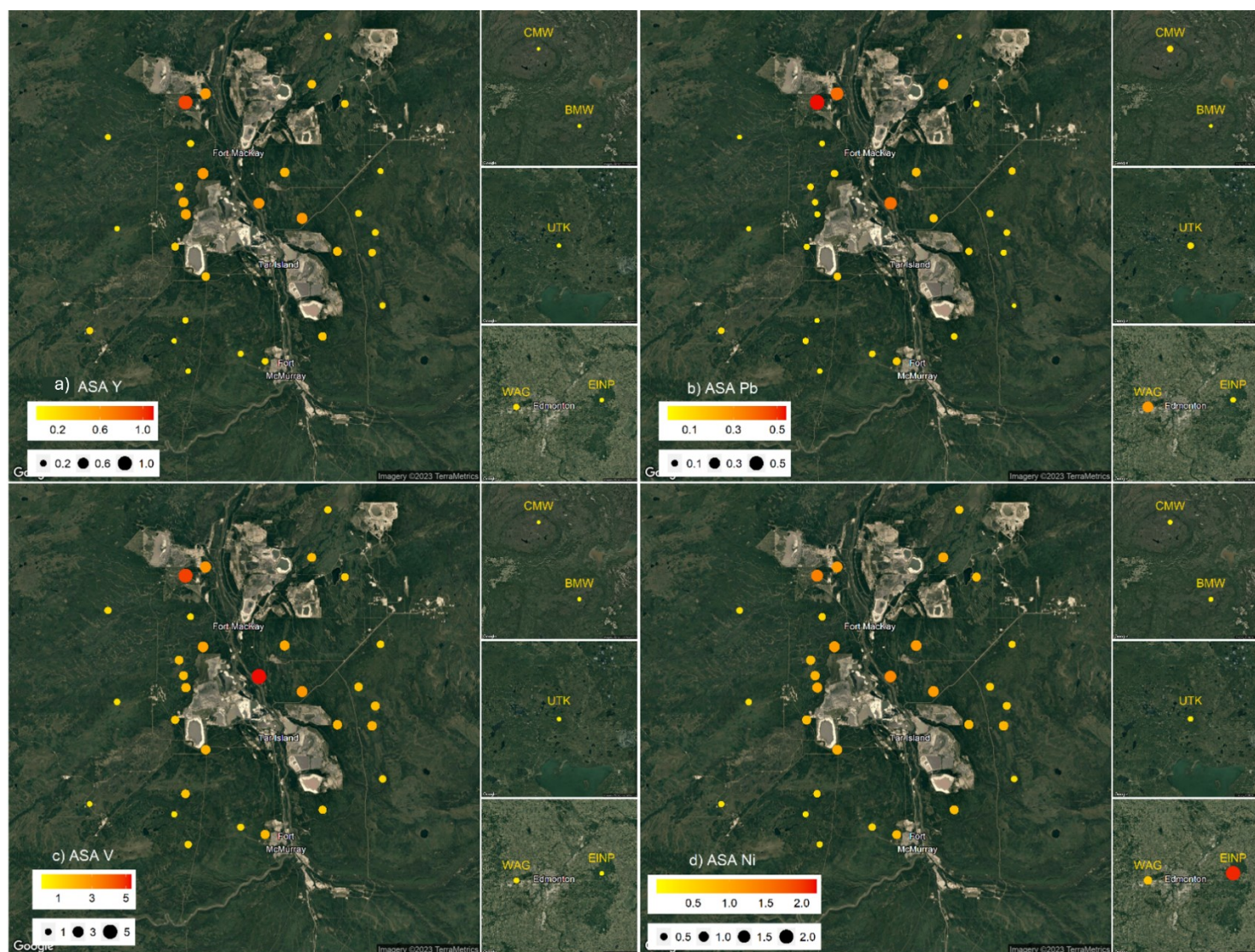




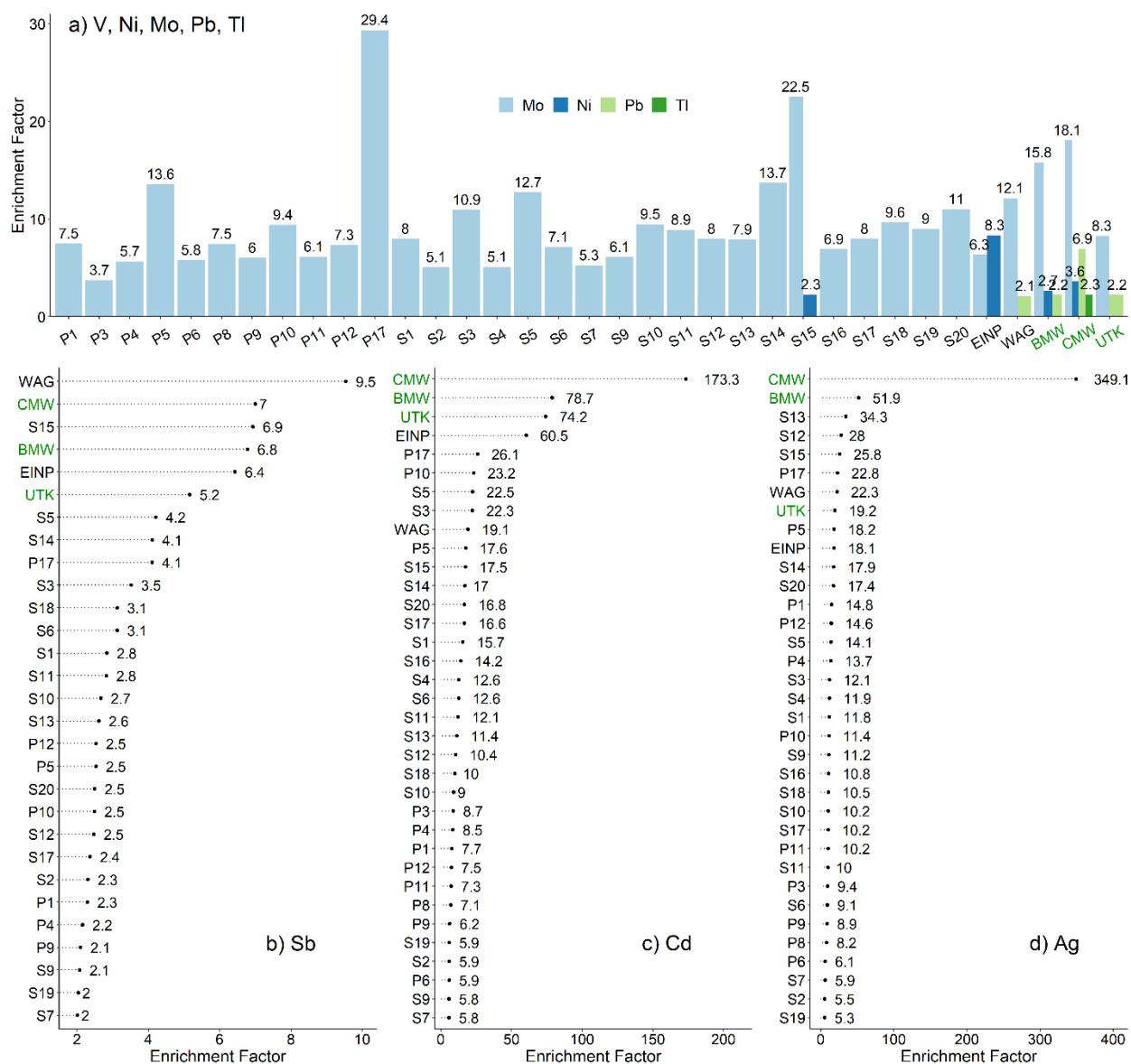
**Fig. S20.** Temporal trends of **total** concentration ( $\mu\text{g kg}^{-1}$  dry moss) of selected elements in *Sphagnum* moss collected in fall 2015 (sites EINP and WAG were excluded), 2019, and 2020. Boxplots show the range, 25<sup>th</sup> percentile, median, and 75th percentile of each element.



**Fig. S21.** Temporal trends of trace element concentration ( $\mu\text{g kg}^{-1}$  dry moss) in the Acid Soluble Ash (ASA) fraction of *Sphagnum* moss collected in fall 2015 (sites EINP and WAG were excluded), 2019, and 2020. Boxplots show the range, 25<sup>th</sup> percentile, median, and 75<sup>th</sup> percentile of each element.



**Fig. S22.** Spatial distribution of acid soluble a) **Y**, b) **Pb**, c) **V**, and d) **Ni** concentrations (mg kg<sup>-1</sup> dry moss) in *Sphagnum* mosses collected in fall 2015. ASA = Acid Soluble Ash, CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.



**Fig. S23.