



## Supplement of

# In-depth characterization of submicron particulate matter inter-annual variations at a street canyon site in northern Europe

Luis M. F. Barreira et al.

Correspondence to: Luis M. F. Barreira (luis.barreira@fmi.fi) and Hilkka Timonen (hilkka.timonen@fmi.fi)

The copyright of individual parts of the supplement might differ from the article licence.

## 1 S1 Supplemental graphics and results



Figure S1: Measurement periods of instruments employed in this study.



Figure S2: Density (top plot) and collection efficiency (bottom plot) determined during this study. Median and average values were included in the graphic.



Figure S3: Correction applied to biomass burning (BB) percentage due to a change in the type of filter tape used during measurements. The red lines are the mean BB before correction (top plot) and during the entire period (bottom plot), while black line indicates the period after which the correction was applied.



13

Figure S4: Total ACSM concentrations plus eBC measured by MAAP vs. PM<sub>1</sub> measured by DMPS, coloured according
 to the sampling time, before (a) and after (b) the employed correction. Slope coefficients and corresponding standard
 deviations were included.



19Figure S5: Total ACSM concentrations plus eBC measured by MAAP vs. PM2.5 measured by TEOM, coloured20according to the sampling time. Slope coefficients and corresponding standard deviations were included.21

- $\frac{21}{22}$
- \_\_\_\_
- 23
- 24





Figure S6: PM<sub>1</sub> characterization determined by other studies performed over the globe in the last years (Green=organics; red=SO<sub>4</sub>; blue=NO<sub>3</sub>; yellow=NH<sub>4</sub>; black=eBC). The world map image was taken from Wikimedia commons (<u>https://commons.wikimedia.org/wiki/File:World map blank with blue sea.svg</u>).









34

Figure S8: Monthly variation of local eBC at the sampling site. Local contribution of eBC was calculated by subtracting eBC measured at an urban background site (Kallio, left) and at a regional background site (Luukki, right) (Luoma et al., 2020) from the eBC measured at the street canyon site, and the remaining eBC was divided by the street canyon eBC concentration. In each box, the mid-line shows the median value for each x-value component, whisker bottom and top correspond to the 10<sup>th</sup> and 90<sup>th</sup> percentiles, and box top and bottom correspond to 75<sup>th</sup> and 25<sup>th</sup> percentiles respectively.



Figure S9: Monthly variation of coating factor (a) and compensation parameter ( $K_6$ , (b)). In each box, the mid-line shows the median value for each x-value component, whisker bottom and top correspond to the 10<sup>th</sup> and 90<sup>th</sup> percentiles, and box top and bottom correspond to 75<sup>th</sup> and 25<sup>th</sup> percentiles respectively.





Figure S10: Week variability of eBC (measured by MAAP), BC<sub>FF</sub>, BB and BC<sub>WB</sub> (determined by Aethalometer). In
 each box, the mid-line shows the median value for each x-value component, whisker bottom and top correspond to the
 10<sup>th</sup> and 90<sup>th</sup> percentiles, and box top and bottom correspond to 75<sup>th</sup> and 25<sup>th</sup> percentiles respectively.







56

57 Figure S12: Diurnal and monthly variations of hourly median BC<sub>FF</sub>, NO<sub>x</sub>, BC<sub>WB</sub>, organics and VC during workdays 58 (left plots) and weekends (right plots).



Figure S13: Diurnal variation of NR-PM<sub>1</sub> constituents (Organics, NO<sub>3</sub>, NH<sub>4</sub> and SO<sub>4</sub>). In each box, the mid-line shows the median value for each x-value component, whisker bottom and top correspond to the 10<sup>th</sup> and 90<sup>th</sup> percentiles, and box top and bottom correspond to 75<sup>th</sup> and 25<sup>th</sup> percentiles respectively.



Figure S14: Back trajectories observed during the measurement period for the long-range transport event (a) and forthe local pollution event (b).





Figure S15: Criteria used for evaluating the most intense pollution events occurring in Helsinki during the measurement period.



Figure S16: Seasonal variation in atmospheric aerosol particle constituent's median concentration and their
 corresponding relative amounts during the pollution episodes. The monthly pollution frequency was also included
 (brown line); Mar–May in spring, Jun–Aug in summer, Sep–Nov in autumn, and Dec–Feb in winter.

