

Supplement Figures

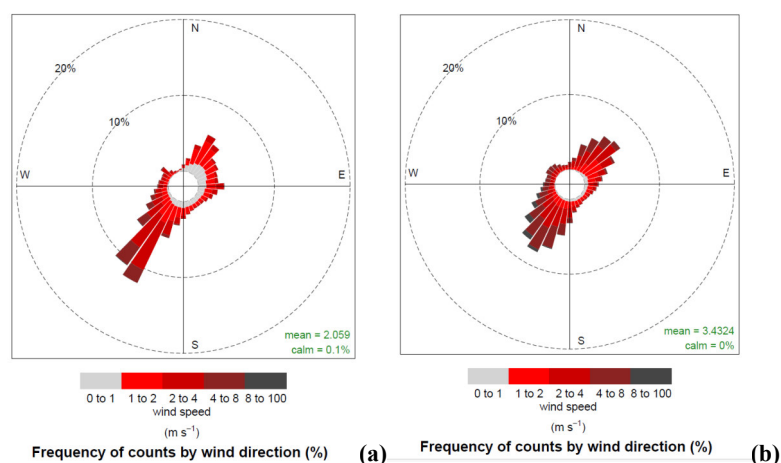


Figure S1. Wind roses on hourly averages for years 2015-2021 (2016 excluded) for LAN (a) and FFM_AP (b) based.

