Supplementary material

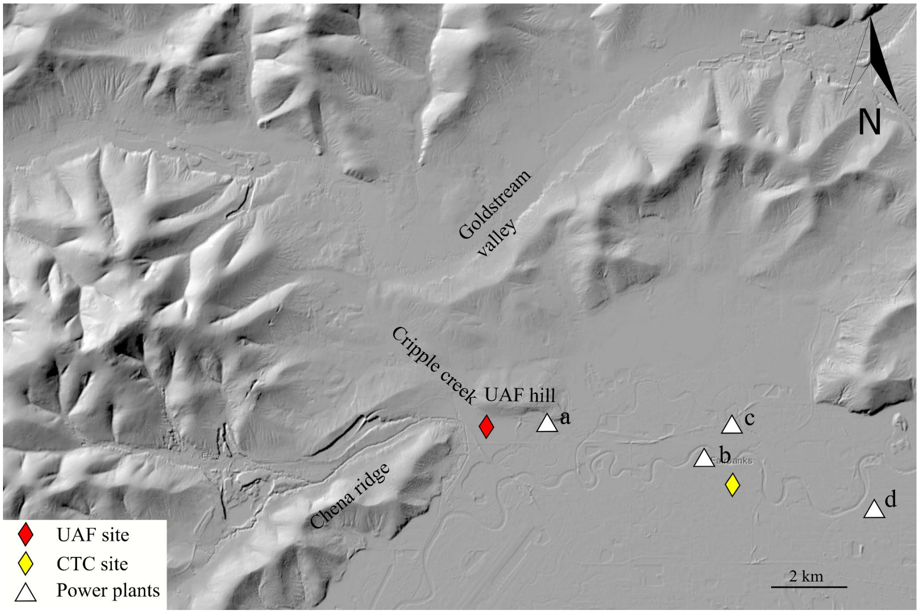

Figure S1 Topographic map of Fairbanks, Alaska, USA. The red and yellow diamonds represent the location of the UAF and CTC study sites, respectively. White triangles indicate the location of the power plants in Fairbanks. (a) UAF, (b) Aurora, (c) Zehnder and (d) Doyon (Fort Wainwright). The map was obtained and adapted from the United States Geological Survey (https://apps.nationalmap.gov/).

Table S1 List of flights. For the synoptic conditions, AC = anticyclonic and C = cyclonic. Instruments flown on specific flights are indicated by the following numbers: 1 = POPS, 2 = mSEMS, 3 = STAP, 4 = CO_2 monitor, 5 = CO monitor (Pico), 6 = O_3 monitor, 7 = MICROMEGAS multi gas sensor (see Table 1). A '-' in the last column indicates that no meteorological measurements are available for the flight.

Flight nr.	Date	Time	# of profiles	Maximum altitude	Instruments	Synoptic conditions	Temperature profile structure
1	2022-01-26	14:00 - 16:00	14	85	1, 4, 6,	AC	-
2	2022-01-27	23:00 - 00:20	4	290	1, 5, 6, 7	AC	Convex SBI[a]
3	2022-01-28	14:00 - 14:30	4	80	1, 3, 4, 6	AC	No SBI
4[d]	2022-01-30	6:00 - 10:40	8	350	1, 2, 4, 7	AC	Convex SBI

5	2022-01-31	14:00 - 16:00	10	85	1, 2, 3, 4, 7	AC	-
6[d]	2022-01-31	22:00 - 2:00	6	275	1, 2, 3, 4, 7	AC	Convex / S-shaped SBI
7[d]	2022-02-03	22:00 - 01:25	8	250	1, 3, 4, 5, 7	C	Convex SBI
8[d]	2022-02-04	02:00 - 03:05	4	180	1, 3, 4, 6, 7	C	Convex SBI
9[d]	2022-02-04	15:20 - 17:10	8	125	1, 3, 4, 5, 6	C	Convex SBI
10[c,d]	2022-02-06	22:50 - 00:30	2	225	1, 4	C	Convex SBI
11	2022-02-07	15:00 - 17:00	8	80	1, 2, 3, 4, 6, 7	C	No SBI
12	2022-02-08	22:00 - 01:00	8	250	1, 2, 3, 4, 7	C	No SBI
13	2022-02-09	01:00 - 03:00	4	330	1, 2, 3, 4, 7	C	No SBI
14[c]	2022-02-09	23:00 - 03:50	4	300	1, 4	C	Convex SBI[a]
15[d]	2022-02-10	17:00 - 19:00	10	140	1, 2, 3, 4, 5	C	Convex / S-shaped SBI
16[d]	2022-02-10	22:30 - 00:30	4	240	1, 2, 3, 4, 5, 7	C	S-shaped SBI
17	2022-02-19	15:00-17:00	8	110	1, 2, 3, 4, 5, 7	AC	Convex SBI / No SBI
18[c]	2022-02-19	21:30 - 03:30	2	280	4	AC	Convex SBI[b]
19[d]	2022-02-20	6:00 - 11:05	8	300	1, 2, 4, 7	AC	S-shaped SBI
20	2022-02-21	13:00 - 15:00	8	150	1, 2, 4, 7	AC	No SBI
21	2022-02-22	22:00 - 03:00	2	300	1, 4	C	Convex SBI
22	2022-02-23	13:00 - 15:00	8	125	1, 2, 4, 5, 6, 7	C	-
23[c,d]	2022-02-23	21:30 - 03:00	2	300	1, 4	C	S-shaped SBI
24[d]	2022-02-25	09:50 - 12:40	4	175	1, 2, 4, 6, 7	C	Convex SBI

[a]The SBI was not observed on all profiles due to either a SBI erosion or SBI formation during in between profiles.

