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Diurnal evolution of negative atmospheric ions above the boreal forest: from ground level to the free troposphere

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Table S1 Overview of time and height levels during the vertical profiling above SMEAR II station**.** All times are given in East European Time (UTC + 2). The height levels are given in m above ground level. For simplification, the height levels are rounded to the nearest hundred. The times shown in the table correspond to the measurement above SMEAR II station and do not include the route from the Airport to SMEAR II.

Date	**Profiles (height & time)**				
02 May 2017	100 m 04:15 – 04:26	700 m 04:32 – 04:42	3200 m 05:08 – 05:19	700 m 05:40 – 05:50	100 m 05:57 – 06:05
02 May 2017	100 m 09:04 – 09:14	300 m 09:15 – 09:28	700 m 09:31 – 09:44	500 m 09:46 – 10:00	300 m 10:03 – 10:15
02 May 2017	100 m 13:49 – 14:06	700 m 14:09 – 14:23	2700 m 14:37 – 14:53	1500 m 15:00 – 15:10	400 m 15:15 – 15:31
03 May 2017	100 m 04:14 – 04:26	700 m 04:30 – 04:42	2700 m 04:56 – 05:10	1500 m 05:20 – 05:31	300 m 05:38 – 05:49
03 May 2017	100 m 08:57 – 09:08	600 m 09:11 – 09:22	2700 m 09:36 – 09:47	1600 m 09:54 – 10:05	100 m 10:29 – 10:35
04 May 2017	100 m 04:10 – 04:26	700 m 04:31 – 04:44	2700 m 04:58 – 05:16	600 m 05:28 – 05:39	100 m 05:43 – 05:48
04 May 2017	100 m 08:46 – 08:57	700 m 09:02 – 09:13	2800 m 09:30 – 09:43	700 m 10:00 – 10:10	100 m 10:15 – 10:24
04 May 2017	100 m 13:42 – 14:02	700 m 14:06 – 14:18	2700 m 14:32 – 14:44	700 m 15:00 – 15:10	100 m 15:15 – 15:28
05 May 2017	100 m 04:15 – 04:30	700 m 04:34 – 04:46	2700 m 05:02 – 05:15	700 m 05:32 – 05:42	100 m 05:48 – 06:01
05 May 2017	100 m 09:21 – 09:38	700 m 09:42 – 09:51	2700 m 10:08 – 10:19	700 m 10:38 – 10:50	100 m 10:55 – 11:09
05 May 2017	100 m 14:31 – 14:44	700 m 14:49 – 15:00	2800 m 15:17 – 15:27	700 m 15:42 – 15:51	100 m 15:56 – 16:17
12 May 2017	100 m 03:44 – 03:58	700 m 04:03 – 04:16	2500 m 04:30 – 04:43	700 m 04:57 – 05:07	100 m 05:11 – 05:22
12 May 2017	100 m 08:12 – 08:25	700 m 08:30 – 08:41	2600 m 08:56 – 09:06	700 m 09:18 – 09:27	100 m 09:31 – 09:48
16 May 2017	100 m 03:35 – 03:52	700 m 03:57 – 04:10	2600 m 04:26 – 04:40	1500 m 04:48 – 05:00	100 m 05:10 – 05:21
16 May 2017	100 m 08:34 – 08:45	700 m 08:49 – 09:03	500 m 09:08 – 09:27	100 m 09:29 – 09:42	500 m 09:45 – 09:59
16 May 2017	100 m 13:16 – 13:32	700 m 13:35 – 13:49	1500 m 13:56 – 14:05	700 m 14:09 – 14:20	400 m 14:22 – 14:37

Table S2 Times of sunrise and sunset at SMEAR II station during the six measurement flights: All times are given in East European Time (UTC + 2). The SMEAR II station is located at 61° 51' N, 24° 17' E. Source: NOAA (https://gml.noaa.gov/grad/solcalc/sunrise.html)

Date	Sunrise	Sunset
02 May 2017	04:07	20:34
03 May 2017	04:04	20:37
04 May 2017	04:01	20:40
05 May 2017	03:59	20:43
12 May 2017	03:29	21:02
16 May 2017	03:28	21:13