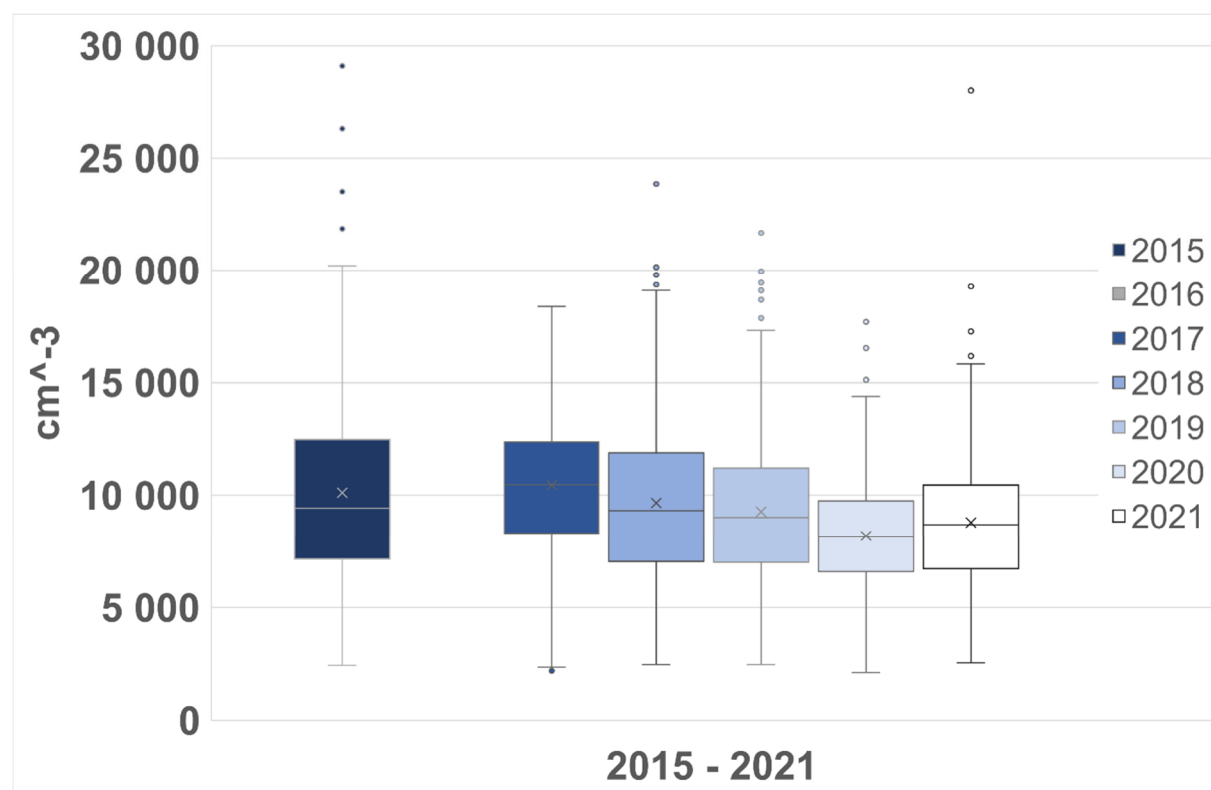


Figure S2. Box plot of daily TNC (10 – 500 nm), 2015 – 2021 in LAN. Quartiles upper box 75 percentile, lower 25 perc.; median center line, upper/lower end of line 98/2 percent.

