

Potential source areas for atmospheric lead reaching Ny-Ålesund from 2010 to 2018

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Supplementary material

The entire data analysis and figures presented in sections 3.1–3.4 are available as R script both on Zenodo at <https://doi.org/10.5281/zenodo.4484122> and on GitHub at https://github.com/andreabz/psa_pb_ny-alesund.

Table S1. Pb concentration (pg/m³), enrichment factors (EFs) and Pb isotope ratio values for PM₁₀ samples collected at Ny-Ålesund from 2010 to 2018. The entire dataset is available in electronic format at <https://doi.org/10.5281/zenodo.4484136>.

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 01-10 | 2010-03-16 | 1062.68 | 122.1 | 370 | 2.106 | 0.006 | 0.862 | 0.002 |
| 02-10 | 2010-03-20 | 1582.75 | 8.9 | 63 | 2.108 | 0.008 | 0.869 | 0.003 |
| 03-10 | 2010-03-23 | 1004.71 | 441.9 | 255 | 2.102 | 0.005 | 0.864 | 0.002 |
| 04-10 | 2010-03-27 | 1016.6 | 173.8 | 82 | 2.106 | 0.005 | 0.865 | 0.002 |
| 05-10 | 2010-03-31 | 826.14 | 1428.1 | 183 | 2.097 | 0.006 | 0.863 | 0.002 |
| 06-10 | 2010-04-08 | 1073.51 | 150.9 | 53 | 2.104 | 0.005 | 0.865 | 0.002 |
| 07-10 | 2010-04-12 | 1053.19 | 59.4 | 19 | 2.103 | 0.005 | 0.862 | 0.001 |
| 08-10 | 2010-04-16 | 739.76 | 71.8 | 21 | 2.105 | 0.004 | 0.863 | 0.001 |
| 09-10 | 2010-04-22 | 348.84 | 44.9 | 32 | 2.087 | 0.008 | 0.863 | 0.003 |
| 10-10 | 2010-04-24 | 1042.69 | 38.3 | 60 | 2.100 | 0.008 | 0.864 | 0.004 |
| 11-10 | 2010-04-28 | 1052.68 | 68.2 | 34 | 2.101 | 0.006 | 0.864 | 0.002 |
| 12-10 | 2010-05-02 | 2081.14 | 100.9 | 31 | 2.101 | 0.004 | 0.863 | 0.002 |
| 13-10 | 2010-05-10 | 2178.91 | 42.4 | 38 | 2.105 | 0.006 | 0.863 | 0.001 |
| 14-10 | 2010-05-18 | 1097.84 | 98.8 | 41 | 2.110 | 0.004 | 0.863 | 0.002 |
| 17-10 | 2010-06-17 | 1148.89 | <4.5 | - | - | - | - | - |
| 18-10 | 2010-06-22 | 1047.04 | <4.5 | - | - | - | - | - |
| 19-10 | 2010-06-26 | 1031.11 | 17.3 | 7 | 2.086 | 0.007 | 0.860 | 0.005 |
| 20-10 | 2010-06-30 | 1002.89 | 84.3 | 9 | 2.095 | 0.006 | 0.853 | 0.003 |
| 21-10 | 2010-07-04 | 1302.86 | 13.5 | 2 | 2.078 | 0.009 | 0.851 | 0.004 |
| 22-10 | 2010-07-09 | 1329.44 | 6.1 | 11 | 2.084 | 0.012 | 0.859 | 0.003 |
| 23-10 | 2010-07-14 | 1267.92 | 10.7 | 2 | 2.061 | 0.008 | 0.845 | 0.003 |
| 24-10 | 2010-07-19 | 1316.31 | 52.8 | 174 | 2.092 | 0.007 | 0.853 | 0.002 |
| 25-10 | 2010-07-24 | 1319.02 | 17.6 | 24 | 2.076 | 0.004 | 0.856 | 0.001 |
| 26-10 | 2010-07-29 | 1041.54 | <4.5 | - | - | - | - | - |
| 27-10 | 2010-08-02 | 445.29 | 31.6 | 17 | 2.078 | 0.012 | 0.852 | 0.004 |
| 29-10 | 2010-08-08 | 1348.79 | 5.3 | 4 | 2.085 | 0.005 | 0.859 | 0.003 |
| 30-10 | 2010-08-14 | 1323.73 | 17.3 | 26 | 2.092 | 0.005 | 0.860 | 0.003 |
| 31-10 | 2010-08-19 | 425.35 | 62.5 | 27 | 2.057 | 0.008 | 0.842 | 0.002 |
| 32-10 | 2010-08-24 | 928.35 | 14 | 13 | 2.079 | 0.007 | 0.857 | 0.004 |
| 33-10 | 2010-08-28 | 1061.56 | <4.5 | - | - | - | - | - |
| 34-10 | 2010-09-01 | 1009.39 | 6.6 | - | - | - | - | - |
| 35-10 | 2010-09-05 | 960 | <4.5 | - | - | - | - | - |
| 36-10 | 2010-09-13 | 445.03 | 43.5 | 23 | 2.101 | 0.006 | 0.859 | 0.004 |
| 01-11 | 2011-03-29 | 1148.88 | 555.1 | 183 | 2.109 | 0.003 | 0.865 | 0.001 |
| 02-11 | 2011-04-03 | 956.87 | 163.2 | 60 | 2.103 | 0.008 | 0.864 | 0.003 |
| 03-11 | 2011-04-07 | 1008.58 | 126.2 | 63 | 2.116 | 0.005 | 0.870 | 0.002 |
| 04-11 | 2011-04-11 | 1294.32 | <4.5 | - | 2.098 | 0.005 | 0.860 | 0.002 |

