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Supplementary Information of

Intervention of pollution episodes from nearby sawmills to ecosystem-atmosphere interactions studied in a boreal forest at SMEAR II

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S1. Wind conditions at SMEAR II

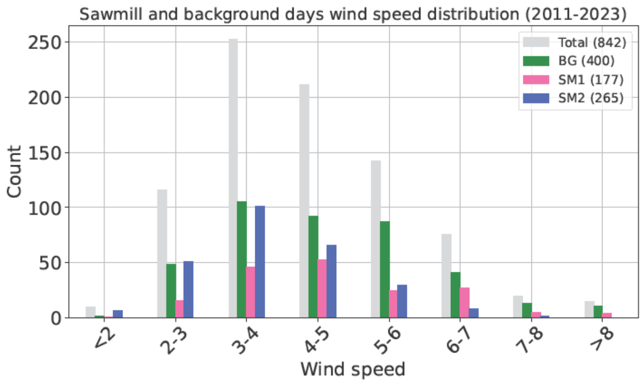

Figure S1. Daily average wind speed distribution at SMEAR II from 67.2 m height for each sector.

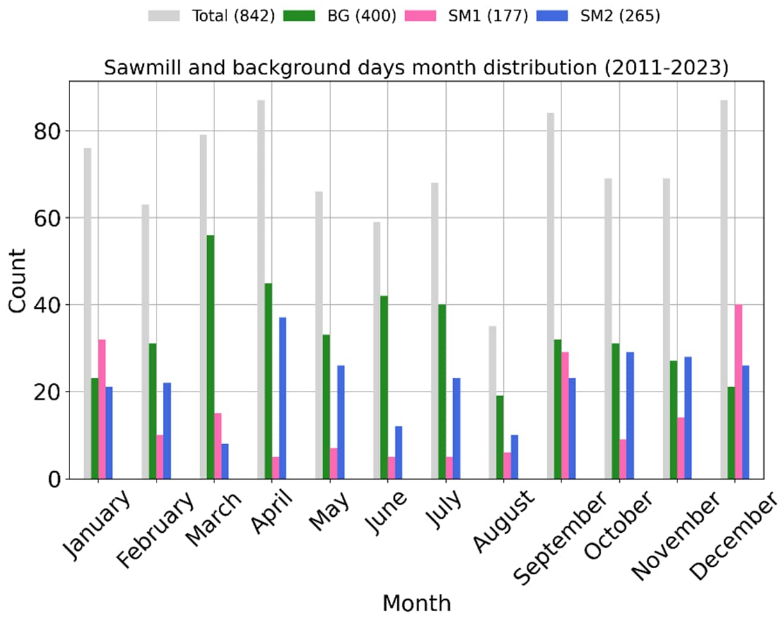

Figure S2. Monthly distribution of sawmill episode (SM1 / SM2) and background (BG) days.

S2. VOC concentration dynamics

Table S1. Difference in the seasonal median concentration of individual VOCs between SM1 and BGs days, and SM2 and BG days, respectively. The difference is expressed in ppt and as a factor showing how much higher the concentrations during sawmill episode days are compared to the BG days.

VOC	Δ_{SM1} (ppt)	Δ_{SM1} (factor)	Δ_{SM2} (ppt)	Δ_{SM2} (factor)
Monoterpenes				
spring	30	2.31	10	1.49
summer	20	1.15	60	1.42
autumn	280	7.08	40	1.88
winter	100	4.46	30	2.25
Acetaldehyde				
spring	240	2.08	20	1.09
summer	200	1.94	50	1.25
autumn	220	2.47	4	1.03
winter	160	1.94	90	1.53
Methacrolein+MVK				
spring	20	2.65	-1	0.90
summer	30	1.93	20	1.50
autumn	2	3.46	10	1.91
winter	10	1.52	1	1.01
Benzene				
spring	90	1.97	10	1.17
summer	30	2.68	4	1.23
autumn	60	2.32	1	1.03
winter	130	1.93	70	1.53
Toluene+p-cymene				
spring	70	4.78	-10	0.95
summer	90	1.16	30	1.05
autumn	90	3.50	10	1.22
winter	70	2.29	40	1.64
Isoprene+MBO				
spring	30	2.89	3	1.23
summer	-2	0.96	20	1.25
autumn	40	3.03	10	1.36
winter	30	3.32	10	1.59
MEK				
spring	40	1.57	20	1.35
summer	120	2.91	10	1.24
autumn	70	2.73	10	1.21
winter	20	1.34	30	1.55

S3. Trace and greenhouse gases

Table S2. The number of trace and greenhouse data points found for each sector.