[b]Instrument issue during the flight

[c]Flights with an instrumental payload for aerosol filter sampling for chemical analysis (details not discussed here, see Pohorsky et al., 2024 for details)

[d]Flights used for the analysis of the mixing layer height and temperature inversion profile (Table 2).

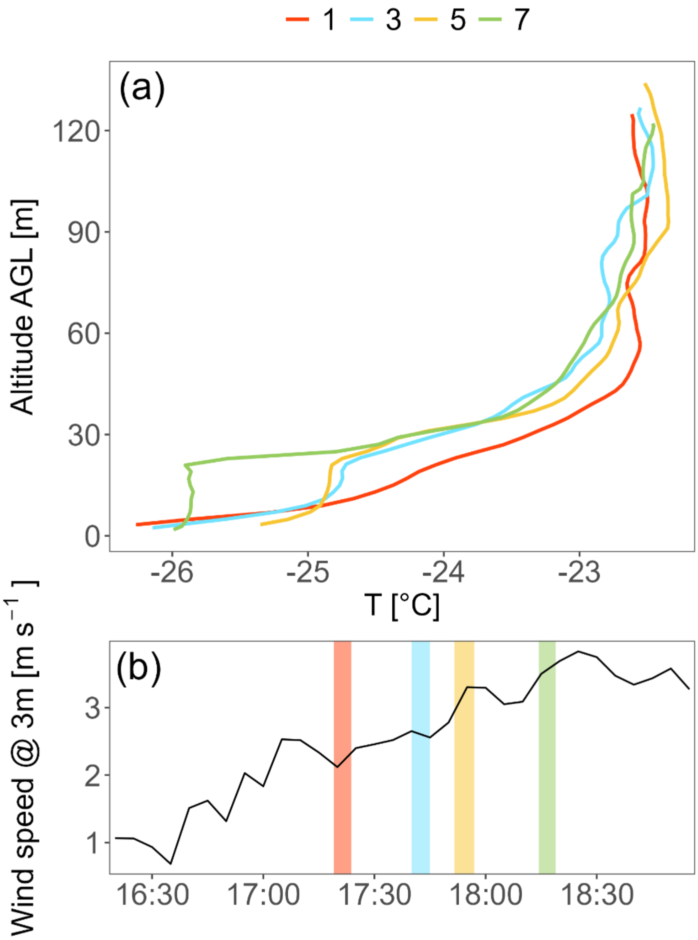

Figure S2 (a) Temperature profiles on February 10, 2022 (Flight 15) and (b) wind speed at 3 m. The colour of the profiles and of the shaded rectangles represent the profile numbers of the flight.