#### 76 S2 Meteorology at the Supersite station during the measurement period in 2015–2019

The meteorological parameters measured during the period of 2015–2019 were evaluated to gain information on the influence of local meteorological conditions on observed aerosol particle concentrations. The minimum temperature was -24.5 °C (January 1<sup>st</sup>, 2016) and the maximum temperature was +30.8 °C (August 2<sup>nd</sup>, 2018).
The coldest month was January 2016 (-8.6 °C), which was exceptionally cold. On the other hand, the warmest month was July 2018 (21.5 °C) that broke the earlier record of high monthly average temperature in Helsinki.
Generally, the coldest month in Helsinki is February with average temperature of -7.0 °C (1981–2010, https://www.ilmatieteenlaitos.fi/helmikuu) and the warmest month is July with average temperature of 17.8 °C

84 (1981–2010, https://www.ilmatieteenlaitos.fi/heinakuu).

85 The relative humidity (RH) stayed mostly around 75-80 % during wintertime, while the driest periods 86 were observed in May with monthly average RH of 48–54 %. The values are close to what is commonly observed 87 in Helsinki. During the coldest seasons of the year, RH is usually about 90 %, while in spring and summer RH is 88 around 65 % (Pirinen et al., 2012). Wet deposition also influences significantly the measured atmospheric 89 concentrations by washing out the particles during precipitation events. During this study, the rainiest months 90 were between June to September, with maximum monthly precipitation sum observed in June 2016 (150,9 mm). 91 This result is in line with statistics from Helsinki between years 1981 and 2010, showing that the maximums of 92 monthly average rain accumulations are observed in August (80 mm) (https://www.ilmatieteenlaitos.fi/elokuu).

The maximum monthly average wind speeds were observed during winter with values between 5–5.5 m/s, while summertime average wind speeds were between 3.4–4.5 m s<sup>-1</sup>. Although the winds were stronger in wintertime, the surface layer of atmosphere is usually more stable in wintertime than in summertime and mixing in vertical direction is then more efficient during summer. The prevailing wind direction was south. The sampling site was thus severely affected by pollutants transported from other regions of Europe.

98 The planetary boundary layer height (PBLH) had a strong seasonal variation, as expected in northern latitudes 99 where seasonality is particularly evident. The mixing height was relatively constant during wintertime, with the 100 median value of about 400 m. This value is consistent with stagnant conditions during this time of the year, 101 favoring the accumulation of PM in a relatively shallow boundary layer. On the other hand, summertime mixing-102 heights had the lowest yearly values during night and early mornings (23h-10h) but increased rapidly up to above 103 2000 m during afternoon promoting dilution of atmospheric pollutants. The role of atmospheric dilution in 104 pollutant concentrations was evaluated by determining the ventilation coefficient (VC=wind speed x PBLH), that 105 has been often used in other studies as a parameter to characterize pollutants dilution (e.g. Gani et al., 2019; 106 Sujatha et al., 2016). The VC dilution rate was much lower during the coldest periods of the year (Fig. S7), 107 contributing to increased amounts of PM. The highest VC during spring and summer coincides with an increase 108 in temperature and solar radiation that enhances convection and PBL growth.