Gas	BG	SM1	SM2
CO_2	39 162	16 225	31 624
CH_4	19 449	7165	17 022
CO	56 690	26 524	38 551
NO	37 640	18 751	25 887
NO_x	52 607	27 201	37 192
O_3	58 019	27 490	39 806
SO_2	221 898	127 403	179 663

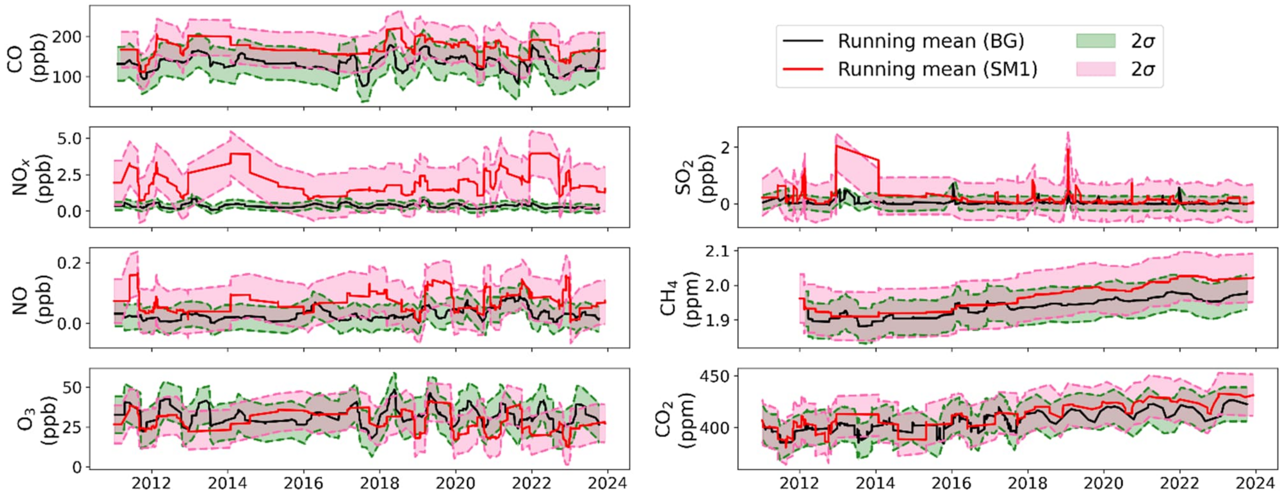

Figure S3. Running mean time series for trace and greenhouse gases in SM1 and BG sectors with 2σ deviation.

For time series analysis, a sector-based approach was implemented, focusing on SM1 and BG sectors (Fig. S2). The time series were discontinuous due to the sector selection, and for that reason, a running mean with a window size of 1000 points was applied. The choice of the window size was driven by the need for statistical stability in the analysis. The chosen value allowed smoothing out short-term fluctuations and revealed the underlying trends in the data. The variations were evaluated based on a 95 % confidence interval, which was smoothed by computing the coefficients of interpolating B-spline. The 2σ standard deviation around the running mean highlights the overlap in the observed concentration values between sectors SM1 and BG.

Due to the non-stationary trend observed in the CO_2 and CH_4 data (Fig. S2), a linear detrending was applied, which consists of performing a linear regression and subtracting the trend from the whole time series to obtain only the variations. This was essential to remove linear trend, ensuring our analysis accurately reflected variations around the mean in Fig. 8.

In Fig. S3, the polar plots illustrate the daily median gas concentrations as a function of both wind direction and wind speed. These plots reveal the patterns of gas dispersion in relation to wind direction, indicating that the BG sector (North-West) exhibits lower (higher for O_3) concentrations compared to other directions. Additionally, the polar plots increased concentrations of CO, NO_x, SO_2, CH_4, and O_3 (decreased) for directions spanning from North-East to South-West. This includes directions where wind originates from the same direction as SM2 (NE), SM1 (SE), and Tampere (SW).