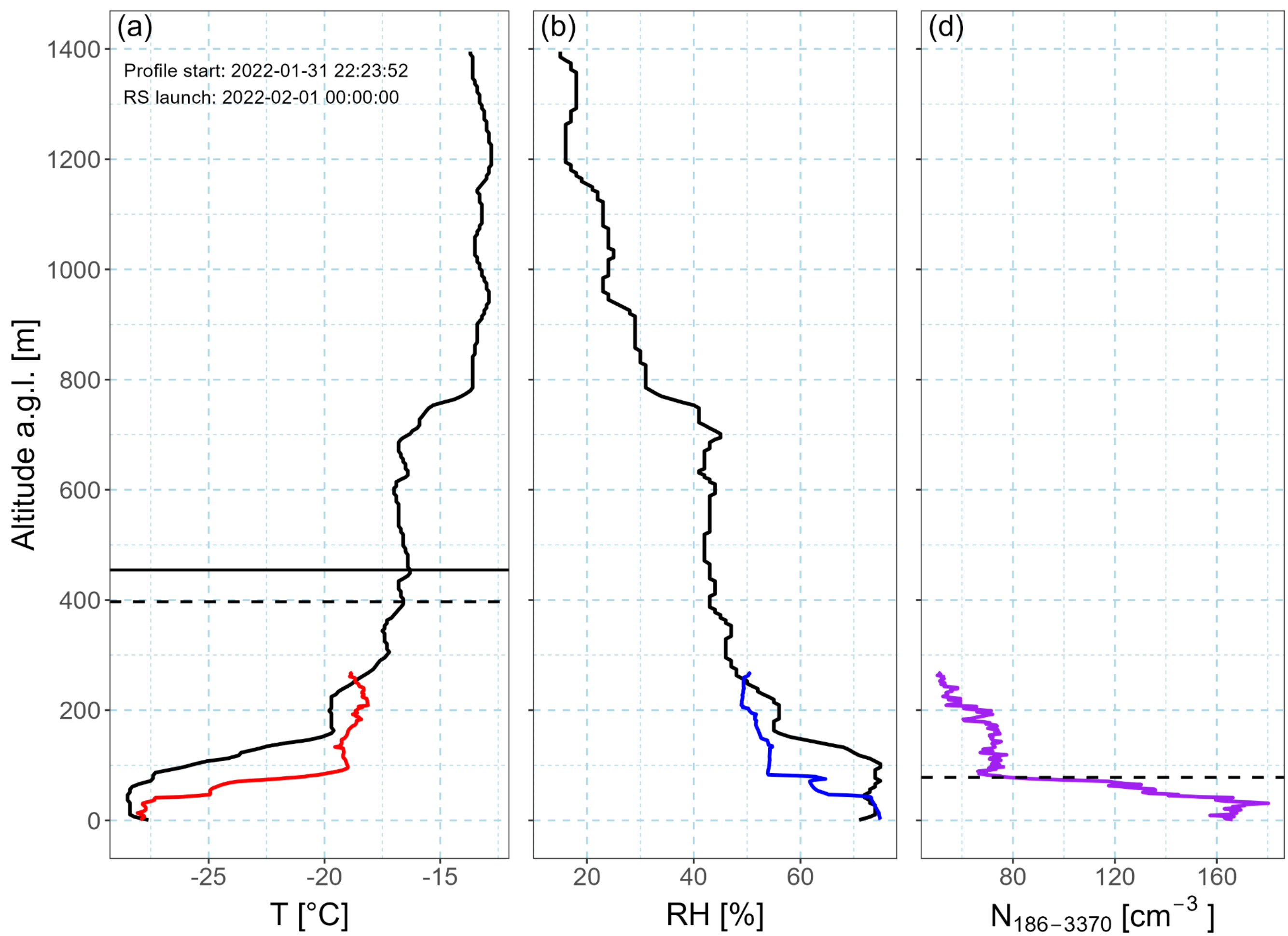

Figure S3 (a) Temperature profiles measured by a radiosonde (RS) at the Fairbanks airport (PAFA) on February 1st at 00:00 LT (black profile) and by the Helikite on January 31st at 22:24 LT (red profile). The horizontal full (dashed) line indicates the top of the SBI identified with a 0 °C / 100 m (0.65 °C / 100 m) threshold. (b) Relative humidity profiles from the radiosonde (black) and Helikite (blue). (c) $N_{186-3370}$ profile. The horizontal dashed line indicates the top of the mixing layer (h_{mix}).

Table S2 Ranges of surface flux parameters used for the calculation of the stable boundary layer height in Sect. 4.2

	25th	50th	75th
u_* [m s^{-1}]	0.031	0.078	0.163
L	0.26	3.8	17
H [W m^{-2}]	-5.7	-0.9	-0.1
$\|B_s\|$	-2.26 x 10^{-4}	-4.51 x 10^{-5}	-1.9 x 10^{-5}
N [Hz]	0.039	0.042	0.045

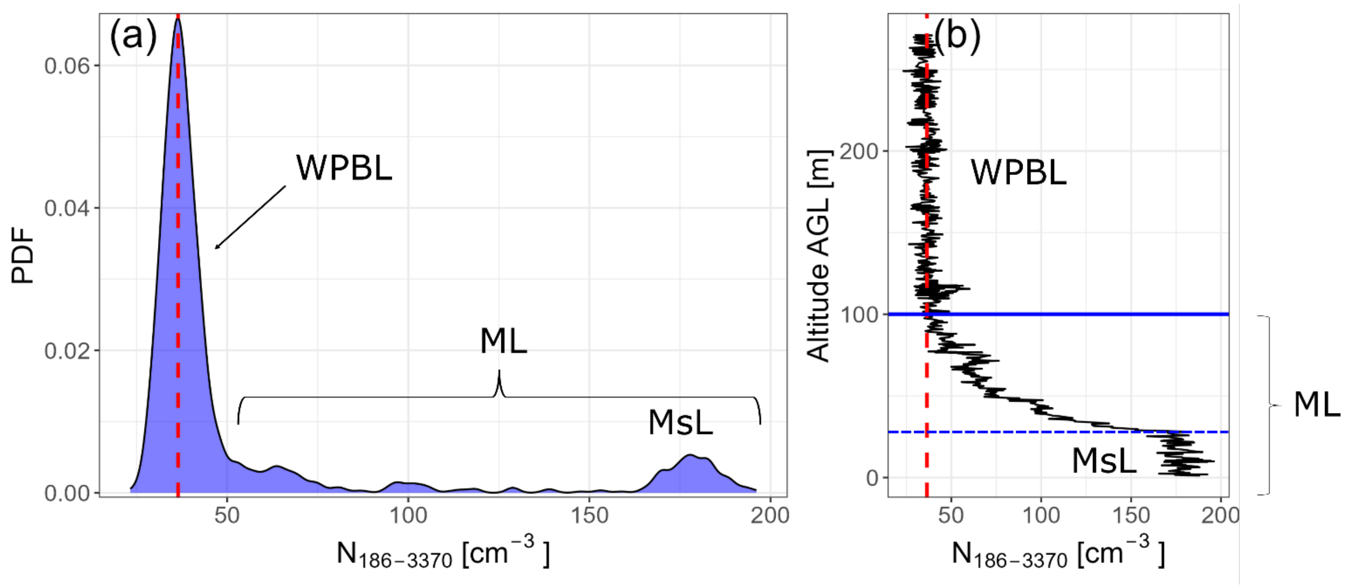

Figure S4 (a) Probability density function of the particle number concentration from 186 to 3370 nm ($N_{186\text{-}3370}$) in a vertical profile on January 27. The mode on the left represents the WPBL. The vertical red dashed line is the average of the WPBL distribution used as a background value for the concentration differences calculated in Sect. 5. (b) Vertical profile of $N_{186\text{-}3370}$. The red dashed line is the WPBL average concentration. The horizontal blue dashed and full lines represent the height of the MsL and ML respectively.