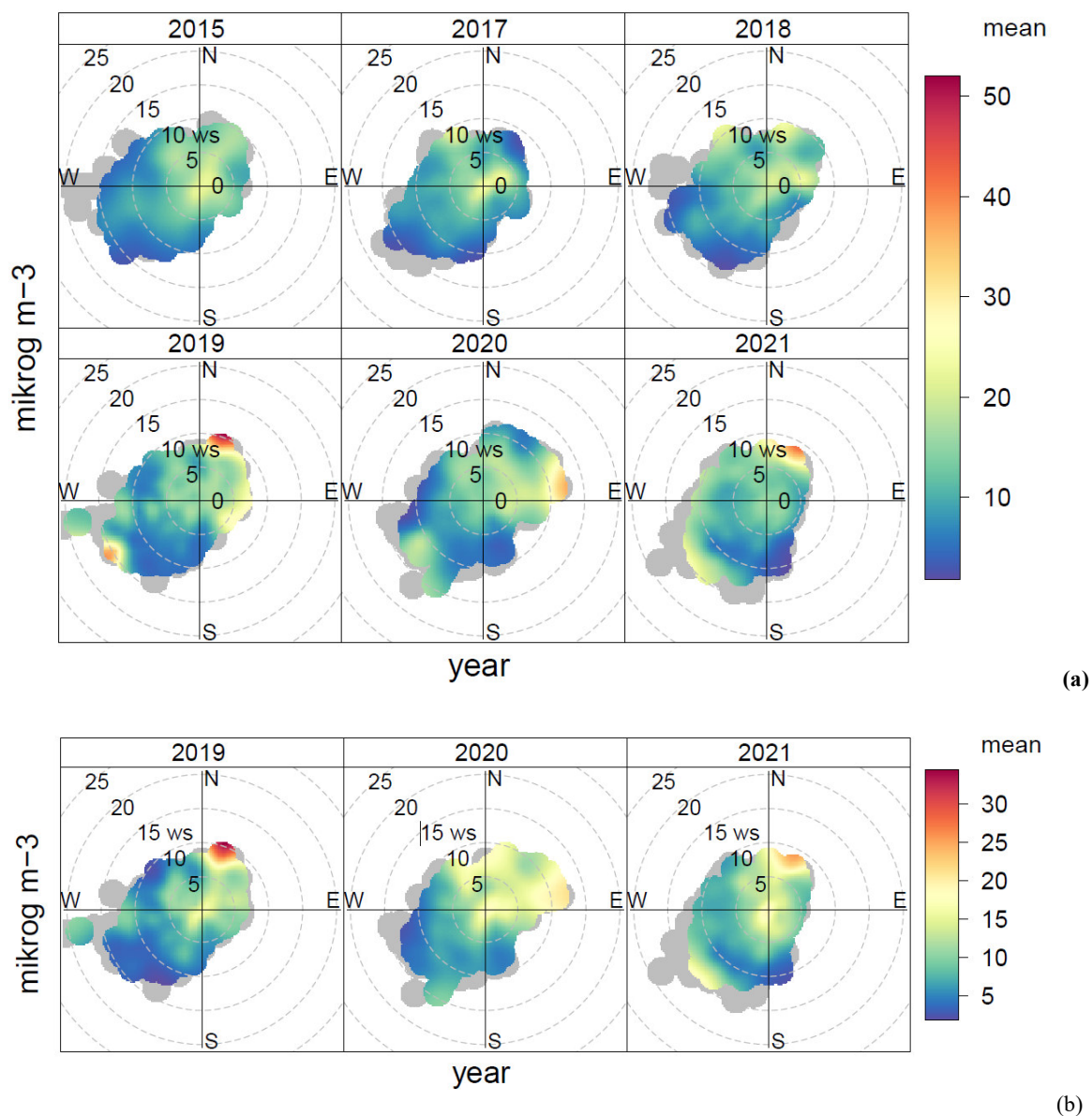


Figure S3. Polarplot of hourly averages of PM10 for separate years, DAR: 2015-2021 (2016 excluded), (a) and LAN: 2019-2021 (b).

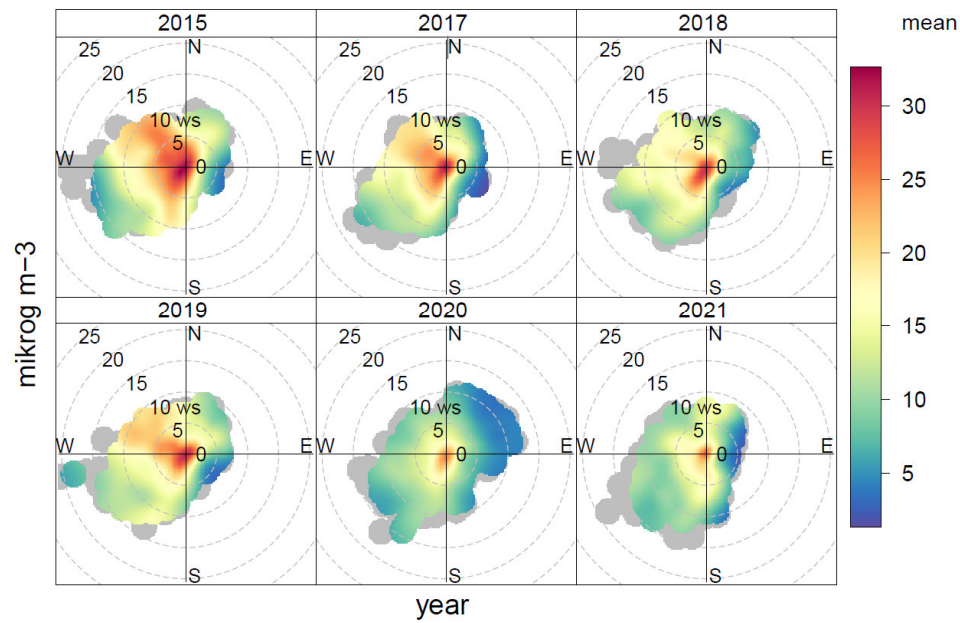


Figure S4. Polarplot of hourly averages of NO₂, separate years, DAR: 2015-2021 (2016 excluded).