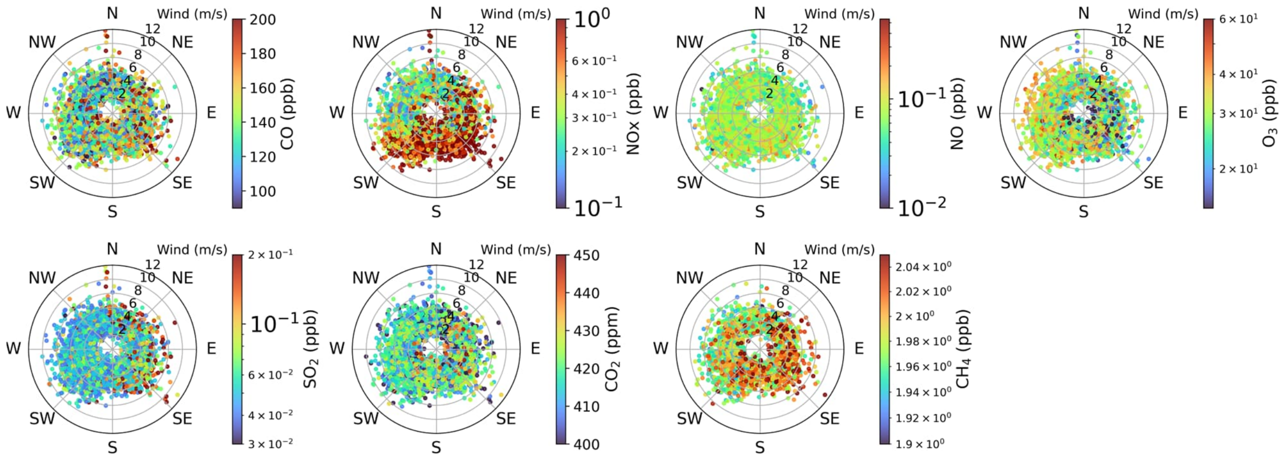

Figure S4. Polar plots of daily median gas concentrations as a function of wind direction and wind speed.

S4. HOMs

The ratio between HOMs and MTs versus O_3 concentration shows that in general, the O_3 concentration is lower for SM1 compared to the BG data (Fig. S5). In the BG data the separation after 2022 and other years is clear, but in SM1 there is no clear distinction in a trend between data before and after 2022. In addition, in none of the cases there is a linear correlation between ratio and O_3. This is confirmed by the correlation coefficient, which remained below 0.15.

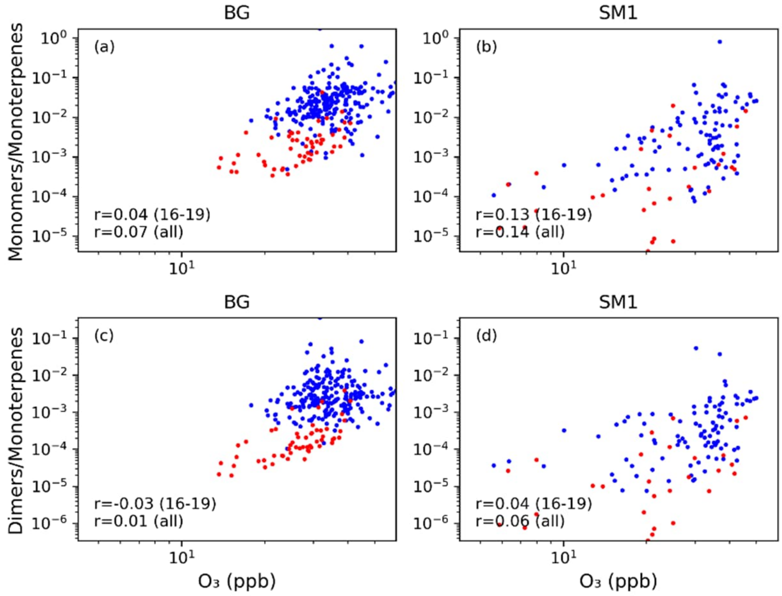

Figure S5. Ratio of HOM monomers and MTs (a, b), and ratio of HOM dimers and MTs (c, d) versus O_3. Blue dots represent the hourly median of data measured between 2016 and 2019; red dots represent the hourly median of data measured after 2022. Each subplot contains the correlation coefficient for data measured between 2016 and 2019 (*r* (16-19)), and for all data available (*r* (all)).