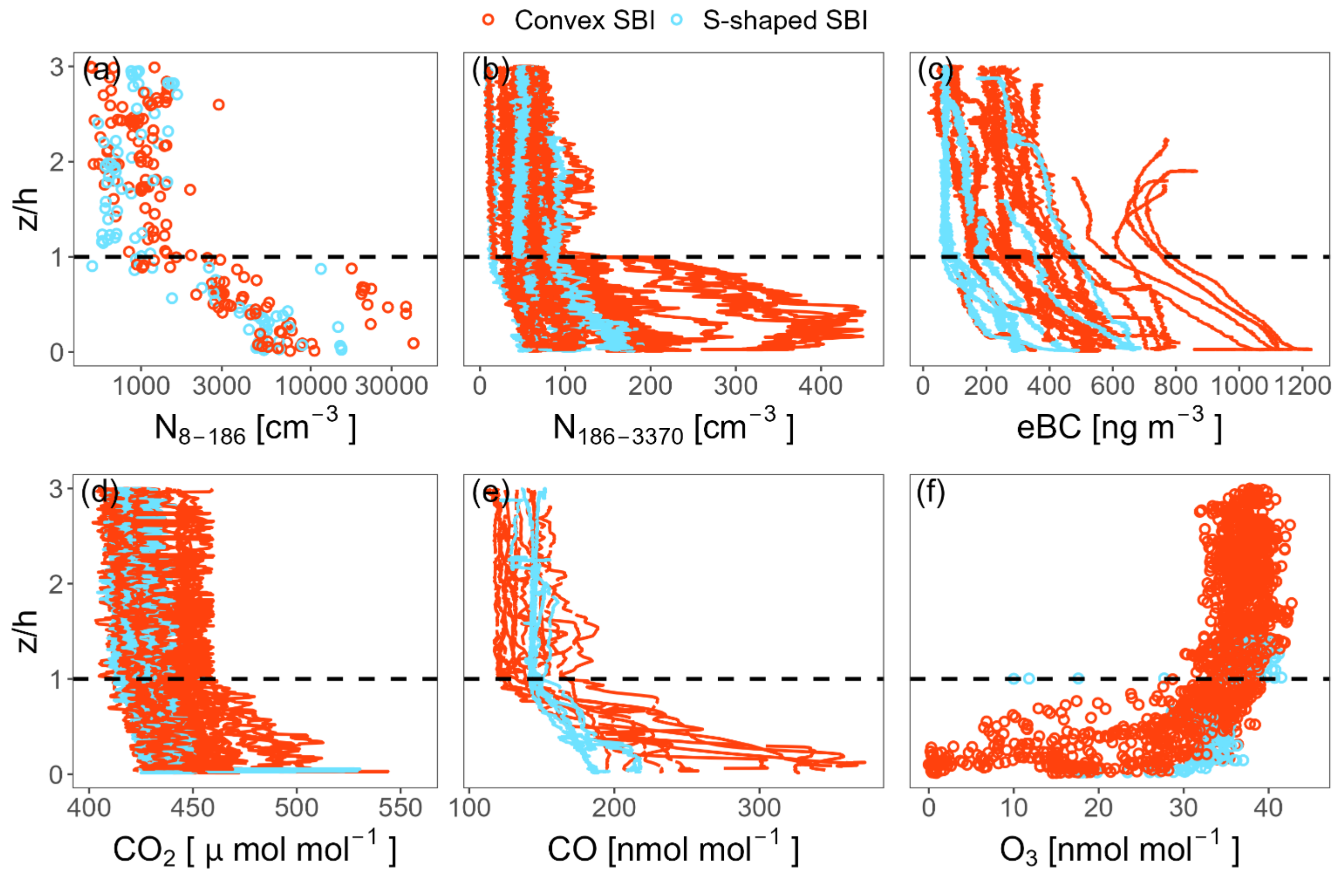

Figure S5 Vertically normalized profiles. (a) $N_{8\text{-}186}$, (b) $N_{186\text{-}3370}$, (c) eBC (d) CO_2 mixing ratio, (e) CO mixing ratio and (f) O_3 mixing ratio. The altitude (z) is normalized by the observed stable boundary layer height (h_{mix}). The profiles correspond to those in Fig. 11 but in absolute values. Profiles are color-coded based on the SBI type.