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 09-11 | 2011-05-01 | 787.44 | 88.1 | 81 | 2.099 | 0.004 | 0.862 | 0.002 |
| 10-11 | 2011-05-05 | 786.05 | 113.8 | 54 | 2.102 | 0.004 | 0.865 | 0.001 |
| 11-11 | 2011-05-09 | 805.96 | 44.1 | 39 | 2.097 | 0.005 | 0.863 | 0.003 |
| 12-11 | 2011-05-13 | 790.35 | 95.4 | 46 | 2.107 | 0.007 | 0.864 | 0.003 |
| 13-11 | 2011-05-17 | 761.33 | 74.6 | 16 | 2.109 | 0.008 | 0.863 | 0.003 |
| 14-11 | 2011-05-21 | 791.53 | 60.4 | 24 | 2.105 | 0.007 | 0.861 | 0.003 |
| 15-11 | 2011-05-25 | 756.53 | 42.3 | 27 | 2.094 | 0.006 | 0.865 | 0.003 |
| 16-11 | 2011-05-29 | 861.66 | 27.3 | 16 | 2.097 | 0.006 | 0.870 | 0.002 |
| 17-11 | 2011-06-05 | 803.32 | 21.4 | 43 | 2.088 | 0.011 | 0.866 | 0.003 |
| 18-11 | 2011-06-09 | 776.37 | 18.8 | 13 | 2.099 | 0.008 | 0.862 | 0.005 |
| 19-11 | 2011-06-13 | 776.02 | 91.9 | 20 | 2.102 | 0.006 | 0.864 | 0.003 |
| 20-11 | 2011-06-17 | 768.09 | 39.6 | - | - | - | - | - |
| 21-11 | 2011-06-21 | 772.73 | <4.5 | - | - | - | - | - |
| 22-11 | 2011-06-25 | 591.2 | 11.2 | - | - | - | - | - |
| 23-11 | 2011-06-29 | 764.76 | 9.9 | - | - | - | - | - |
| 24-11 | 2011-07-03 | 771.7 | 11.1 | 18 | 2.098 | 0.007 | 0.86 | 0.003 |
| 25-11 | 2011-07-07 | 775.62 | 22.8 | 22 | 2.093 | 0.008 | 0.86 | 0.003 |
| 26-11 | 2011-07-10 | NA | <4.5 | - | - | - | - | - |
| 27-11 | 2011-07-15 | 763.59 | <4.5 | - | - | - | - | - |
| 28-11 | 2011-07-19 | 766.87 | 19 | - | - | - | - | - |
| 29-11 | 2011-07-23 | 760.57 | 26.8 | 43 | 2.098 | 0.008 | 0.862 | 0.003 |
| 08-12 | 2012-04-21 | 1090.68 | 52 | 80 | 2.108 | 0.005 | 0.864 | 0.003 |
| 09-12 | 2012-04-25 | 1132.36 | 65.8 | 95 | 2.106 | 0.006 | 0.863 | 0.004 |
| 10-12 | 2012-04-29 | 1098.59 | 61 | 73 | 2.103 | 0.005 | 0.864 | 0.003 |
| 11-12 | 2012-05-03 | 1083.1 | 50.6 | 82 | 2.113 | 0.007 | 0.863 | 0.005 |
| 12-12 | 2012-05-07 | 1059.76 | 51.3 | 47 | 2.113 | 0.005 | 0.864 | 0.005 |
| 14-12 | 2012-05-15 | 1087.72 | 121 | 36 | 2.102 | 0.006 | 0.859 | 0.003 |
| 15-12 | 2012-05-19 | 1051.59 | 71.4 | 33 | 2.107 | 0.004 | 0.861 | 0.004 |
| 16-12 | 2012-05-23 | 997.82 | 72.3 | 52 | 2.099 | 0.006 | 0.860 | 0.004 |
| 17-12 | 2012-05-27 | 1091.42 | 29.3 | 27 | 2.107 | 0.008 | 0.860 | 0.005 |
| 18-12 | 2012-05-31 | 1040.93 | 39.9 | 23 | 2.109 | 0.009 | 0.864 | 0.005 |
| 19-12 | 2012-06-04 | 1054.97 | 45.4 | 39 | 2.106 | 0.007 | 0.861 | 0.003 |
| 20-12 | 2012-06-08 | 1092.18 | 22.8 | 41 | 2.107 | 0.009 | 0.863 | 0.005 |
| 21-12 | 2012-06-12 | 998.35 | 26.1 | 32 | 2.108 | 0.008 | 0.863 | 0.005 |
| 22-12 | 2012-06-16 | 1016.03 | 30.8 | 124 | 2.110 | 0.008 | 0.865 | 0.006 |
| 23-12 | 2012-06-20 | 1013.04 | 7 | 33 | 2.107 | 0.012 | 0.865 | 0.005 |
| 24-12 | 2012-06-24 | 1011.73 | <4.5 | - | - | - | - | - |
| 25-12 | 2012-06-28 | 1026.34 | 14.9 | 130 | 2.094 | 0.013 | 0.859 | 0.008 |
| 26-12 | 2012-07-02 | 1003.29 | 29.9 | 57 | 2.092 | 0.007 | 0.856 | 0.005 |
| 27-12 | 2012-07-06 | 1037.11 | 5.3 | 5 | 2.091 | 0.027 | 0.852 | 0.01 |
| 28-12 | 2012-07-10 | 1026.3 | 118.2 | 73 | 2.078 | 0.005 | 0.851 | 0.003 |
| 29-12 | 2012-07-14 | 1029.54 | 28.8 | 49 | 2.092 | 0.005 | 0.857 | 0.004 |
| 30-12 | 2012-07-18 | 1004.2 | 15.4 | 11 | 2.099 | 0.011 | 0.860 | 0.005 |
| 31-12 | 2012-07-22 | 1039.81 | 35.5 | 71 | 2.077 | 0.005 | 0.851 | 0.005 |
| 32-12 | 2012-07-26 | 1045.77 | 24 | 15 | 2.099 | 0.008 | 0.860 | 0.007 |
| 33-12 | 2012-07-30 | 1098.77 | 44.2 | 67 | 2.098 | 0.004 | 0.860 | 0.003 |
| 34-12 | 2012-08-03 | 1075.06 | 32.7 | - | 2.102 | 0.007 | 0.865 | 0.002 |
| 35-12 | 2012-08-07 | 1074.2 | 45.8 | 115 | 2.090 | 0.007 | 0.857 | 0.003 |
| 36-12 | 2012-08-11 | 919.08 | <4.5 | - | - | - | - | - |
| 37-12 | 2012-08-15 | 1088.64 | 24.5 | 54 | 2.071 | 0.01 | 0.852 | 0.005 |
| 39-12 | 2012-08-23 | 1074.06 | <4.5 | - | - | - | - | - |
| 40-12 | 2012-08-27 | 1073.32 | 41.4 | 141 | 2.090 | 0.011 | 0.858 | 0.005 |
| 41-12 | 2012-08-31 | 1082.66 | 11.1 | 37 | 2.082 | 0.022 | 0.857 | 0.01 |
| 42-12 | 2012-09-04 | 1062.7 | <4.5 | - | - | - | - | - |
| 01-13 | 2013-05-01 | 803.19 | 42.6 | 51 | 2.105 | 0.007 | 0.863 | 0.002 |
| 02-13 | 2013-05-05 | 1083.26 | 82.8 | 63 | 2.107 | 0.003 | 0.863 | 0.002 |
| 03-13 | 2013-05-09 | 1071.76 | 6.7 | - | 2.084 | 0.011 | 0.863 | 0.009 |