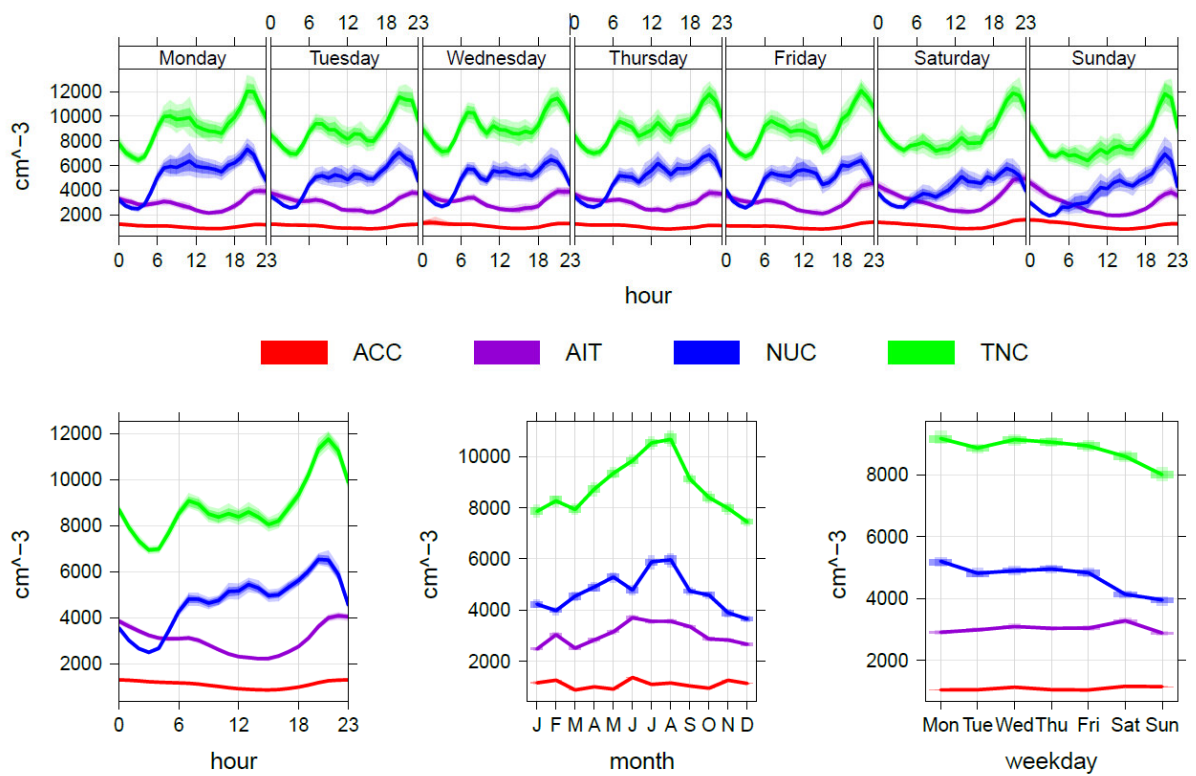


Figure S5. TNC and NUC, AIT, ACC: Timevariations based on hourly averages for the years 2019-2021 in LAN: (a) Weekday diurnal variation in an average week, (b) the diurnal variation of hourly averages, (c) The annual cycle of monthly averages, (d) The weekday variation of single days. Shaded areas represent 75 % and 95% confidence intervall.