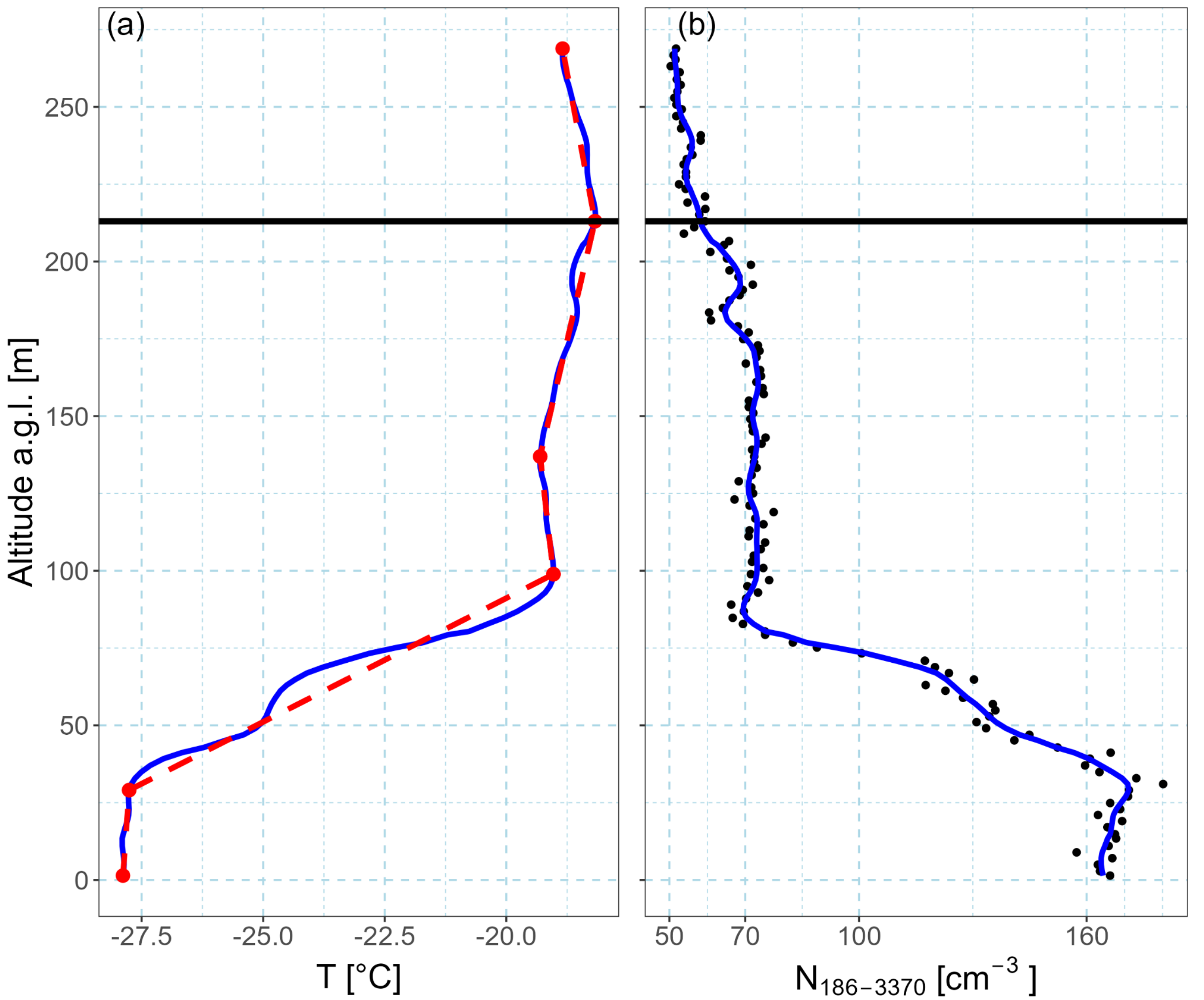

Figure S6 (a) Temperature profile on January 31 at 22:20 LT. The blue line represents the measured temperature with a gaussian smoothing over 20 meters. The red line represents the temperature profile from the temperature layering analysis (c.f. Sect. 2.3). (b) $N_{186\text{-}3370}$ profile. The black dots represent the 2-m averaged profile. The blue line represents the gaussian smoothed profile. The horizontal black line marks the top of an EI and the lower limit of the FTBL.

Comparison of pollution levels in the LBL and WPBL to reported Arctic haze background values

Table S3 indicates median (and interquartile range) values of the various measured tracers during ALPACA in the different layers and situations discussed in Sect. 4 and 5. The last column of Table S3 shows Arctic haze background values for submicron particle number concentrations from the literature. Background values represent either free tropospheric haze values or surface high latitude haze values for same period of the year (January – February) if available.

In April 2008, during the Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) project an aircraft measured the free tropospheric background haze concentrations above north Alaska (Brock et al., 2011). Six flights were carried out from April 11 to April 21, 2008 from Fairbanks. The aircraft was equipped with various aerosol and trace gas instruments. The flight region covered the northern part of Alaska and sea ice to the north. Flights included profiles up to 7400 m in altitude and down to 70 m. Four different air masses were intercepted during those flights, and classified according to the gas-phase composition of the air. Out of those four air masses, a free tropospheric haze background layer was identified and characterized by sulfat-rich aerosol extending from the top a surface-based inversion over sea ice to an altitude of 7400 m. Here, we used reported values of aerosol number concentration, CO and O_3 mixing ratios from Brock et al. (2011). Additional comparison of eBC values was made with data from Schmale et al. (2022) who analyzed seasonal cycles and trends of aerosol properties at 10 Pan-Arctic stations and Boyer et al. (2023) who compared Arctic station's aerosol measurements to aerosol data collected during the MOSAIC expedition. We used their data from Utqiagvik/Barrow for the months of January and February from 1992 to 2019. We also compared our observation to vertical measurements from Mazzola et al. (2016) who performed tethered-balloon measurements over Ny-Ålesund in September 2015 and April-May 2015, collecting vertical profiles of eBC concentrations. We used their profiles from April to compare values at higher elevations.

Freud et al., (2017) analyzed the seasonality and transport patterns driving aerosol number size distribution from 20 to 500 nm across several Pan-Arctic stations from 2007 to 2015 (Alert, Villum Research Station – Station Nord, Zeppelin, Tiksi and Utqiagvik/Barrow). We used results at Utqiagvik/Barrow during January/February to compare with the LBL values from the Helikite flights. Although the measurements from Freud et al. (2017) were taken at the surface, they constitute a useful reference for Arctic haze values for the North American sector of the Arctic. We also compared our PNSD to their haze size distribution identified from a k-means clustering analysis from the Alert, Villum - Station Nord and Zeppelin stations. This comparison is made under the assumption that the average Arctic haze PNSD are homogenous throughout the Arctic. Finally, we used reported values from Engvall et al. (2008) who reported PNSD from 20 to 630 nm from the Zeppelin station for the month of April between 2000 and 2005.

Kinase et al., (2023) analyzed CO measurements from 2016 to 2020 at the Poker flat research range located 30 km north of Fairbanks. This dataset constitutes background boundary layer values for northern Alaska. Given its geographical proximity to Fairbanks, this data constitutes a good regional background reference as it is not directly influenced by fresh pollution emissions, yet close to Fairbanks. Finally, Whaley et al. (2023) reported the seasonality of CO and O_3 mixing ratios, including at Utqiagvik/Barrow and vertical profiles from observations and modelling studies. We used their observations for the months of January and February for comparison.

Table S3 Table of concentrations and mixing ratios measured in the different layers and under various situations by the Helikite during ALPACA. The first value always indicates the median and the values in brackets represent the 25th and 75th percentile, respectively. Background values in the last column refer to measurements of submicron particle number concentrations in Arctic haze at various elevations.