Figure S5 shows that BG data are scattered and there is no correlation between HOMs and negative ions. In SM1 (Fig. S5c, d), the correlation coefficient is about zero if all the data are considered. However, if data after 2022 are disregarded in the computation, then HOMs and negative ions show a weak positive correlation for the SM1 sector. In fact, for HOM monomers the correlation coefficient is 0.2 with p-value of 0.03, while for HOM dimers the correlation coefficient is 0.22 with p-value of 0.02. This means that the correlation is positive and low, but statistically significant because the p-values are lower than the commonly used significance threshold (0.05). The biggest difference between BG and SM1 is that the former shows lower values of negative ions. Nevertheless, the data availability for SM1 is very limited; thus, strong conclusions on the SM1 activity cannot be drawn.

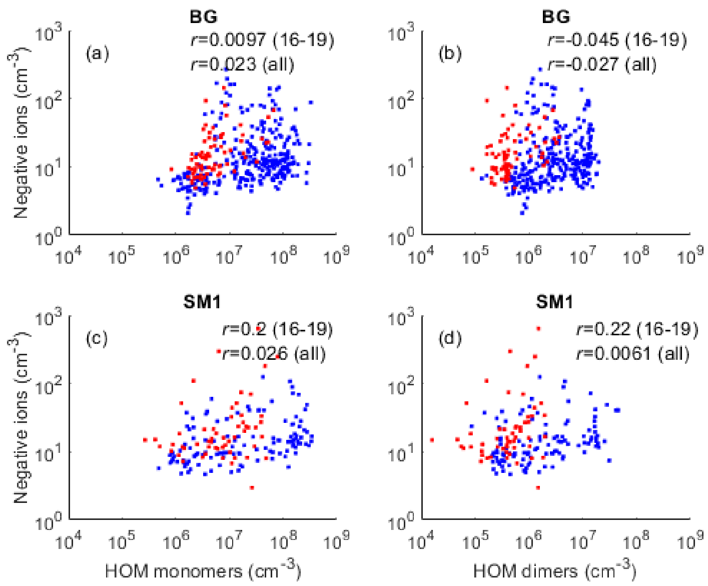

Figure S6. Negative ion concentration (2–7 nm) versus (a,c) HOM monomers and (b,c) HOM dimers. Blue dots represent the hourly median of data measured between 2016 and 2019; red dots represent the hourly median of data measured after 2022. Each subplot contains the correlation coefficient for data measured between 2016 and 2019 (*r* (16-19)), and for all data available (*r* (all)).

S5. VOC/OA

A possible correlation between volatile organic compounds VOCs and organic aerosols (OA) was explored by evaluating the ratios VOC/OA for the BG (43 points), SM1 (23 points), and SM2 (34 points; Fig. S6). Furthermore, the VOC/OA ratios were evaluated as functions of O_3, temperature, and photosynthetically active radiation (PAR; Fig. S7), as these variables may affect the oxidation. No correlation was found for the SM2 sector, while the VOC/OA ratios were slightly lower for MTs, acetaldehyde, methacrolein+MVK, benzene, and toluene+p-cymene for the SM1 sector compared to the BG. While some differences can be seen between the BG and SM1 sector for the relations to O_3, temperature, and PAR, it is not systematic enough and based on too limited data to draw any reliable conclusions.