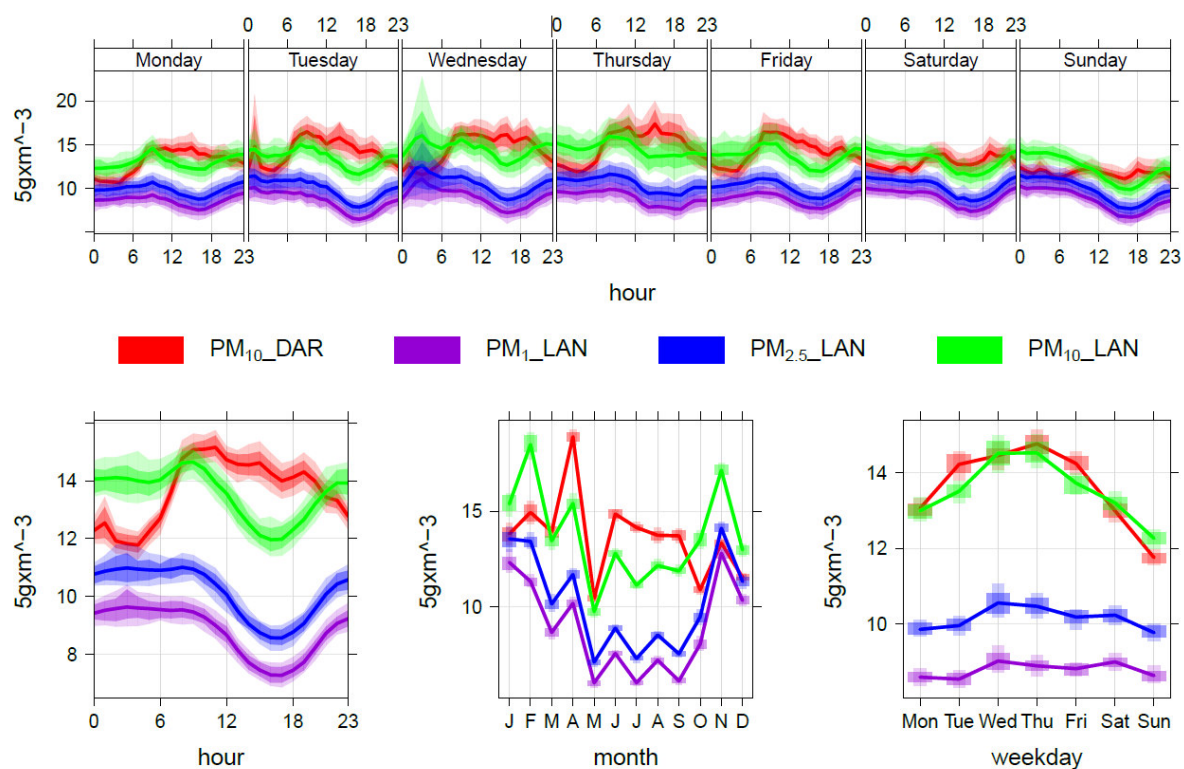


Figure S6. PMmass size classes in LAN and DAR. Detailed description s. caption Fig S3.

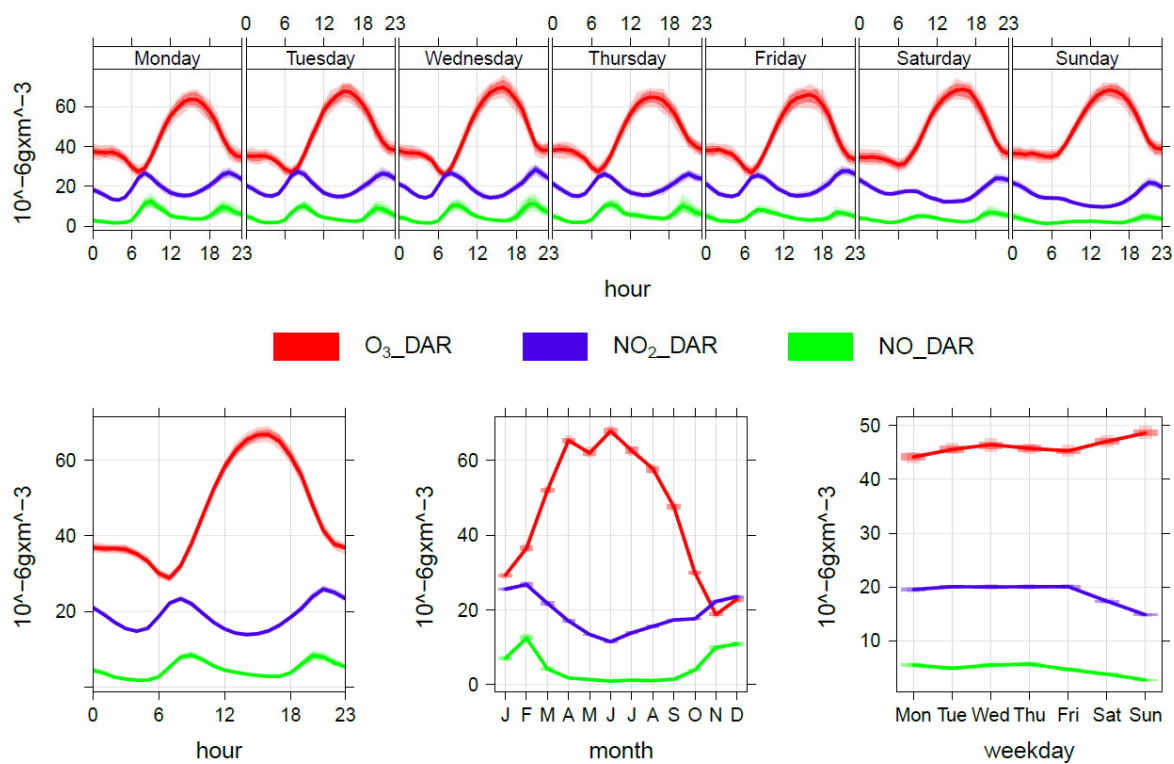


Figure S7. O_3 , NO_2 , NO in DAR. Detailed description s. caption Fig S3.