	MsL (Convex SBI)	MsL (s-shaped SBI)	No SBI flights	WPBL (easterly wind)	WPBL (other wind direction)	LBL	Literature reported background values
N_{8-186} [cm^{-3}]	6000 (4500 - 8490)	5430 (5160-6860)	1000 (800-3600)	1220 (820 - 1480)	670 (310 - 890)	174 (149-215)	371* (Brock et al., 2011) 200 – 250** (Freud et al., 2017)
$N_{186-3370}$ [cm^{-3}]	112 (85-182)	98 (88-110)	55 (53 - 57)	56 (47 - 65)	49 (40 - 58)	45 (42 - 50)	
eBC [ng m^{-3}]	550 (500 - 700)	290 (260 - 490)	126 (112-142)	230 (180 - 290)	80 (65 - 130)	56 (52-74)	60 (Brock et al., 2011) 58 [31 - 103] (Schmale et al., 2022) 100 – 300 (Mazzola et al., 2016)
CO_2 [µmol mol^{-1}]	443 (435 - 458)	436 (432 - 441)	426 (423-435)	423 (416 - 433)	420 (416 - 424)	420 (418 - 422)	-
CO [nmol mol^{-1}]	237 (200 - 255)	185 (182 - 209)	-	132 (121 - 140)	143 (135 - 148)	-	161 ± 8 (Brock et al., 2011) 131 (107 - 150) (Kinase et al., 2023) ~ 140 – 150 (Whaley et al., 2023)
O_3 [nmol mol^{-1}]	16 (6 - 26)	30 (27 - 31)	33 (31 - 34)	38 (36 - 39)	38 (37 - 39)	-	52 ± 14 (Brock et al., 2011) 32 - 35 (Whaley et al., 2023)

*Size range: < 1000 nm

**Size range: 20 – 500 nm

***Size range: 20 – 630 nm

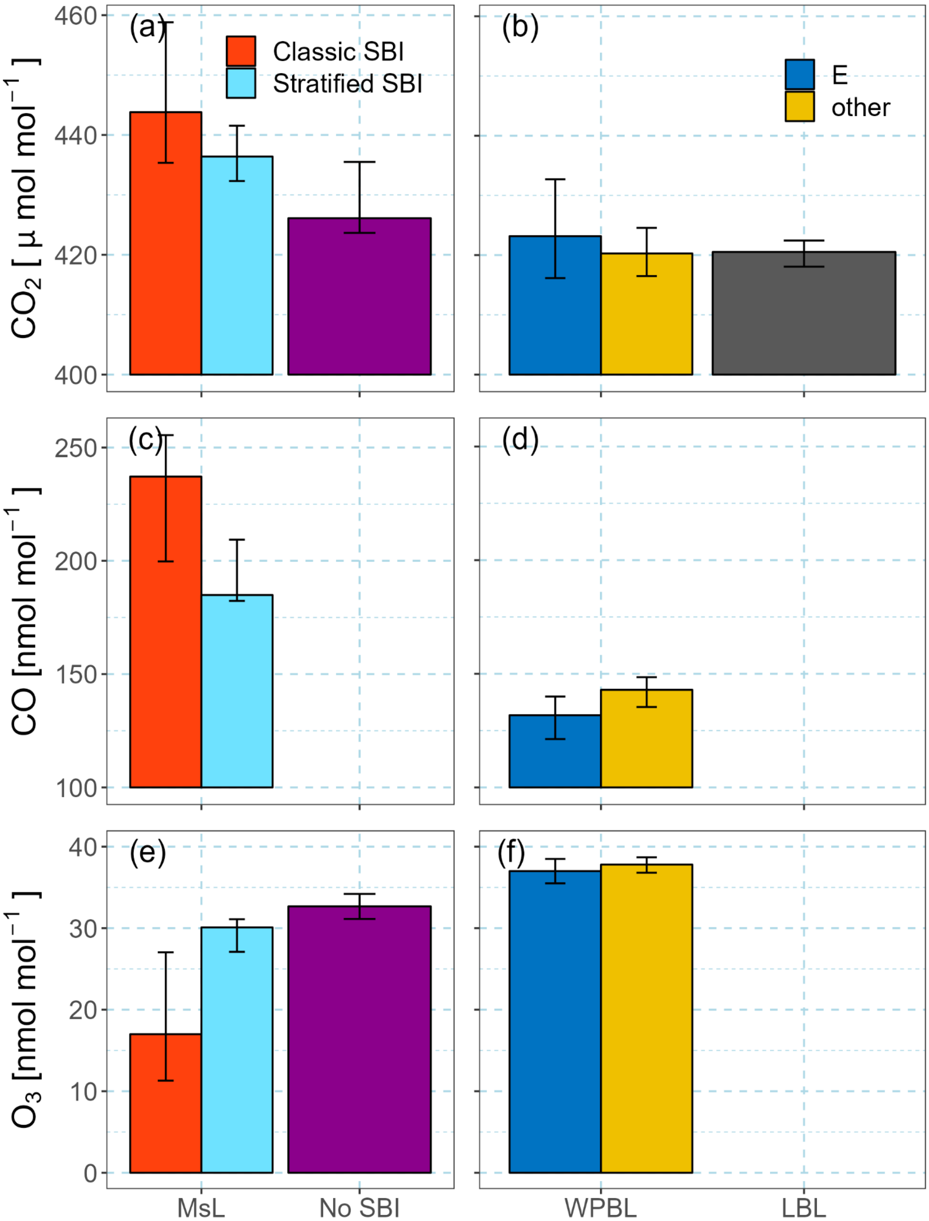

Figure S7 Median mixing ratios of CO_2 (a and b), CO (c and d) and O_3 (e and f). Left panels show values in the mixed sublayer (MsL) under conditions of convex SBI (red) and 's-shaped' SBI (blue) and without an SBI (purple). Right panels show values in the WPBL under different dominant wind directions (blue and yellow) and in the LBL (grey). The error bars indicate the interquartile range. If a bar is not shown, it means that no measurements of the specific tracer are available for the specific layer or situation.