### 109 **References**

- 110 Aurela, M., Saarikoski, S., Niemi, J. V., Canonaco, F., Prevot, A. S. H., Frey, A., Carbone, S., Kousa, A. and
- 111 Hillamo, R.: Chemical and Source Characterization of Submicron Particles at Residential and Traffic Sites in the
- Helsinki Metropolitan Area, Finland, Aerosol Air Qual. Res., 15(4), 1213–1226, doi:10.4209/aaqr.2014.11.0279,
- 113 2015.
- Carbone, S., Saarikoski, S., Frey, A., Reyes, F., Reyes, P., Castillo, M., Gramsch, E., Oyola, P., Jayne, J.,
  Worsnop, D. R. and others: Chemical characterization of submicron aerosol particles in Santiago de Chile,
  Aerosol Air Qual. Res, 13(2), 462–473, doi:10.4209/aaqr.2012.10.0261, 2013.
- 117 Du, W., Sun, Y. L., Xu, Y. S., Jiang, Q., Wang, Q. Q., Yang, W., Wang, F., Bai, Z. P., Zhao, X. D. and Yang, Y.
- 118 C.: Chemical characterization of submicron aerosol and particle growth events at a national background site (3295
- 119 m a.s.l.) on the Tibetan Plateau, Atmos. Chem. Phys., 15(18), 10811–10824, doi:10.5194/acp-15-10811-2015,
- 120 2015.
- 121 Gani, S., Bhandari, S., Seraj, S., Wang, D. S., Patel, K., Soni, P., Arub, Z., Habib, G., Hildebrandt Ruiz, L. and
- 122 Apte, J. S.: Submicron aerosol composition in the world's most polluted megacity: the Delhi Aerosol Supersite
- 123 study, Atmos. Chem. Phys., 19(10), 6843–6859, doi:10.5194/acp-19-6843-2019, 2019.
- Gramsch, E., Reyes, F., Vásquez, Y., Oyola, P. and Rubio, M. A.: Prevalence of Freshly Generated Particles
  during Pollution Episodes in Santiago de Chile, Aerosol Air Qual. Res., 16(9), 2172–2185,
  doi:10.4209/aaqr.2015.12.0691, 2016.
- Hu, W., Hu, M., Hu, W.-W., Zheng, J., Chen, C., Wu, Y. and Guo, S.: Seasonal variations in high time-resolved
  chemical compositions, sources, and evolution of atmospheric submicron aerosols in the megacity Beijing, Atmos.
  Chem. Phys., 17(16), 9979–10000, doi:10.5194/acp-17-9979-2017, 2017.
- Jiang, Q., Wang, F. and Sun, Y.: Analysis of Chemical Composition, Source and Processing Characteristics of
  Submicron Aerosol during the Summer in Beijing, China, Aerosol Air Qual. Res., 19(6), 1450–1462,
  doi:10.4209/aaqr.2018.12.0480, 2019.
- Katsanos, D., Bougiatioti, A., Liakakou, E., Kaskaoutis, D. G., Stavroulas, I., Paraskevopoulou, D., Lianou, M.,
  Psiloglou, B. E., Gerasopoulos, E., Pilinis, C. and Mihalopoulos, N.: Optical Properties of Near-Surface Urban
  Aerosols and their Chemical Tracing in a Mediterranean City (Athens), Aerosol Air Qual. Res., 19(1), 49–70,
  doi:10.4209/aaqr.2017.11.0544, 2019.
- Levy, M. E., Zhang, R., Zheng, J., Tan, H., Wang, Y., Molina, L. T., Takahama, S., Russell, L. M. and Li, G.:
  Measurements of submicron aerosols at the California–Mexico border during the Cal–Mex 2010 field campaign,
  Atmos. Environ., 88, 308–319, doi:10.1016/j.atmosenv.2013.08.062, 2014.
- 140 Lin, C., Ceburnis, D., Huang, R.-J., Xu, W., Spohn, T., Martin, D., Buckley, P., Wenger, J., Hellebust, S., Rinaldi,
- 141 M., Facchini, M. C., O'Dowd, C. and Ovadnevaite, J.: Wintertime aerosol dominated by solid-fuel-burning
- 142 emissions across Ireland: insight into the spatial and chemical variation in submicron aerosol, Atmos. Chem.
- 143 Phys., 19(22), 14091–14106, doi: 10.5194/acp-19-14091-2019, 2019.
- Minguillón, M. C., Ripoll, A., Pérez, N., Prévôt, A. S. H., Canonaco, F., Querol, X. and Alastuey, A.: Chemical
  characterization of submicron regional background aerosols in the western Mediterranean using an Aerosol
  Chemical Speciation Monitor, Atmos. Chem. Phys., 15(11), 6379–6391, doi:10.5194/acp-15-6379-2015, 2015.
- Pirinen, P., Simola, H., Aalto, J., Kaukoranta, J. P., Karlsson, P. and Ruuhela, R.: Statistics on the Finnish climate
  1981-2010, Finnish Meteorological Institute, 2012.
- Qin, Y. M., Tan, H. B., Li, Y. J., Schurman, M. I., Li, F., Canonaco, F., Prévôt, A. S. H. and Chan, C. K.: Impacts of traffic emissions on atmospheric particulate nitrate and organics at a downwind site on the periphery of Guangzhou, China, Atmos. Chem. Phys., 17(17), 10245–10258, doi:10.5194/acp-17-10245-2017, 2017.
- 152 Ripoll, A., Minguillón, M. C., Pey, J., Jimenez, J. L., Day, D. A., Sosedova, Y., Canonaco, F., Prévôt, A. S. H.,
- 153 Querol, X. and Alastuey, A.: Long-term real-time chemical characterization of submicron aerosols at Montsec
- 154 (southern Pyrenees, 1570 m a.s.l.), Atmos. Chem. Phys., 15(6), 2935–2951, doi:10.5194/acp-15-2935-2015, 2015.