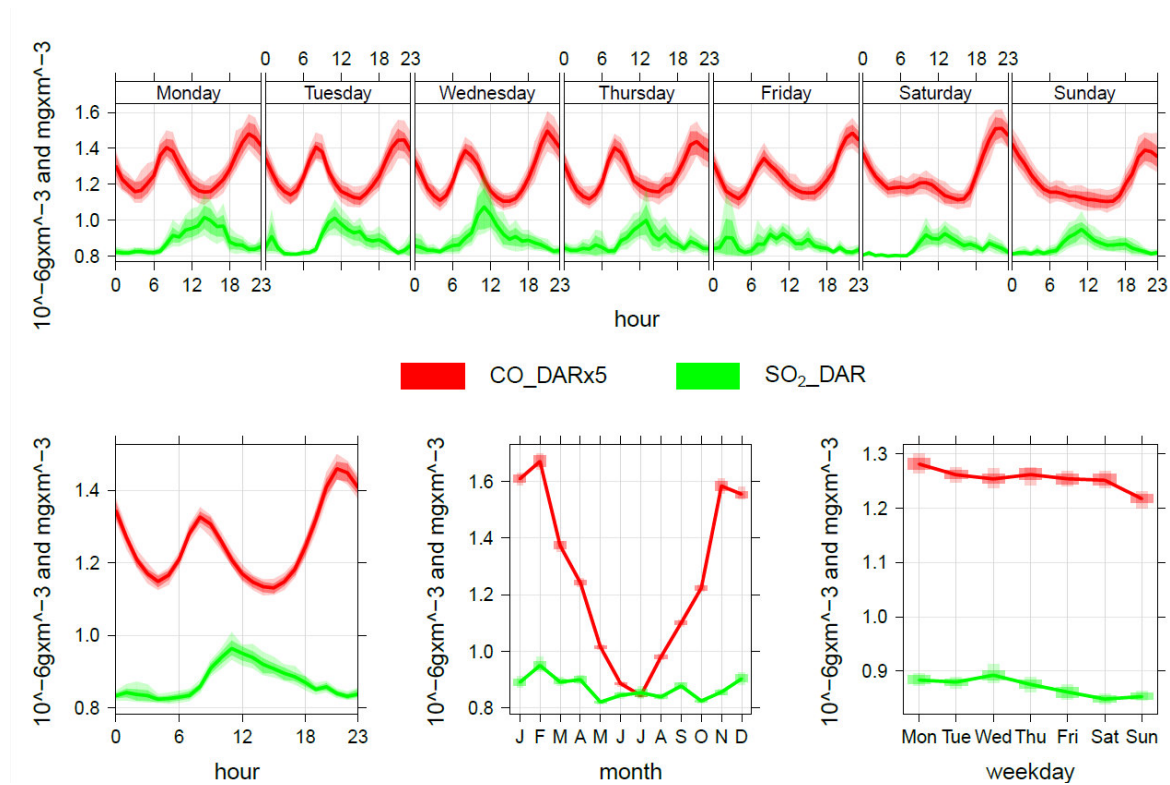


Figure S8. CO and SO₂ in DAR. Detailed description s. caption Fig S3.