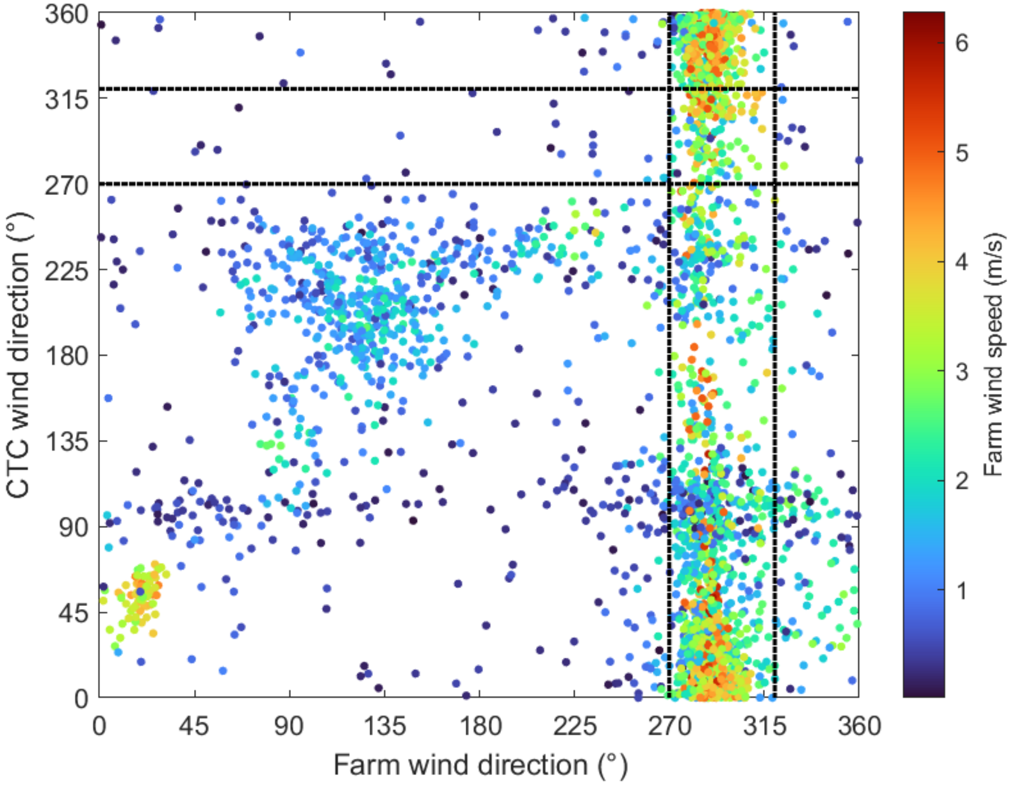

Figure S8 Wind speed and direction measured simultaneously at the UAF farm and CTC sites during the ALPACA campaign. The dashed lines indicate the direction range associated with the SCF. Dots that fall inside this range at both sites indicate that the SCF was also measured at CTC.