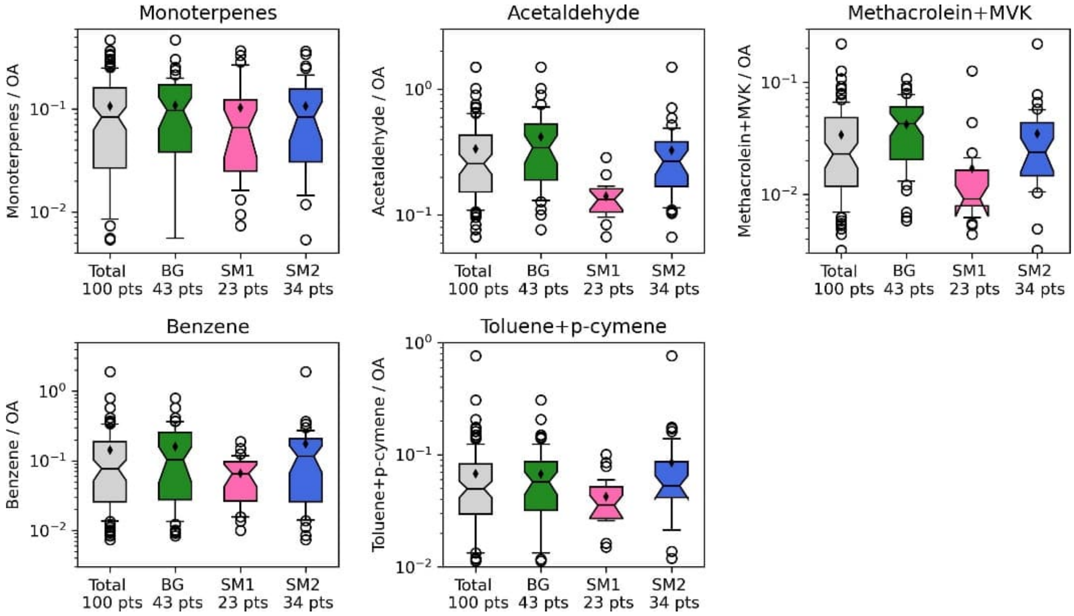

Figure S7. Ratio, in ppb (µg m^{-3})$^{-1}$, of daily median of select VOCs (MTs, acetaldehyde, methacrolein+MVK, benzene, and toluene+p-cymene) to daily median of organic aerosols for the total measurements, background-, SM1-, and SM2-sectors. The median is shown with a black line, the mean value with a black diamond point, and outliers with black circles. The box edges extend to 25th and 75th percentiles while whiskers display the 10th and 90th percentiles. Overlapping notches indicate that the medians do not differ statistically significantly in 95 % confidence level. The number of data points (equal to the number of days) is shown below the plots.