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 04-13 | 2013-05-13 | 1053.42 | 19.8 | 31 | 2.099 | 0.010 | 0.860 | 0.003 |
| 05-13 | 2013-05-17 | 1071.53 | 24.3 | 23 | 2.095 | 0.009 | 0.862 | 0.003 |
| 06-13 | 2013-05-21 | 1052.71 | 21.8 | 42 | 2.106 | 0.004 | 0.866 | 0.002 |
| 07-13 | 2013-05-25 | 1059.05 | 26 | 17 | 2.106 | 0.006 | 0.862 | 0.003 |
| 08-13 | 2013-05-29 | 1091.41 | 20.1 | 55 | 2.099 | 0.006 | 0.866 | 0.002 |
| 10-13 | 2013-06-06 | 1083.81 | 26.3 | 10 | 2.092 | 0.008 | 0.857 | 0.001 |
| 11-13 | 2013-06-10 | 1080.93 | 15.6 | 26 | 2.104 | 0.009 | 0.865 | 0.004 |
| 12-13 | 2013-06-14 | 1082.13 | <4.5 | - | - | - | - | - |
| 13-13 | 2013-06-18 | 1098.8 | 32.1 | 200 | 2.072 | 0.003 | 0.845 | 0.002 |
| 14-13 | 2013-06-22 | 1073.29 | 6.2 | - | 2.101 | 0.004 | 0.873 | 0.008 |
| 15-13 | 2013-06-26 | 1081.78 | 16.8 | 40 | 2.107 | 0.005 | 0.865 | 0.004 |
| 16-13 | 2013-06-30 | 1092.9 | 30.7 | 145 | 2.101 | 0.006 | 0.866 | 0.004 |
| 17-13 | 2013-07-04 | 1081.03 | 32.4 | 91 | 2.090 | 0.007 | 0.856 | 0.003 |
| 18-13 | 2013-07-08 | 1081.96 | <4.5 | - | - | - | - | - |
| 19-13 | 2013-07-12 | 1078.63 | 25.2 | 8 | 2.094 | 0.006 | 0.860 | 0.003 |
| 20-13 | 2013-07-16 | 1078.31 | 14.5 | 3 | 2.098 | 0.007 | 0.864 | 0.005 |
| 21-13 | 2013-07-20 | 1081.95 | 6.7 | 27 | 2.095 | 0.011 | 0.864 | 0.006 |
| 22-13 | 2013-07-24 | 1090.07 | <4.5 | - | - | - | - | - |
| 25-13 | 2013-08-05 | 1080.18 | 7 | 28 | 2.062 | 0.009 | 0.847 | 0.003 |
| 27-13 | 2013-08-13 | 1080.43 | <4.5 | - | - | - | - | - |
| 28-13 | 2013-08-17 | 1089.15 | 87 | 443 | 2.107 | 0.004 | 0.873 | 0.002 |
| 29-13 | 2013-08-21 | 1099.04 | <4.5 | - | - | - | - | - |
| 31-13 | 2013-08-29 | 1083.41 | <4.5 | - | - | - | - | - |
| 32-13 | 2013-09-02 | 1082.28 | 14.9 | 26 | 2.103 | 0.009 | 0.869 | 0.004 |
| 33-13 | 2013-09-06 | 1082.42 | 6.2 | 4 | 2.101 | 0.012 | 0.866 | 0.004 |
| 34-13 | 2013-09-10 | 784.58 | 24 | - | 2.057 | 0.007 | 0.839 | 0.002 |
| 01-14 | 2014-03-31 | 796.76 | 88.5 | 68 | 2.105 | 0.005 | 0.864 | 0.002 |
| 02-14 | 2014-04-04 | 817.68 | 72.1 | 111 | 2.105 | 0.002 | 0.863 | 0.001 |
| 03-14 | 2014-04-08 | 799.46 | <4.5 | - | 2.103 | 0.004 | 0.863 | 0.002 |
| 04-14 | 2014-04-12 | 799.12 | 85.8 | 97 | 2.109 | 0.003 | 0.863 | 0.002 |
| 05-14 | 2014-04-16 | 806.6 | 188.1 | - | 2.113 | 0.004 | 0.866 | 0.002 |
| 06-14 | 2014-04-20 | 772.74 | 307.3 | 135 | 2.107 | 0.003 | 0.865 | 0.002 |
| 07-14 | 2014-04-24 | 783.87 | 109.8 | 135 | 2.102 | 0.007 | 0.863 | 0.002 |
| 08-14 | 2014-04-28 | 799.59 | 81.3 | 53 | 2.102 | 0.005 | 0.861 | 0.002 |
| 09-14 | 2014-05-02 | 861.65 | 62.9 | 195 | 2.104 | 0.004 | 0.864 | 0.003 |
| 10-14 | 2014-05-07 | 821.06 | 51.6 | 29 | 2.100 | 0.006 | 0.861 | 0.003 |
| 11-14 | 2014-05-11 | 788.8 | 41 | 56 | 2.115 | 0.005 | 0.870 | 0.002 |
| 12-14 | 2014-05-15 | 838.49 | 68.7 | 44 | 2.106 | 0.004 | 0.863 | 0.001 |
| 13-14 | 2014-05-19 | 801.65 | 36.6 | 7 | 2.103 | 0.008 | 0.868 | 0.003 |
| 14-14 | 2014-05-23 | 801.73 | 65.3 | 7 | 2.100 | 0.005 | 0.854 | 0.003 |
| 15-14 | 2014-05-27 | 802.98 | 30.3 | 35 | 2.098 | 0.012 | 0.857 | 0.006 |
| 16-14 | 2014-05-31 | 792.31 | 12.6 | 62 | 2.110 | 0.009 | 0.864 | 0.003 |
| 17-14 | 2014-06-04 | 814.58 | 75.1 | 156 | 2.107 | 0.003 | 0.867 | 0.002 |
| 18-14 | 2014-06-08 | 799.12 | 17 | 47 | 2.091 | 0.009 | 0.864 | 0.004 |
| 19-14 | 2014-06-12 | 810.67 | 15.3 | - | - | - | - | - |
| 20-14 | 2014-06-16 | 799.85 | 11.1 | - | 2.114 | 0.006 | 0.867 | 0.002 |
| 21-14 | 2014-06-20 | 805.91 | 34.9 | 10 | 2.101 | 0.009 | 0.871 | 0.004 |
| 22-14 | 2014-06-24 | 804.64 | <4.5 | - | - | - | - | - |
| 23-14 | 2014-06-28 | 861.69 | 26.6 | 3 | 2.049 | 0.008 | 0.837 | 0.003 |
| 24-14 | 2014-07-03 | 814.61 | 11 | 14 | 2.092 | 0.010 | 0.855 | 0.005 |
| 25-14 | 2014-07-07 | 792.08 | 24.5 | 16 | 2.072 | 0.010 | 0.852 | 0.006 |
| 26-14 | 2014-07-12 | 454.1 | 66.9 | 476 | 2.072 | 0.008 | 0.850 | 0.004 |
| 27-14 | 2014-07-17 | 811.22 | 43.2 | 415 | 2.110 | 0.005 | 0.868 | 0.003 |
| 28-14 | 2014-07-21 | 818.24 | 18.3 | 18 | 2.099 | 0.007 | 0.863 | 0.004 |
| 29-14 | 2014-07-25 | 810.25 | <4.5 | - | - | - | - | - |
| 30-14 | 2014-07-29 | 813.75 | <4.5 | - | - | - | - | - |
| 31-14 | 2014-08-02 | 794.5 | 8.7 | 25 | 2.095 | 0.018 | 0.860 | 0.007 |