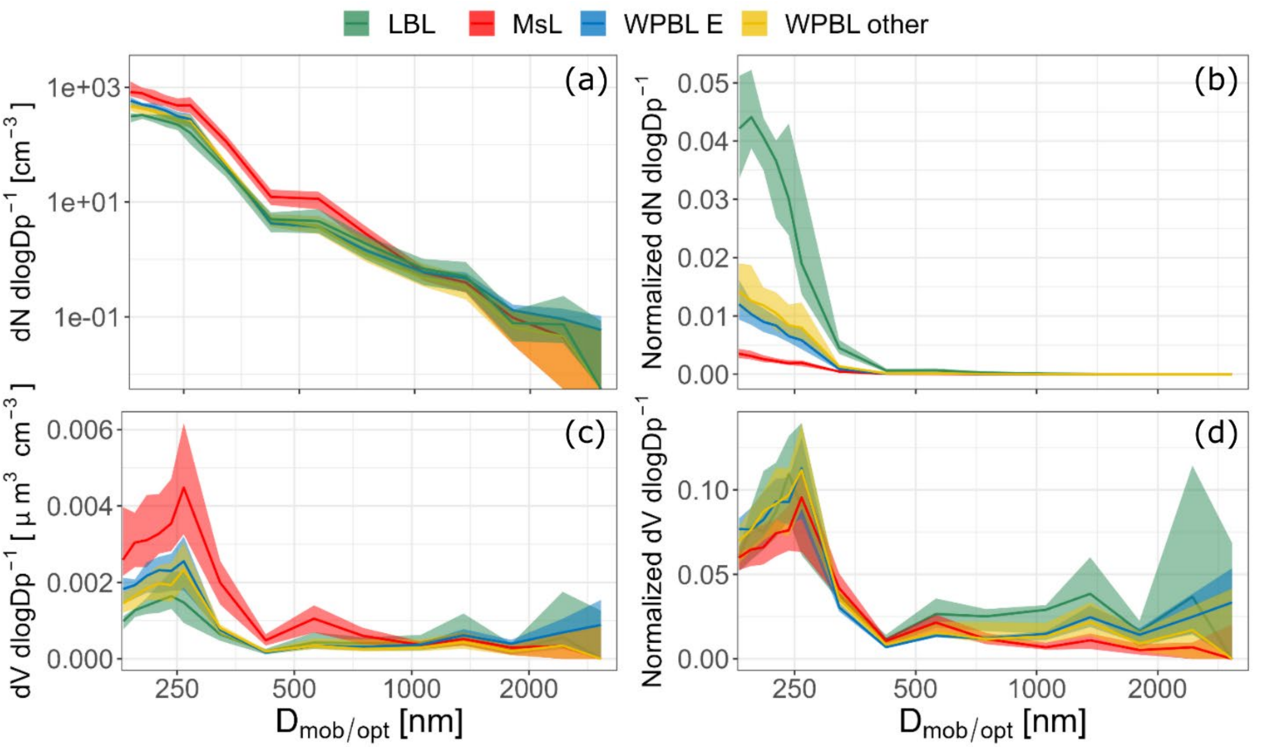

Figure S9 (a) Particle number size distribution in the mixed sub-layer (red), in the weakly polluted background layer under easterly dominant winds (blue) and other wind directions (yellow) and above the EIs in the lowest background layer (green). (b) Normalized PNSD in the same layers. (c) PVSD in the same layers. (d) Normalized PVSD in the same layers. The displayed size range is from 180 to 3370 nm and is merged from the mSEMS and the POPS.

Table S4 Particle number size distribution fit parameters from Fig. 13. The μ is the mode diameter and σ is the standard deviation of each respective mode.

Layer	μ [nm]	σ [nm]	mode
MsL	28.8 ± 1.8	22.1	Aitken
	181 ± 1.4	86.2	Accumulation
WPBL (Easterly wind)	26 ± 1.7	34.1	Aitken
	182 ± 1.4	87.2	Accumulation
WPBL (other)	32.6 ± 2.1	35.5	Aitken
	187 ± 6.0	100.5	Accumulation
LBL	32.7 ± 2.1	124	Aitken
	193.1 ± 6.7	90.8	Accumulation

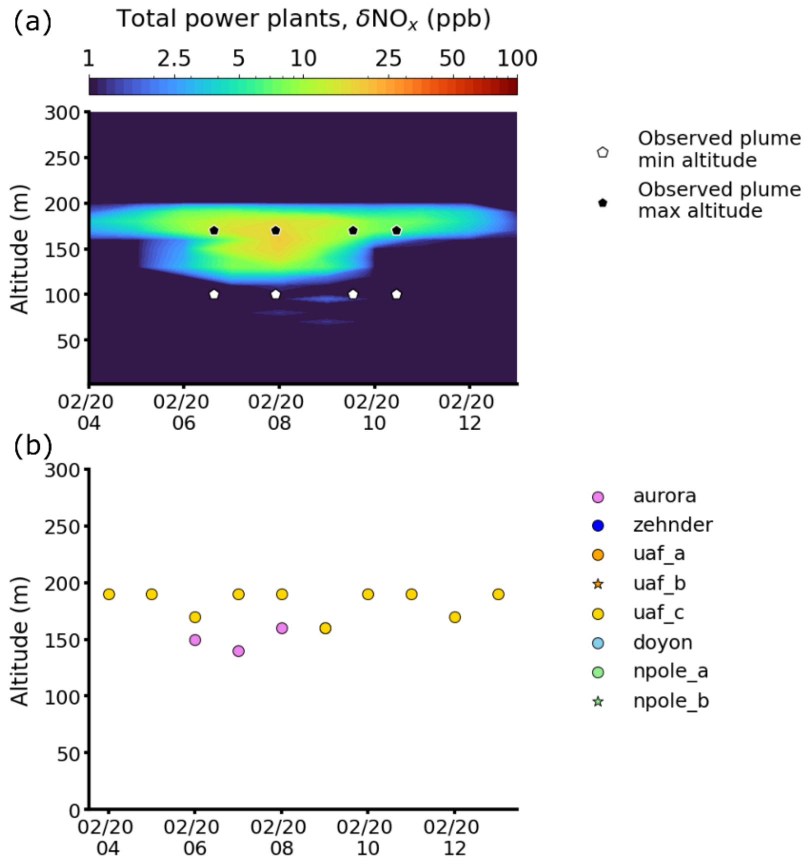

Figure S10: Figure S6: (a) Timeseries of the vertical distribution of simulated NO_x tracer enhancements above background (ppb) from the different power plants in Fairbanks for the 1.33 x 1.33 km grid box covering the UAF site from 0400 to 1400 AKST on 20 February 2022. (b) Altitudes of the different power plant plumes simulated over the UAF site for the same period. Results are from FLEXPART-WRF pollution dispersion model simulations for the ALPACA-2022 campaign. See Brett et al. (2024) for details.

Table S5 List of power plants in Fairbanks with the respective fuel type and stack height

Power plant	Fuel type	Stack height [m]
UAF A	Diesel	20
UAF B	Diesel	20
UAF C	Coal	64
Aurora	Coal	48
Zehnder	Diesel	18
Doyon	Coal	26

Plume identification details:

In Table 6 (Sect. 6.2) a question mark indicates that the source is unknown or uncertain. For two plumes, the recorded flight data was insufficient to perform an analysis of tracer ratios (rows in grey shading). These plumes were therefore not used in the analysis. On February 9, two plumes (plume ID 141 and 142) with distinct tracer ratios were observed at very similar heights and were partially overlapping. After careful comparison with FLEXPART-WRF model tracer results (see Brett et al., 2024), plume 142 was attributed to UAF C as it showed similar ratios to those observed in other UAF C plumes. For plume 141, it is uncertain whether the observed peak belongs to plume 141 despite the vertical displacement or if the origin is different. According to the FLEXPART-WRF results, Zehnder could also be a potential source in this case. Given the slight overlap of these two plumes, their tracer ratios might therefore be different as a result of mixing of the different species. On February 23, a plume (ID 231) was observed at 50 m above ground with wind directions of 290°. Given the wind direction, a likely source could be located on Chena ridge (Fig. 1).