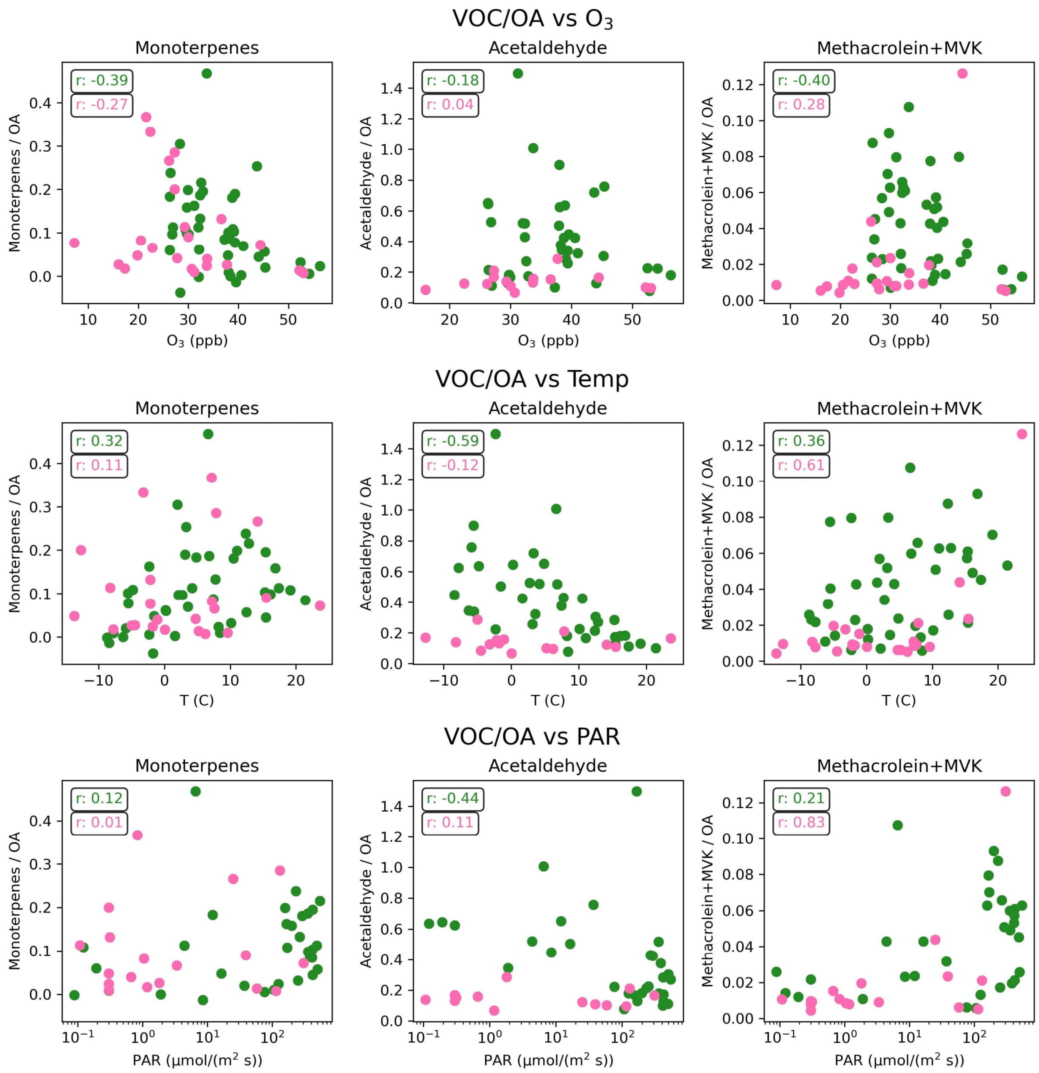

Figure S8. Scatter plots for the ratio, in ppb (μg m^{-3})$^{-1}$, of the daily median of MTs, acetaldehyde, and methacrolein+MVK to the daily median of organic aerosols as functions of the daily medians of O_3 (top row), temperature (middle row), and PAR (bottom row). Background is indicated by green, and SM1 by pink, and Pearson's correlation coefficient is *r* respectively. SM2 is not shown as it behaved like the background.

S6. Case study

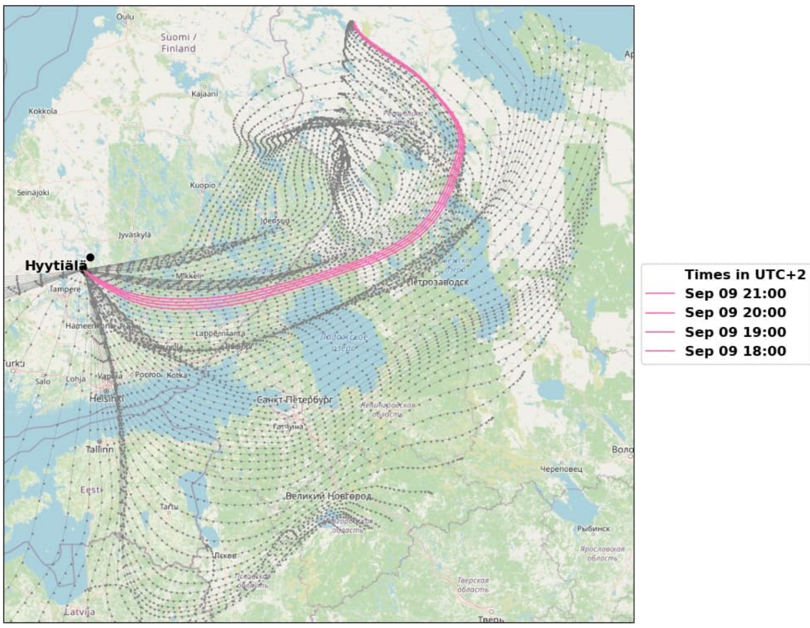

Figure S9. Air mass trajectories reaching SMEAR II during the case study of 8.9.2018 to 13.9.2018 are shown with gray color, and trajectories passing SM1 closer than 1 km are shown in pink. The time stamps indicate when the air masses reach SMEAR II at 67.2 m height.