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 32-14 | 2014-08-06 | 816.95 | 14.1 | 19 | 2.075 | 0.014 | 0.848 | 0.005 |
| 33-14 | 2014-08-10 | 811.79 | 13.6 | 13 | 2.081 | 0.010 | 0.854 | 0.005 |
| 34-14 | 2014-08-18 | 801.84 | <4.5 | - | - | - | - | - |
| 35-14 | 2014-08-22 | 814.68 | 12.5 | 48 | 2.081 | 0.009 | 0.856 | 0.005 |
| 36-14 | 2014-08-26 | 707.32 | 23.5 | 24 | 2.097 | 0.010 | 0.860 | 0.005 |
| 37-14 | 2014-08-30 | 817.99 | 61.5 | 134 | 2.095 | 0.005 | 0.860 | 0.002 |
| 38-14 | 2014-09-03 | 830.1 | <4.5 | - | - | - | - | - |
| 39-14 | 2014-09-07 | 772.85 | <4.5 | - | - | - | - | - |
| 01-15 | 2015-02-28 | 790.33 | 82 | 59 | 2.100 | 0.006 | 0.863 | 0.004 |
| 02-15 | 2015-03-04 | 781.96 | 11.8 | 176 | 2.076 | 0.014 | 0.855 | 0.01 |
| 03-15 | 2015-03-08 | 774.24 | 48.9 | 349 | 2.100 | 0.007 | 0.867 | 0.003 |
| 04-15 | 2015-03-12 | 793.24 | 358.1 | 355 | 2.111 | 0.004 | 0.864 | 0.002 |
| 05-15 | 2015-03-16 | 798.81 | 185.3 | 233 | 2.107 | 0.004 | 0.862 | 0.002 |
| 06-15 | 2015-03-20 | 804.22 | 125 | 132 | 2.087 | 0.004 | 0.862 | 0.002 |
| 07-15 | 2015-03-24 | 799.4 | 121.4 | 138 | 2.087 | 0.003 | 0.861 | 0.002 |
| 08-15 | 2015-03-28 | 792.52 | 73.8 | 116 | 2.095 | 0.005 | 0.865 | 0.002 |
| 09-15 | 2015-04-01 | 813.64 | 141.7 | 52 | 2.10 | 0.004 | 0.857 | 0.002 |
| 10-15 | 2015-04-05 | 802.53 | 520.5 | 144 | 2.113 | 0.006 | 0.867 | 0.002 |
| 11-15 | 2015-04-09 | 809.16 | 514.8 | 512 | 2.101 | 0.005 | 0.865 | 0.003 |
| 12-15 | 2015-04-13 | 829.4 | 505.3 | 413 | 2.103 | 0.005 | 0.862 | 0.003 |
| 13-15 | 2015-04-17 | 818.13 | 20.1 | 82 | 2.101 | 0.011 | 0.861 | 0.005 |
| 14-15 | 2015-04-21 | 814.81 | 16.9 | 49 | 2.095 | 0.010 | 0.862 | 0.005 |
| 15-15 | 2015-04-25 | 820.35 | 20.9 | 45 | 2.093 | 0.009 | 0.862 | 0.006 |
| 16-15 | 2015-04-29 | 813.8 | 29.8 | 33 | 2.086 | 0.009 | 0.857 | 0.004 |
| 17-15 | 2015-05-03 | 823.41 | 17.5 | 38 | 2.087 | 0.015 | 0.857 | 0.006 |
| 18-15 | 2015-05-07 | 814.88 | 17.6 | 26 | 2.088 | 0.010 | 0.856 | 0.005 |
| 19-15 | 2015-05-11 | 809.78 | 13.5 | 51 | 2.086 | 0.010 | 0.858 | 0.006 |
| 20-15 | 2015-05-15 | 809.52 | 5.4 | 24 | 2.120 | 0.014 | 0.867 | 0.01 |
| 21-15 | 2015-05-19 | 807.75 | 26.1 | 42 | 2.085 | 0.006 | 0.855 | 0.004 |
| 22-15 | 2015-05-23 | 814.18 | 29.3 | 46 | 2.072 | 0.006 | 0.853 | 0.002 |
| 13-15 | 2015-05-27 | 816.16 | 29.8 | 37 | 2.091 | 0.007 | 0.858 | 0.004 |
| 24-15 | 2015-05-31 | 807.14 | 18.8 | 17 | 2.059 | 0.013 | 0.845 | 0.01 |
| 25-15 | 2015-06-04 | 808.27 | 4.5 | 11 | 2.088 | 0.011 | 0.854 | 0.006 |
| 26-15 | 2015-06-08 | 804.09 | 5.6 | 10 | 2.072 | 0.012 | 0.849 | 0.008 |
| 27-15 | 2015-06-12 | 809.83 | 13 | 4 | 2.069 | 0.009 | 0.849 | 0.005 |
| 28-15 | 2015-06-16 | 809.55 | 12.4 | 12 | 2.095 | 0.014 | 0.861 | 0.009 |
| 29-15 | 2015-06-20 | 805.57 | 32.3 | 72 | 2.065 | 0.009 | 0.842 | 0.005 |
| 30-15 | 2015-06-24 | 812.34 | 7.4 | 19 | 2.053 | 0.009 | 0.844 | 0.006 |
| 31-15 | 2015-06-28 | 809.95 | 6.1 | 21 | 2.073 | 0.017 | 0.855 | 0.007 |
| 32-15 | 2015-07-02 | 813.26 | 15.7 | 103 | 2.073 | 0.009 | 0.851 | 0.004 |
| 33-15 | 2015-07-06 | 807.97 | 6.2 | 9 | 2.062 | 0.008 | 0.841 | 0.005 |
| 34-15 | 2015-07-10 | 810.35 | 30.5 | 11 | 2.081 | 0.009 | 0.853 | 0.005 |
| 35-15 | 2015-07-14 | 811.86 | 6.2 | 10 | 2.079 | 0.010 | 0.860 | 0.006 |
| 36-15 | 2015-07-18 | 807.64 | 3.5 | 9 | 2.081 | 0.017 | 0.861 | 0.009 |
| 37-15 | 2015-07-22 | 812.89 | 3.5 | 14 | 2.065 | 0.015 | 0.848 | 0.010 |
| 38-15 | 2015-07-26 | 809.79 | 27.4 | 211 | 2.082 | 0.017 | 0.85 | 0.009 |
| 39-15 | 2015-07-30 | 811.55 | 12.7 | 20 | 2.095 | 0.013 | 0.856 | 0.005 |
| 40-15 | 2015-08-03 | 803 | 13.2 | 16 | 2.054 | 0.009 | 0.846 | 0.005 |
| 41-15 | 2015-08-07 | 810.61 | 5.1 | 22 | 2.064 | 0.016 | 0.85 | 0.008 |
| 42-15 | 2015-08-11 | 792.37 | 8.6 | 15 | 2.073 | 0.020 | 0.843 | 0.012 |
| 43-15 | 2015-08-15 | 809.02 | 8.5 | 31 | 2.113 | 0.018 | 0.866 | 0.009 |
| 44-15 | 2015-08-19 | 810.54 | 2.9 | 20 | 2.080 | 0.018 | 0.850 | 0.010 |
| 45-15 | 2015-08-23 | 818.81 | 4.3 | 38 | 2.109 | 0.026 | 0.864 | 0.017 |
| 46-15 | 2015-08-27 | 819.92 | 7.1 | 26 | 2.079 | 0.015 | 0.858 | 0.007 |
| 47-15 | 2015-08-31 | 809.93 | 2.8 | 9 | 2.073 | 0.021 | 0.844 | 0.012 |
| 48-15 | 2015-09-04 | 810.22 | 5.1 | 15 | 2.072 | 0.017 | 0.845 | 0.008 |
| 49-15 | 2015-09-08 | 604.14 | 10.8 | 8 | 2.072 | 0.014 | 0.844 | 0.009 |