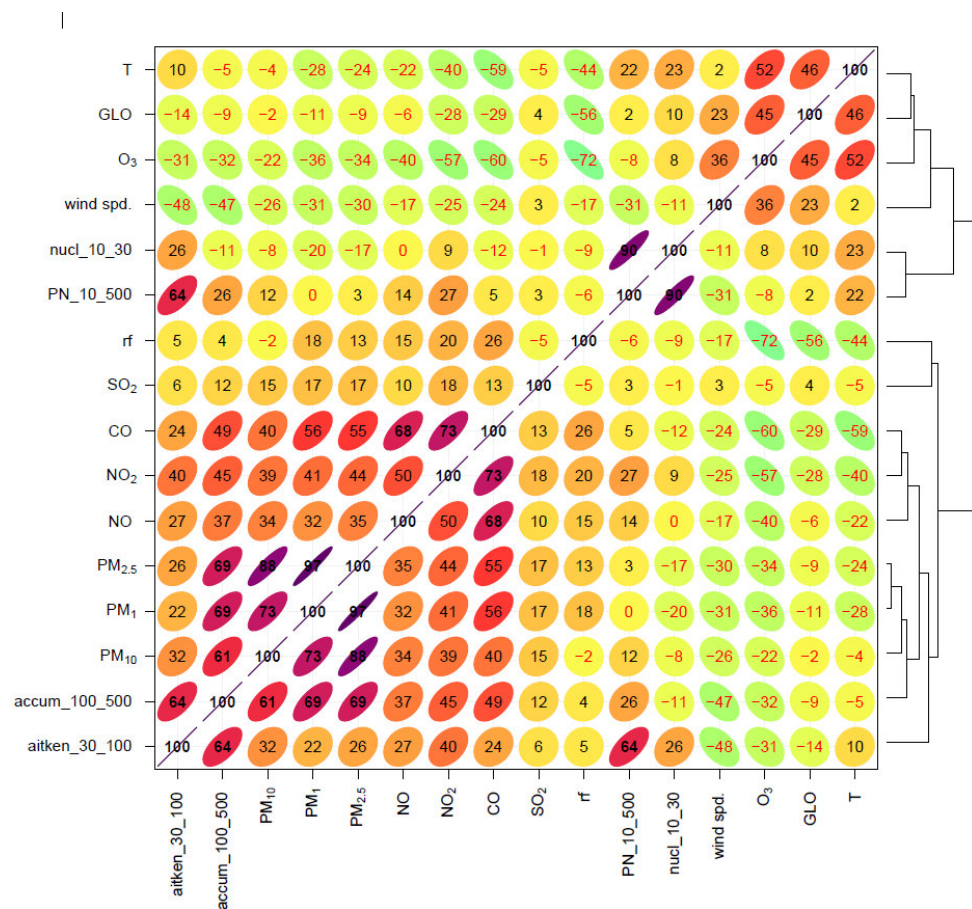


Figure S9. Correlation matrix, 2021, in LAN: Particle fractions, FFM_{AP}: Meteorology and DAR: gaseous compounds. Numbers representing $x = r \cdot 100$, printed in bold if $r > 0.6$, in red $r < 0$. Dendrogram on the right visualizes connected variables

Supplement Tables

Table S1. Instruments used to measure UFP-PSD and PM mass at LAN. ND, Not-determined; SMPS, Scanning Mobility Particle Sizer; CPC, Condensation Particle Counter; DMA, Differential Mobility Analyser; UB, Urban Background.

Parameter	Equipment	Type	Time Interval	Size range
PNSD	SMPS + CPC	TSI 3936 with X-Ray neutraliser, TSI 3010 (until 06-2018), TSI 3772 (from 07-2018)	2015 – 2021 (-2016)	10-500 nm
PNC	CPC	TSI 3750	2021	10 – 1000 nm
PM mass	OPC	PALAS, FIDAS 200E	2019 – 2021	<1 µm, <2.5 µm, <10 µm
Solar radiation	pyranometer	Kipp+Zonen, CMP21m	2018 – 2021	-
T, rH, p, wd, ws	weather station	LUFFT WS600:	2015 – 2021	-

Table S2. Hourly Concentrations 2015-2021 (2016 excluded) of auxiliary parameters and PM10, at DAR. Data coverage in % p.a..

	in	Min	1perc	Median	Mean	SD	99perc	Max	coverage (%)
PM10	µg/m ³	0.5	2.2	12.9	16	12	55.6	499.9	99
SO2	µg/m ³	0.8	0.8	0.8	1	1	3.1	32.5	100
NO	µg/m ³	0.4	0.4	1.3	6	15	77.2	288.9	100
NO2	µg/m ³	0.6	3.6	17.6	21	13	65	124.5	100
O3	µg/m ³	0.6	0.6	42.3	45	32	138.2	253	100
CO	mg/m ³	0.03	0.08	0.23	0	0	0.63	1.36	100

Table S3. Hourly Concentrations for 2021 at Langen (LAN) for size modes, PM10, PM2.5 and PM1, T, Temp T; rH, relative humidity; GLO, global radiation and ws, wind speed; Darmstadt (DAR) for auxiliary parameters and PM10, PM2.5.