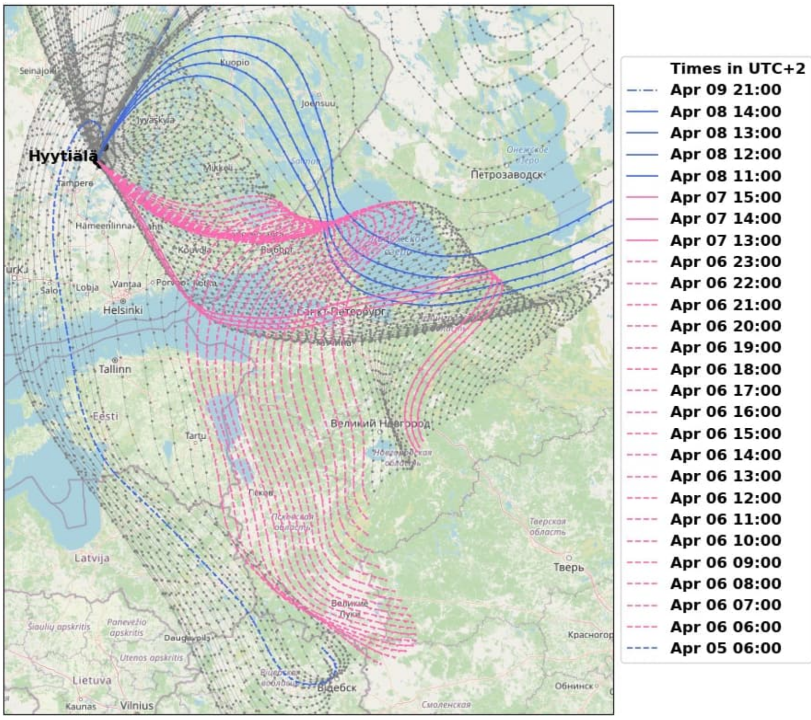

Figure S10. Air mass trajectories reaching SMEAR II during the case study of 5.4.2019 to 12.4.2019 are shown with gray color and trajectories passing SM1 and SM2 closer than 1 km are marked in pink and in blue, respectively. The time stamps indicate when the air masses reach SMEAR II at

33.6 m height. Consecutive hours with air coming from the same sawmill are marked with the same line style.