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 50-15 | 2015-09-12 | 812.5 | 19.5 | 32 | 2.086 | 0.009 | 0.862 | 0.006 |
| 51-15 | 2015-09-16 | 809.78 | 128.3 | 36 | 2.089 | 0.005 | 0.848 | 0.004 |
| 52-15 | 2015-09-20 | 578.63 | 9.3 | 3 | 2.077 | 0.014 | 0.851 | 0.008 |
| 53-15 | 2015-09-24 | 807.74 | 91.6 | 883 | 2.099 | 0.008 | 0.864 | 0.004 |
| 54-15 | 2015-09-28 | 804.16 | 7.5 | 12 | 2.089 | 0.018 | 0.856 | 0.011 |
| 55-15 | 2015-10-02 | 809.98 | 9.2 | 8 | 2.094 | 0.010 | 0.852 | 0.004 |
| 56-15 | 2015-10-06 | 741.24 | 14.4 | 61 | 2.096 | 0.014 | 0.864 | 0.010 |
| 57-15 | 2015-10-10 | 782.29 | 27 | 21 | 2.082 | 0.008 | 0.848 | 0.004 |
| 58-15 | 2015-10-14 | 779.42 | 6.6 | 22 | 2.075 | 0.018 | 0.852 | 0.012 |
| 01-16 | 2016-03-04 | 774.77 | 37.5 | NA | 2.070 | 0.005 | 0.848 | 0.001 |
| 02-16 | 2016-03-08 | 770.62 | 15.7 | 135 | 2.103 | 0.005 | 0.865 | 0.003 |
| 03-16 | 2016-03-12 | 777.31 | 1.5 | 10 | 2.103 | 0.008 | 0.870 | 0.004 |
| 04-16 | 2016-03-16 | 784.08 | 35.3 | 45 | 2.081 | 0.002 | 0.855 | 0.001 |
| 05-16 | 2016-03-20 | 798.53 | 263.5 | 120 | 2.100 | 0.002 | 0.862 | 0.001 |
| 06-16 | 2016-03-24 | 782.44 | 510.1 | 282 | 2.092 | 0.002 | 0.856 | 0.001 |
| 07-16 | 2016-03-28 | 796.63 | 80.6 | 93 | 2.099 | 0.003 | 0.862 | 0.002 |
| 08-16 | 2016-04-03 | 861.65 | 57.7 | 26 | 2.094 | 0.003 | 0.859 | 0.001 |
| 09-16 | 2016-04-08 | 708.22 | 73.7 | 130 | 2.099 | 0.003 | 0.862 | 0.003 |
| 10-16 | 2016-04-12 | 709.85 | 97.6 | 46 | 2.099 | 0.004 | 0.861 | 0.002 |
| 11-16 | 2016-04-16 | 731.93 | 85.2 | 66 | 2.100 | 0.003 | 0.862 | 0.001 |
| 12-16 | 2016-04-20 | 747.67 | 52.2 | 78 | 2.095 | 0.001 | 0.858 | 0.001 |
| 13-16 | 2016-04-24 | 728.43 | 42.8 | 41 | 2.099 | 0.004 | 0.861 | 0.002 |
| 14-16 | 2016-04-28 | 820.83 | 49.3 | 50 | 2.100 | 0.006 | 0.862 | 0.001 |
| 15-16 | 2016-05-02 | 824.39 | 12.2 | 51 | 2.098 | 0.004 | 0.863 | 0.004 |
| 16-16 | 2016-05-06 | 806.5 | <1.0 | - | - | - | - | - |
| 17-16 | 2016-05-10 | 817.54 | 16.1 | 36 | 2.076 | 0.004 | 0.853 | 0.002 |
| 18-16 | 2016-05-14 | 757.41 | 7.8 | 68 | 2.109 | 0.004 | 0.870 | 0.002 |
| 19-16 | 2016-05-18 | 301.55 | 29.6 | 35 | 2.111 | 0.006 | 0.865 | 0.005 |
| 04-17 | 2017-03-01 | 773.89 | 135.9 | 304 | 2.101 | 0.002 | 0.871 | 0.001 |
| 05-17 | 2017-03-05 | 861.61 | 77.1 | 113 | 2.108 | 0.004 | 0.867 | 0.002 |
| 06-17 | 2017-03-10 | 775.83 | 170.5 | 120 | 2.100 | 0.002 | 0.865 | 0.001 |
| 07-17 | 2017-03-22 | 790.84 | 298.7 | 300 | 2.103 | 0.003 | 0.865 | 0.001 |
| 08-17 | 2017-03-26 | 762.34 | 386.8 | 160 | 2.105 | 0.003 | 0.865 | 0.002 |
| 09-17 | 2017-03-30 | 809.45 | 520.9 | 229 | 2.102 | 0.002 | 0.865 | 0.002 |
| 10-17 | 2017-04-03 | 802.64 | 241.1 | 178 | 2.106 | 0.001 | 0.868 | 0.001 |
| 11-17 | 2017-04-07 | 793.91 | 124.9 | 130 | 2.096 | 0.002 | 0.864 | 0.002 |
| 12-17 | 2017-04-11 | 813.72 | 577.3 | 455 | - | - | - | - |
| 13-17 | 2017-04-15 | 788.77 | 252.5 | 109 | 2.102 | 0.002 | 0.864 | 0.002 |
| 14-17 | 2017-04-19 | 776.94 | 158.7 | 69 | 2.085 | 0.005 | 0.859 | 0.003 |
| 15-17 | 2017-04-23 | 774.28 | 67.3 | 46 | 2.101 | 0.003 | 0.861 | 0.002 |
| 16-17 | 2017-04-27 | 785.51 | 39.5 | 58 | 2.099 | 0.004 | 0.856 | 0.001 |
| 17-17 | 2017-05-01 | 757.91 | 117.8 | 147 | 2.096 | 0.003 | 0.867 | 0.002 |
| 18-17 | 2017-05-05 | 782.42 | 72.5 | 50 | 2.100 | 0.002 | 0.861 | 0.003 |
| 19-17 | 2017-05-09 | 794.33 | 163.5 | 65 | 2.110 | 0.002 | 0.873 | 0.002 |
| 20-17 | 2017-05-13 | 794.05 | 80.3 | 48 | 2.103 | 0.003 | 0.862 | 0.004 |
| 21-17 | 2017-05-17 | 780.74 | 63.9 | 36 | 2.088 | 0.006 | 0.857 | 0.003 |
| 22-17 | 2017-05-21 | 791.01 | 18 | 27 | 2.101 | 0.004 | 0.863 | 0.004 |
| 23-17 | 2017-05-25 | 784.84 | 26.9 | 21 | 2.096 | 0.004 | 0.860 | 0.002 |
| 24-17 | 2017-05-29 | 796.33 | 46.1 | 13 | 2.097 | 0.003 | 0.854 | 0.002 |
| 25-17 | 2017-06-02 | 781.23 | 45.2 | 20 | 2.099 | 0.002 | 0.854 | 0.003 |
| 26-17 | 2017-06-06 | 796.67 | 153.3 | 73 | 2.108 | 0.005 | 0.874 | 0.002 |
| 27-17 | 2017-06-10 | 792.23 | 24 | 61 | - | - | - | - |
| 28-17 | 2017-06-14 | 778.48 | 9.4 | 33 | 2.099 | 0.004 | 0.861 | 0.003 |
| 29-17 | 2017-06-18 | 794.71 | 9.7 | 21 | 2.117 | 0.007 | 0.863 | 0.005 |
| 30-17 | 2017-06-22 | 782.42 | 11.2 | 22 | 2.107 | 0.006 | 0.859 | 0.002 |
| 31-17 | 2017-06-28 | 790.19 | 39.8 | 88 | 2.100 | 0.002 | 0.865 | 0.002 |
| 32-17 | 2017-06-30 | 796.33 | 2.4 | 5 | 2.104 | 0.012 | 0.859 | 0.004 |