	in	Min.	1perc	Median	Mean	SD	99perc	Max	Coverage In %
TNC, N10-500	1/cm ³	957	1 799	7 857	8 793	5 046	26 537	54 212	92
NUC, N10-30	1/cm ³	165	649	3 851	4 932	4 177	21 565	45 555	92
AIT, N30-100	1/cm ³	143	417	2 482	2 858	1 731	8 425	12 429	92
ACC, N100-500	1/cm ³	46	129	884	1 002	644	3 106	5 804	92
N30-500	1/cm ³	189	604	3 431	3 861	2 201	10 693	16 553	92
PM1	µg/m ³	0.3	0.9	7.6	8.9	6.2	28.1	36.8	100
PM2.5	µg/m ³	0.4	1.2	8.9	10.3	6.9	33.7	54.0	100
PM10	µg/m ³	0.7	2.1	12.5	14.4	10.6	48.3	148.0	100
PM2.5 DA	µg/m ³	0.5	1.3	7.4	8.8	5.9	28.1	89.5	99
PM10 DA	µg/m ³	0.5	2.3	11.1	13.2	9.6	46.7	126.3	99
SO2	µg/m ³	0.8	0.8	0.8	0.8	0.3	2.2	8.9	100
NO	µg/m ³	0.4	0.4	1.2	4.2	10.2	52.9	145.9	100
NO2	µg/m ³	0.9	3.0	14.3	17.1	11.2	57.2	113.5	100
O3	µg/m ³	0.6	0.6	39.9	42.3	28.8	115.7	166.7	100
CO	mg/m ³	0.08	0.11	0.21	0.24	0.09	0.54	0.83	100
T	°C	-10.5	-5.8	9.6	11	8	27.8	34.2	100
rH	%	23	32	79	76	17	98	100	100
Ws	m/s	0.2	0.6	2.8	3	2	10.1	17.8	99
GLO	W/m ²	27	29	39	165	213	871	991	98

Table S4. Theil-Sen trend in % per year for 2015 to 2021 (2016 excluded) and in additional column for comparison 2015 to 2019 of TNC and modes based on monthly measurements based on hourly concentrations at Langen (LAN) and Darmstadt (DAR) for PM10 and auxiliary components and T (FFM_AP). *** p< 0.001, ** p< 0.01, * p< 0.05. D.o.s. = Degree of significance.

		2015-2021				2015-2019			
	in	Mean	Min	Max	D.o.s.	Mean	Min	Max	D.o.s.
TNC, N10-500	1/cm ³	-2.3	3.1	-1.4	***	-1.9	-2.8	-0.3	*
NUC, N10-30	1/cm ³	-2.9	-3.9	-1.7	***	-2.0	-3.1	-0.7	**
AIT, N30-100	1/cm ³	-2.0	-3.0	-0.8	**	1.2	-0.9	4.4	-
ACC, N100-500	1/cm ³	-0.9	-3.2	1.8	-	3.3	0.3	9.0	*
PM10 DA	µg/m ³	-4.3	-5.1	-3.1	***	-4.4	-5.4	-2.5	***
SO2	µg/m ³	-2.3	-2.7	-1.9	***	-1.9	-2.7	-0.9	**
NO2	µg/m ³	-4.9	-5.4	-4.4	***	-3.9	-5.4	-2.2	***
O3	µg/m ³	1.5	0.1	3.3	*	4.4	2.0	7.0	**
CO	mg/m ³	3.5	1.7	6.0	***	7.3	3.8	14.3	***

Table S5. WHO values above certain amount of TNC 10 – 500 nm 2015 -2021 in LAN. Days per year TNC > 10 000 1/cm and hours per year TNC > 20 000 1/cm³. Only coverage >=55% are used.

year	days > x In %	coverage day in %	hours > y In %	coverage hour in %
2015	46	71	6	73
2016	<i>n.n.</i>	35	<i>n.n.</i>	37
2017	59	55	6	56
2018	42	59	5	60
2019	37	80	5	83
2020	23	73	2	76
2021	30	90	3	92