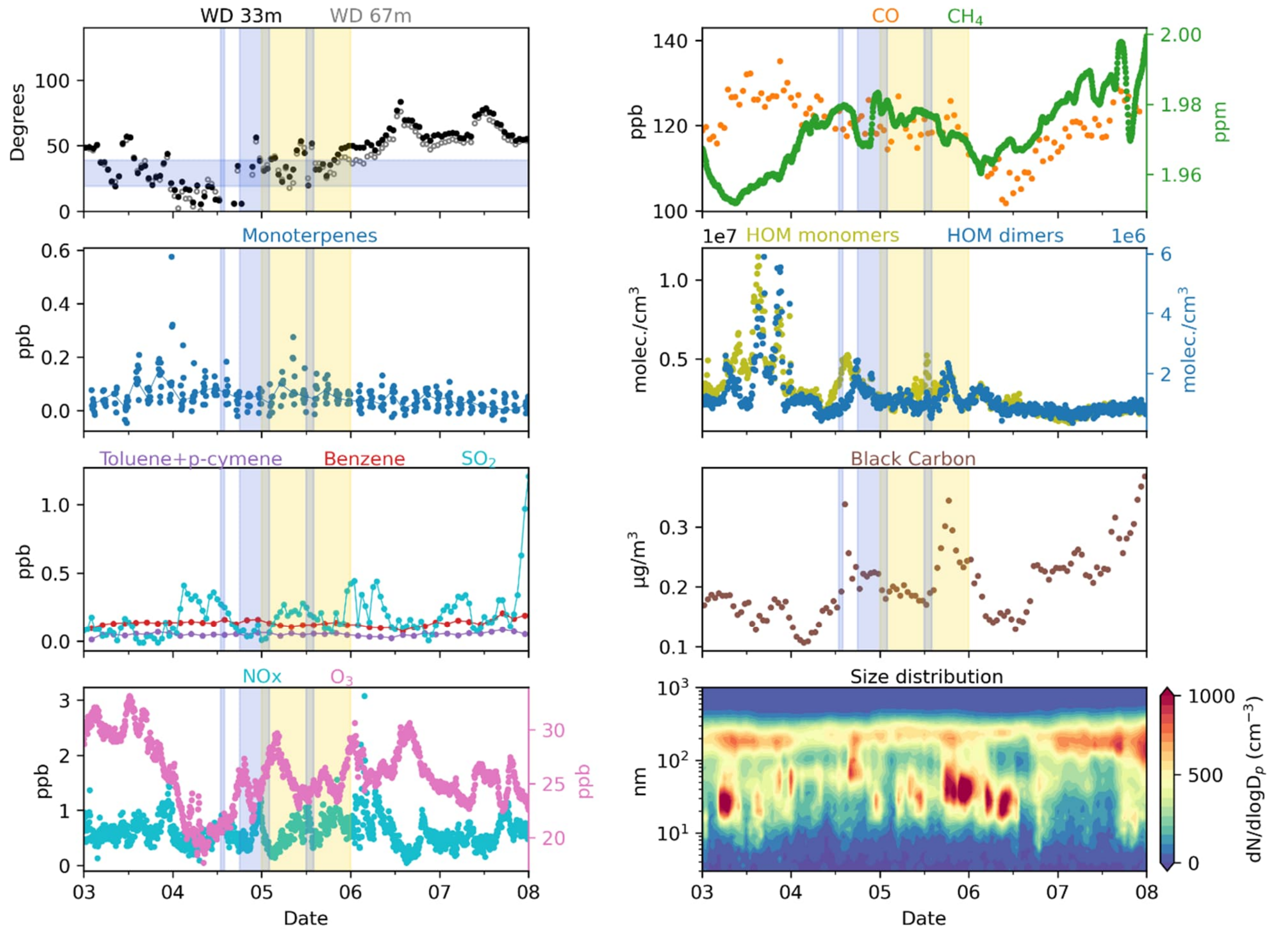

Figure S11. Case study of 3.11.2016 to 8.11. containing wind directions, concentrations of MTs, benzene, toluene+p-cymene, SO_2, NO_x, O_3, CO, CH_4, HOM monomers and HOM dimers, black carbon, and OA as well as particle size distribution. Organic aerosol (OA) concentration was missing for this time period. The day classified as sawmill episode day is indicated by the yellow shaded area, the times when air mass trajectories crossed the SM1 sector are indicated by the vertical blue shaded areas, and the direction of the SM2 sector is indicated by the horizontal blue shaded area. The wind directions, black carbon, benzene, toluene+p-cymene, and SO_2 are one-hour medians, and the one-hour median for the MTs are indicated by the blue line.

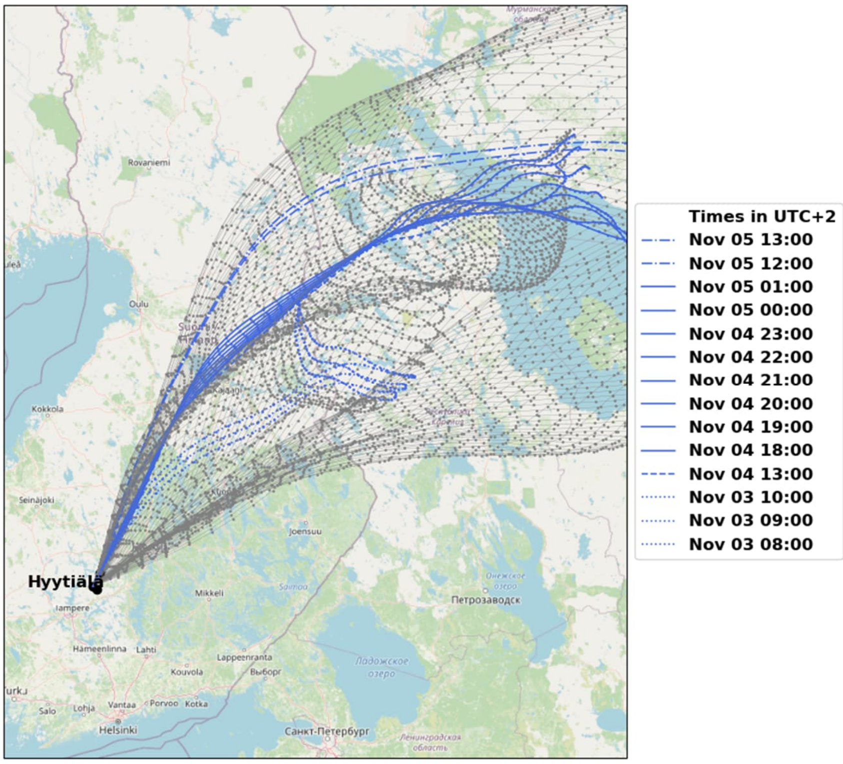

Figure S12. Air mass trajectories reaching SMEAR II during the case study of 3.11.2016 to 8.11.2016 are shown with gray color and trajectories passing SM2 closer than 1 km are marked in blue. The time stamps indicate when the air masses reach SMEAR II at 33.6 m height. Consecutive hours with air coming from the same sawmill are marked with the same line style.