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 33-17 | 2017-07-04 | 811.22 | <1.0 | - | - | - | - | - |
| 34-17 | 2017-07-08 | 796.61 | 1.1 | 10 | 2.107 | 0.007 | 0.859 | 0.011 |
| 35-17 | 2017-07-12 | 787.93 | 17.2 | 20 | - | - | - | - |
| 36-17 | 2017-07-16 | 801.73 | 11.8 | 13 | 2.079 | 0.006 | 0.841 | 0.003 |
| 37-17 | 2017-07-20 | 806.11 | 7.4 | 16 | - | - | - | - |
| 38-17 | 2017-07-24 | 796.41 | 3.9 | 5 | - | - | - | - |
| 39-17 | 2017-07-28 | 794.16 | 10.7 | 3 | - | - | - | - |
| 40-17 | 2017-08-01 | 792.14 | 5.8 | 4 | 2.092 | 0.004 | 0.86 | 0.004 |
| 41-17 | 2017-08-05 | 800.6 | 2.7 | 9 | 2.114 | 0.019 | 0.862 | 0.005 |
| 42-17 | 2017-08-09 | 815.77 | 5.7 | 6 | 2.090 | 0.003 | 0.859 | 0.002 |
| 43-17 | 2017-08-13 | 772.28 | 2.8 | 3 | - | - | - | - |
| 44-17 | 2017-08-17 | 789.26 | <1.0 | - | - | - | - | - |
| 45-17 | 2017-08-21 | 794.37 | 1.3 | 4 | 2.069 | 0.012 | 0.857 | 0.004 |
| 46-17 | 2017-08-25 | 801.04 | 4.7 | 5 | 2.071 | 0.006 | 0.854 | 0.004 |
| 47-17 | 2017-08-29 | 795.51 | 5.1 | 5 | - | - | - | - |
| 48-17 | 2017-09-02 | 795.15 | 18.8 | 32 | 2.099 | 0.006 | 0.864 | 0.004 |
| 49-17 | 2017-09-06 | 776.42 | 138.7 | 80 | - | - | - | - |
| 50-17 | 2017-09-10 | 791.37 | 8.2 | 6 | 2.103 | 0.014 | 0.853 | 0.006 |
| 51-17 | 2017-09-14 | 789.42 | 17.2 | 47 | 2.077 | 0.006 | 0.849 | 0.002 |
| 52-17 | 2017-09-18 | 803.62 | 7.9 | 6 | 2.061 | 0.006 | 0.849 | 0.005 |
| 53-17 | 2017-09-22 | 891.61 | 11.8 | 115 | - | - | - | - |
| 01-18 | 2018-02-23 | 817.62 | 8.6 | 30 | 2.113 | 0.008 | 0.865 | 0.004 |
| 02-18 | 2018-02-28 | 773.09 | 36.4 | 21 | 2.095 | 0.007 | 0.856 | 0.002 |
| 03-18 | 2018-03-04 | 774.37 | 30.8 | 23 | 2.104 | 0.005 | 0.858 | 0.001 |
| 04-18 | 2018-03-08 | 783.71 | 42.8 | 47 | 2.094 | 0.004 | 0.854 | 0.003 |
| 05-18 | 2018-03-12 | 775.04 | 41.7 | 24 | 2.097 | 0.005 | 0.863 | 0.001 |
| 06-18 | 2018-03-16 | 772.87 | 42.2 | 27 | 2.101 | 0.003 | 0.854 | 0.003 |
| 07-18 | 2018-03-20 | 772.25 | 166.6 | 41 | 2.098 | 0.003 | 0.861 | 0.001 |
| 08-18 | 2018-03-24 | 762.82 | 101 | 97 | 2.098 | 0.005 | 0.865 | 0.002 |
| 09-18 | 2018-03-28 | 762.68 | 55.7 | 76 | 2.107 | 0.003 | 0.862 | 0.003 |
| 10-18 | 2018-04-01 | 766.17 | 50.2 | 163 | 2.107 | 0.005 | 0.862 | 0.002 |
| 11-18 | 2018-04-05 | 784.6 | 193.7 | 75 | 2.104 | 0.003 | 0.862 | 0.001 |
| 12-18 | 2018-04-09 | 758.86 | 96.5 | 45 | 2.092 | 0.002 | 0.862 | 0.002 |
| 13-18 | 2018-04-13 | 760.21 | 107.8 | 28 | 2.103 | 0.003 | 0.860 | 0.001 |
| 14-18 | 2018-04-17 | 768.99 | 91.2 | 40 | 2.100 | 0.003 | 0.860 | 0.001 |
| 15-18 | 2018-04-21 | 764.53 | 179.2 | 29 | 2.106 | 0.003 | 0.859 | 0.001 |
| 16-18 | 2018-04-25 | 771.58 | 56 | 22 | 2.096 | 0.003 | 0.861 | 0.001 |
| 17-18 | 2018-04-29 | 737.42 | 65.3 | 29 | 2.101 | 0.005 | 0.856 | 0.002 |
| 18-18 | 2018-05-03 | 780.77 | 96.9 | 44 | 2.077 | 0.002 | 0.855 | 0.002 |
| 19-18 | 2018-05-07 | 777.38 | 20.7 | 12 | 2.089 | 0.006 | 0.865 | 0.002 |
| 20-18 | 2018-05-11 | 724.97 | 7.2 | 5 | 2.088 | 0.008 | 0.851 | 0.005 |
| 21-18 | 2018-05-15 | 719.74 | 13.1 | 9 | 2.095 | 0.012 | 0.853 | 0.003 |
| 22-18 | 2018-05-19 | 718.02 | <2.2 | - | - | - | - | - |
| 23-18 | 2018-05-23 | 752.66 | 24.9 | 11 | 2.086 | 0.006 | 0.856 | 0.001 |
| 24-18 | 2018-05-27 | 801.75 | 42.8 | 67 | 2.056 | 0.005 | 0.841 | 0.002 |
| 25-18 | 2018-05-31 | 810.45 | <2.2 | - | - | - | - | - |
| 26-18 | 2018-06-04 | 799.08 | 28.2 | 9 | 2.092 | 0.005 | 0.852 | 0.002 |
| 27-18 | 2018-06-08 | 802.48 | 13.9 | 13 | 2.103 | 0.003 | 0.855 | 0.003 |
| 28-18 | 2018-06-12 | 791.7 | 9.3 | 5 | 2.091 | 0.010 | 0.847 | 0.005 |
| 29-18 | 2018-06-16 | 808.02 | 9.5 | 7 | 2.087 | 0.010 | 0.852 | 0.004 |
| 30-18 | 2018-06-20 | 817 | 20.9 | 12 | 2.082 | 0.008 | 0.844 | 0.002 |
| 31-18 | 2018-06-24 | 767.13 | 6.8 | 8 | 2.082 | 0.007 | 0.855 | 0.003 |
| 32-18 | 2018-06-28 | 807.52 | 2.2 | 8 | 2.094 | 0.014 | 0.852 | 0.006 |
| 34-18 | 2018-07-06 | 799.77 | 14.3 | 11 | 2.087 | 0.006 | 0.854 | 0.002 |
| 35-18 | 2018-07-10 | 809.82 | 4.1 | 14 | 2.093 | 0.013 | 0.853 | 0.004 |
| 36-18 | 2018-07-14 | 799.07 | 11 | 15 | 2.091 | 0.009 | 0.857 | 0.002 |
| 37-18 | 2018-07-18 | 785.34 | 8.2 | 27 | 2.058 | 0.006 | 0.844 | 0.005 |

| ID | Sampling date | Volume (m ³) | Pb (pg/m ³) | EF(Pb/Al) _c | ²⁰⁸ Pb/ ²⁰⁶ Pb | U (95%-conf) | ²⁰⁷ Pb/ ²⁰⁶ Pb | U (95%-conf) |
|-------|---------------|--------------------------|-------------------------|------------------------|--------------------------------------|--------------|--------------------------------------|--------------|
| 38-18 | 2018-07-22 | 781.9 | 3.1 | 10 | 2.090 | 0.006 | 0.865 | 0.003 |
| 39-18 | 2018-07-26 | 781.16 | 2.1 | 3 | 2.082 | 0.017 | 0.854 | 0.003 |
| 40-18 | 2018-07-30 | 805.75 | 104.3 | 35 | 2.098 | 0.003 | 0.863 | 0.002 |
| 41-18 | 2018-08-03 | 766.89 | 5.8 | 5 | 2.084 | 0.016 | 0.846 | 0.006 |
| 42-18 | 2018-08-08 | 770.13 | 4.7 | 3 | 2.092 | 0.008 | 0.854 | 0.007 |
| 43-18 | 2018-08-12 | 806.24 | 4.6 | 4 | 2.093 | 0.006 | 0.858 | 0.006 |
| 44-18 | 2018-08-16 | 829.66 | 20.8 | 13 | 2.122 | 0.008 | 0.883 | 0.002 |
| 45-18 | 2018-08-20 | 804.1 | 11.3 | 18 | 2.099 | 0.010 | 0.865 | 0.004 |
| 46-18 | 2018-08-25 | 762.55 | 20.6 | 26 | 2.049 | 0.009 | 0.837 | 0.002 |

Table S2. Percentage of monthly BTs calculated from 2010 to 2018 associated to the different geographical macro-sector.

| Year | Month | Eurasia | North America | Arctic Ocean |
|-------------|--------------|----------------|----------------------|---------------------|
| 2010 | January | 26 | 20 | 54 |
| 2010 | February | 100 | 0 | 0 |
| 2010 | March | 40 | 0 | 60 |
| 2010 | April | 0 | 0 | 100 |
| 2010 | May | 42 | 0 | 58 |
| 2010 | June | 0 | 33 | 67 |
| 2010 | July | 0 | 64 | 36 |
| 2010 | August | 0 | 40 | 60 |
| 2010 | September | 36 | 25 | 39 |
| 2010 | October | 55 | 45 | 0 |
| 2010 | November | 0 | 34 | 64 |
| 2010 | December | 0 | 48 | 52 |
| 2011 | January | 52 | 48 | 0 |
| 2011 | February | 67 | 33 | 0 |
| 2011 | March | 0 | 39 | 61 |
| 2011 | April | 0 | 69 | 32 |
| 2011 | May | 0 | 44 | 56 |
| 2011 | June | 9 | 26 | 66 |
| 2011 | July | 0 | 50 | 50 |
| 2011 | August | 0 | 0 | 100 |
| 2011 | September | 0 | 38 | 63 |
| 2011 | October | 11 | 38 | 51 |
| 2011 | November | 0 | 46 | 54 |
| 2011 | December | 30 | 70 | 0 |
| 2012 | January | 24 | 76 | 0 |
| 2012 | February | 39 | 34 | 27 |
| 2012 | March | 59 | 41 | 0 |
| 2012 | April | 0 | 0 | 100 |
| 2012 | May | 0 | 0 | 100 |
| 2012 | June | 0 | 13 | 87 |
| 2012 | July | 0 | 0 | 100 |
| 2012 | August | 0 | 23 | 77 |
| 2012 | September | 0 | 56 | 44 |
| 2012 | October | 33 | 43 | 24 |
| 2012 | November | 32 | 25 | 43 |
| 2012 | December | 44 | 0 | 56 |
| 2013 | January | 30 | 17 | 52 |
| 2013 | February | 25 | 41 | 33 |
| 2013 | March | 34 | 45 | 21 |
| 2013 | April | 59 | 41 | 0 |
| 2013 | May | 0 | 79 | 21 |
| 2013 | June | 0 | 51 | 49 |
| 2013 | July | 0 | 56 | 44 |
| 2013 | August | 0 | 100 | 0 |
| 2013 | September | 60 | 40 | 0 |
| 2013 | October | 0 | 24 | 76 |