- Salcedo, D., Alvarez-Ospina, H., Peralta, O. and Castro, T.: PM1 Chemical Characterization during the ACU15
   Campaign, South of Mexico City, Atmosphere, 9(6), 232, doi:10.3390/atmos9060232, 2018.
- 157 Schlag, P., Kiendler-Scharr, A., Blom, M. J., Canonaco, F., Henzing, J. S., Moerman, M., Prévôt, A. S. H. and
- 158 Holzinger, R.: Aerosol source apportionment from 1-year measurements at the CESAR towerin Cabauw, the
- 159 Netherlands, Atmos. Chem. Phys., 16(14), 8831–8847, doi:10.5194/acp-16-8831-2016, 2016.
- Sujatha, P., Mahalakshmi, D. V., Ramiz, A., Rao, P. V. N. and Naidu, C. V.: Ventilation coefficient and boundary
  layer height impact on urban air quality, edited by Z. Wang, Cogent Environ. Sci., 2(1),
  doi:10.1080/23311843.2015.1125284, 2016.
- 163 Sun, Y.-L., Zhang, Q., Schwab, J. J., Demerjian, K. L., Chen, W.-N., Bae, M.-S., Hung, H.-M., Hogrefe, O.,
- 164 Frank, B., Rattigan, O. V. and Lin, Y.-C.: Characterization of the sources and processes of organic and inorganic
- aerosols in New York city with a high-resolution time-of-flight aerosol mass apectrometer, Atmos. Chem. Phys.,
- 166 11(4), 1581–1602, doi:10.5194/acp-11-1581-2011, 2011.
- 167 Teinilä, K., Aurela, M., Niemi, J. V., Kousa, A., Petäjä, T., Järvi, L., Hillamo, R., Kangas, L., Saarikoski, S. and
- 168 Timonen, H.: Concentration variation of gaseous and particulate pollutants in the Helsinki city centre —
- 169 observations from a two-year campaign from 2013–2015, Boreal Env. Res., 24, 22, 2019.
- 170 Tiitta, P., Vakkari, V., Croteau, P., Beukes, J. P., van Zyl, P. G., Josipovic, M., Venter, A. D., Jaars, K., Pienaar,
- 171 J. J., Ng, N. L., Canagaratna, M. R., Jayne, J. T., Kerminen, V.-M., Kokkola, H., Kulmala, M., Laaksonen, A.,
- Worsnop, D. R. and Laakso, L.: Chemical composition, main sources and temporal variability of PM 1 aerosols
- 173 in southern African grassland, Atmos. Chem. Phys., 14(4), 1909–1927, doi:10.5194/acp-14-1909-2014, 2014.
- Xu, W., Sun, Y., Wang, Q., Zhao, J., Wang, J., Ge, X., Xie, C., Zhou, W., Du, W., Li, J., Fu, P., Wang, Z.,
  Worsnop, D. R. and Coe, H.: Changes in Aerosol Chemistry From 2014 to 2016 in Winter in Beijing: Insights
  From High-Resolution Aerosol Mass Spectrometry, J. Geophys. Res. Atmos., 124(2), 1132–1147,
  doi:10.1029/2018JD029245, 2019.
- Young, D. E., Kim, H., Parworth, C., Zhou, S., Zhang, X., Cappa, C. D., Seco, R., Kim, S. and Zhang, Q.:
  Influences of emission sources and meteorology on aerosol chemistry in a polluted urban environment: results
  from DISCOVER-AQ California, Atmos. Chem. Phys., 16(8), 5427–5451, doi:10.5194/acp-16-5427-2016, 2016.
- 181 Zhang, Y., Favez, O., Petit, J.-E., Canonaco, F., Truong, F., Bonnaire, N., Crenn, V., Amodeo, T., Prévôt, A. S.
- 182 H., Sciare, J., Gros, V. and Albinet, A.: Six-year source apportionment of submicron organic aerosols from near-183 continuous highly time-resolved measurements at SIRTA (Paris area, France), Atmos. Chem. Phys., 19(23), 14755 14776 bio 105104/ up 1014755 2010 2010
- 184 14755–14776, doi: 10.5194/acp-19-14755-2019, 2019.