** Enrichment factors using Y as the reference element with values greater than 2 of a) V, Ni, Mo, Pb, Tl, b) Sb, c) Ag, and d) Cd of *Sphagnum* mosses collected in fall 2015. Enrichment factors of V were all below 2 and are not shown in the plot. CMW = Caribou Mountains Wildland, BMW = Birch Mountains Wildland, UTK = Utikuma, WAG = Wagner Natural Area, EINP = Elk Island National Park.



## References

- (1) Rudnick, R. L.; Gao, S. Composition of the continental crust. In *Treatise on Geochemistry*, 2<sup>nd</sup> ed.; Holland, H., Turekian, K., Eds.; Elsevier, 2014; pp 1–51. DOI: 10.1016/B978-0-08-095975-7.00301-6.
- (2) R Core Team. *R: A language and environment for statistical computing*; R Foundation for Statistical Computing, 2021. <https://www.r-project.org/>.
- (3) Carslaw, D. C.; Ropkins, K. openair - An R package for air quality data analysis. *Environ. Model. Softw.* **2012**, 27–28 (0), 52–61. DOI: 10.1016/j.envsoft.2011.09.008.
- (4) Mullan-Boudreau, G.; Belland, R.; Devito, K.; Noernberg, T.; Pelletier, R.; Shotyk, W. *Sphagnum* moss as an indicator of contemporary rates of atmospheric dust deposition in the Athabasca Bituminous Sands Region. *Environ. Sci. Technol.* **2017**, 51 (13), 7422–7431. DOI: 10.1021/acs.est.6b06195.