| Year | Month | Eurasia | North America | Arctic Ocean |
|-------------|--------------|----------------|----------------------|---------------------|
| 2013 | November | 38 | 29 | 33 |
| 2013 | December | 70 | 30 | 0 |
| 2014 | January | 0 | 0 | 100 |
| 2014 | February | 100 | 0 | 0 |
| 2014 | March | 0 | 27 | 73 |
| 2014 | April | 0 | 0 | 100 |
| 2014 | May | 0 | 19 | 81 |
| 2014 | June | 0 | 35 | 65 |
| 2014 | July | 0 | 71 | 29 |
| 2014 | August | 0 | 63 | 37 |
| 2014 | September | 44 | 56 | 0 |
| 2014 | October | 0 | 34 | 66 |
| 2014 | November | 40 | 60 | 0 |
| 2014 | December | 32 | 36 | 32 |
| 2015 | January | 30 | 21 | 38 |
| 2015 | February | 49 | 51 | 0 |
| 2015 | March | 35 | 32 | 32 |
| 2015 | April | 29 | 0 | 71 |
| 2015 | May | 0 | 58 | 42 |
| 2015 | June | 0 | 60 | 40 |
| 2015 | July | 4 | 32 | 64 |
| 2015 | August | 0 | 67 | 33 |
| 2015 | September | 46 | 15 | 39 |
| 2015 | October | 0 | 42 | 58 |
| 2015 | November | 34 | 66 | 0 |
| 2015 | December | 21 | 27 | 52 |
| 2016 | January | 39 | 61 | 0 |
| 2016 | February | 48 | 28 | 24 |
| 2016 | March | 47 | 53 | 0 |
| 2016 | April | 0 | 25 | 75 |
| 2016 | May | 0 | 75 | 25 |
| 2016 | June | 0 | 83 | 16 |
| 2016 | July | 39 | 18 | 43 |
| 2016 | August | 0 | 48 | 52 |
| 2016 | September | 25 | 36 | 39 |
| 2016 | October | 49 | 51 | 0 |
| 2016 | November | 70 | 30 | 0 |
| 2016 | December | 0 | 32 | 68 |
| 2017 | January | 28 | 18 | 54 |
| 2017 | February | 50 | 50 | 0 |
| 2017 | March | 0 | 38 | 62 |
| 2017 | April | 52 | 48 | 0 |
| 2017 | May | 0 | 72 | 28 |
| 2017 | June | 0 | 0 | 100 |
| 2017 | July | 0 | 70 | 30 |
| 2017 | August | 0 | 59 | 41 |
| 2017 | September | 13 | 42 | 46 |
| 2017 | October | 52 | 0 | 48 |

| Year | Month | Eurasia | North America | Arctic Ocean |
|-------------|--------------|----------------|----------------------|---------------------|
| 2017 | November | 22 | 34 | 44 |
| 2017 | December | 57 | 12 | 30 |
| 2018 | January | 54 | 18 | 27 |
| 2018 | February | 0 | 31 | 69 |
| 2018 | March | 0 | 36 | 64 |
| 2018 | April | 25 | 76 | 0 |
| 2018 | May | 0 | 51 | 49 |
| 2018 | June | 0 | 50 | 50 |
| 2018 | July | 0 | 100 | 0 |
| 2018 | August | 0 | 31 | 69 |
| 2018 | September | 0 | 37 | 63 |
| 2018 | October | 0 | 34 | 66 |
| 2018 | November | 0 | 33 | 67 |
| 2018 | December | 21 | 40 | 40 |

Table S3. Percentage of BTs, distance between the first and last endpoints and altitude of the last endpoint for the classes winter and summer from 2010 to 2018.

| Year | Class | %BT | | | Distance (km) | | | Altitude (m a.g.l.) | | |
|------|--------|---------|---------------|--------------|---------------|---------------|--------------|---------------------|---------------|--------------|
| | | Eurasia | North America | Arctic Ocean | Eurasia | North America | Arctic Ocean | Eurasia | North America | Arctic Ocean |
| 2010 | winter | 32 | 12 | 56 | 1230 | 2906 | 1997 | 2162 | 2018 | 1989 |
| 2010 | summer | 0 | 54 | 46 | - | 1124 | 2044 | - | 1973 | 1955 |
| 2011 | winter | 29 | 30 | 41 | 813 | 2686 | 1864 | 1974 | 2514 | 1893 |
| 2011 | summer | 0 | 44 | 56 | - | 1560 | 1838 | - | 2175 | 1952 |
| 2012 | winter | 39 | 27 | 34 | 1297 | 2804 | 2322 | 2149 | 2370 | 2101 |
| 2012 | summer | 0 | 41 | 59 | - | 1147 | 1791 | - | 1958 | 1727 |
| 2013 | winter | 19 | 33 | 48 | 1568 | 2007 | 2298 | 1804 | 2443 | 2137 |
| 2013 | summer | 0 | 51 | 49 | - | 1949 | 1259 | - | 1948 | 1988 |
| 2014 | winter | 40 | 21 | 39 | 1296 | 2787 | 2367 | 1942 | 2519 | 2220 |
| 2014 | summer | 0 | 51 | 49 | - | 1493 | 1842 | - | 1994 | 1843 |
| 2015 | winter | 54 | 18 | 28 | 2268 | 2994 | 1723 | 1359 | 2286 | 1427 |
| 2015 | summer | 0 | 38 | 62 | - | 1626 | 1646 | - | 1495 | 1176 |
| 2016 | winter | 30 | 24 | 46 | 1743 | 2619 | 1704 | 1387 | 1822 | 1114 |
| 2016 | summer | 39 | 27 | 34 | 982 | 2201 | 1657 | 1299 | 1548 | 1165 |
| 2017 | winter | 16 | 13 | 72 | 2444 | 3104 | 1473 | 1357 | 1611 | 1420 |
| 2017 | summer | 17 | 26 | 57 | 1396 | 2184 | 1483 | 2194 | 1749 | 1210 |
| 2018 | winter | 20 | 22 | 58 | 1746 | 2434 | 1646 | 2090 | 2266 | 1705 |
| 2018 | summer | 0 | 55 | 45 | - | 1951 | 1254 | - | 1664 | 1632 |

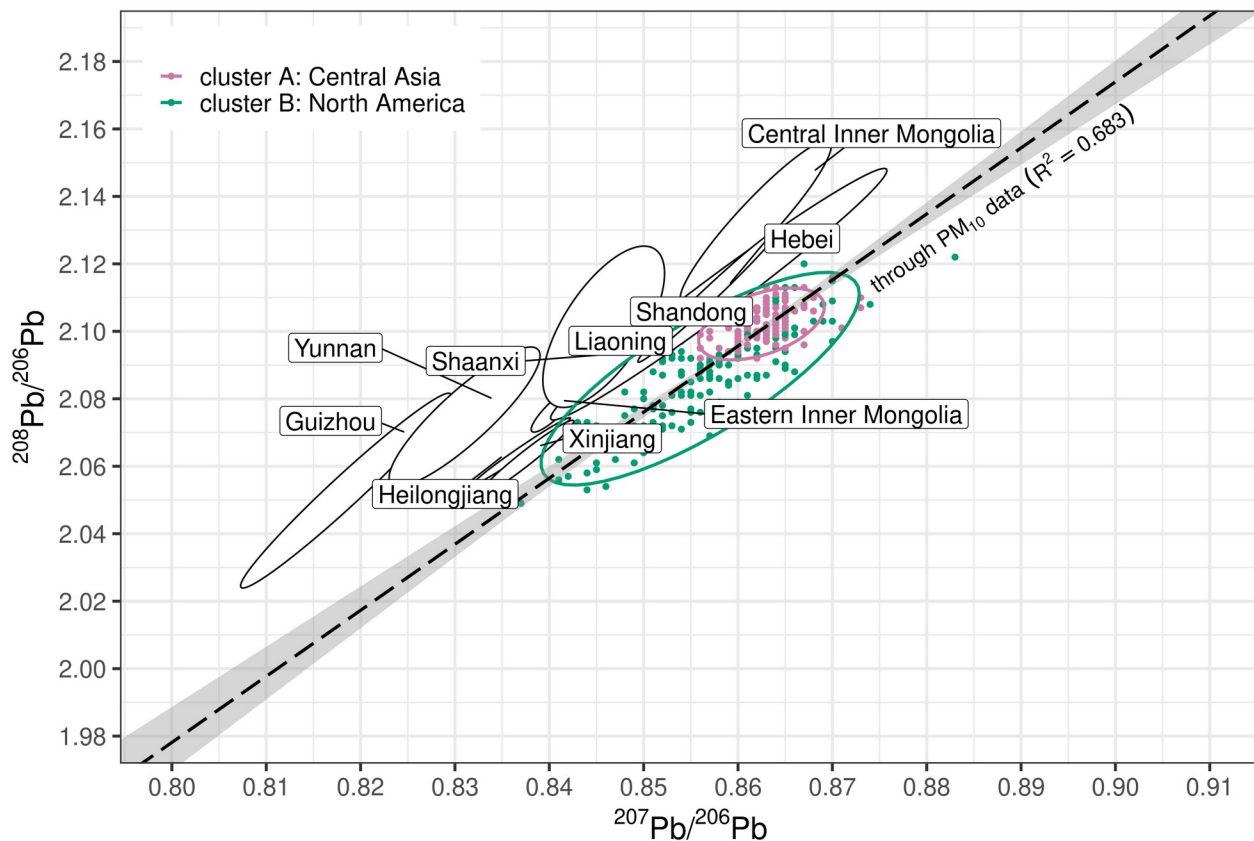


Figure S1: Three isotope plot for PM₁₀ samples collected at Ny-Ålesund from 2010 to 2018, compared with Chinese coals. PM₁₀ data are classified in two clusters according to the result of the Gaussian Mixture Modeling (GMM) for Pb isotope ratio values, enrichment factors (EFs) and Pb concentrations. The regression line for PM₁₀ data was obtained by Deming regression, considering a relative uncertainty on $^{208}\text{Pb}/^{206}\text{Pb}$ values as twice that for $^{207}\text{Pb}/^{206}\text{Pb}$ values. Data for Chinese coals: [46]. Black ellipses represent the 95% confidence interval of the mean for literature data.

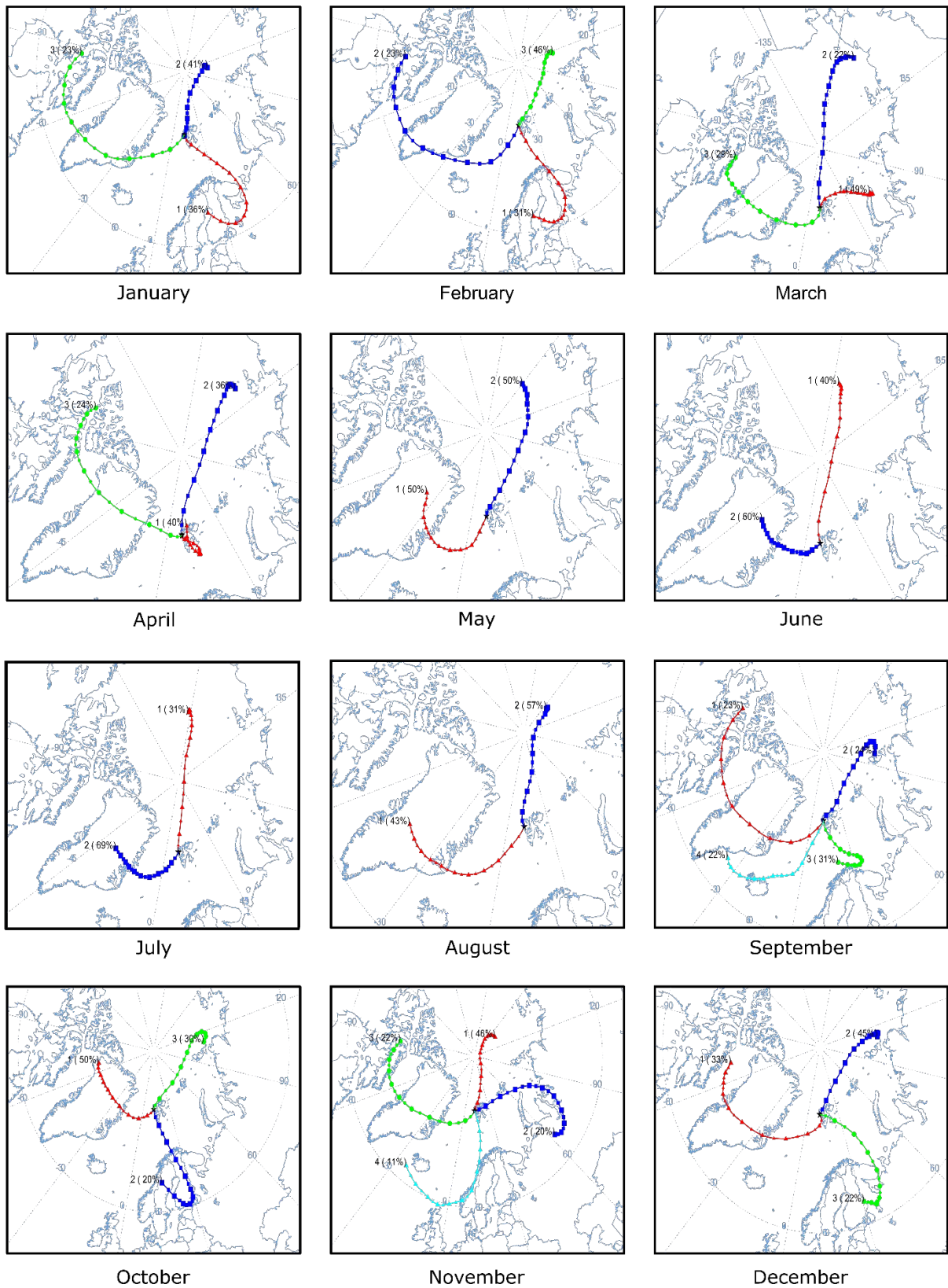


Figure S2: Geographic representation of the centroids obtained from the cluster analysis of the monthly BTs from 2010 to 2018, for 1000 m a.g.l as altitude of the endpoint at Ny-Alesund. For the other altitudes (500 and 1500 m a.g.l.) the results are similar. The percentages of the BTs associated to each cluster are also showed.