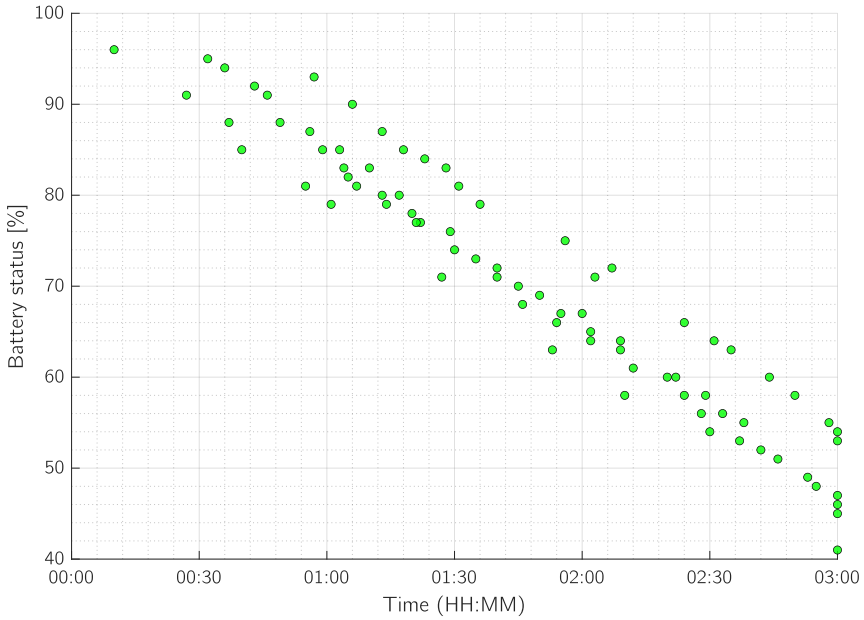

Figure S1 Lifetime of the battery of the autonomous APi-TOF setup. The figure is showing the manually recorded battery status (in %) on six different flights: three flights from 4 May and three from 5 May 2017. The x-axis shows the time passed from unplugging until recharghing (~3 hours).

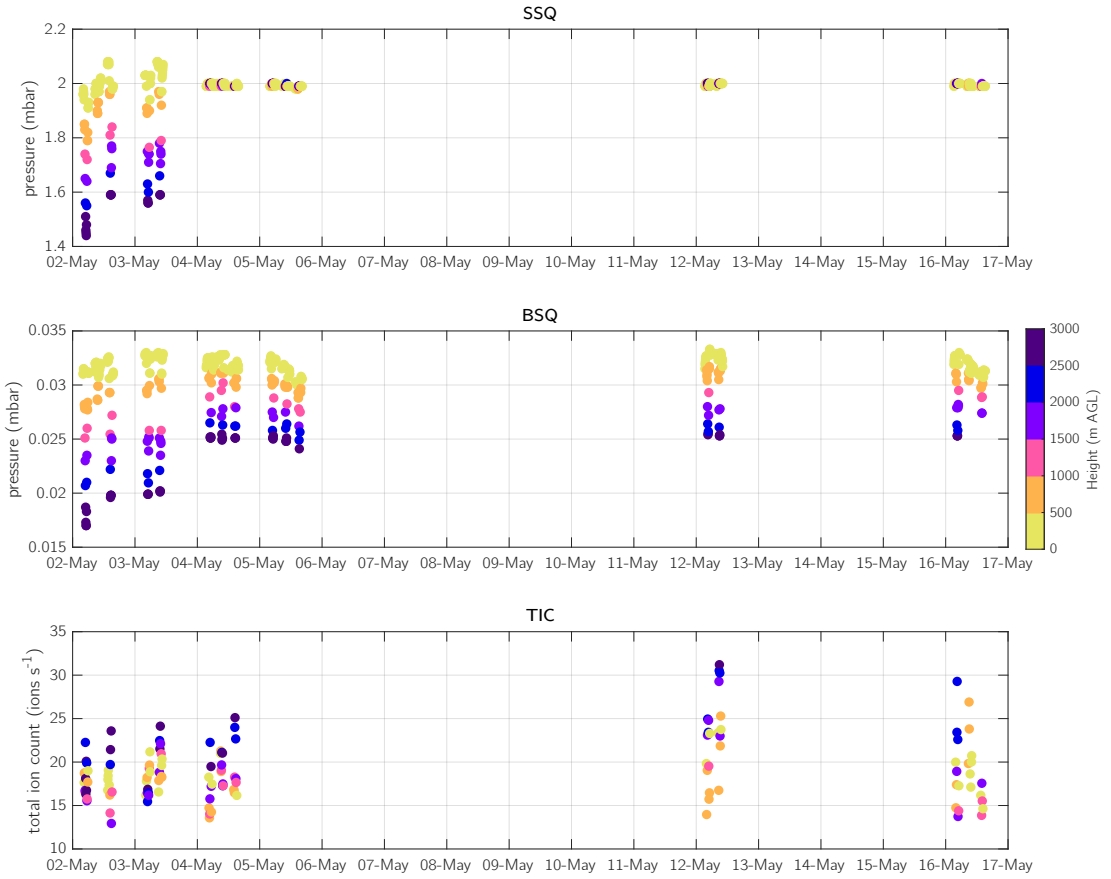

Figure S2 Time series of pressures inside the APi-TOF during the measurement campaign. (a) Pressure of the Small Segmented Quadrupole (SSQ) chamber inside the APi-TOF as well as **(b)** the Big Segmented Quadrupole (BSQ) during all measurement flights. The pressure of the SSQ was kept stable at 2 mbar on 4 out of 6 measurement days (4, 5, 12 and 16 May 2017). The BSQ pressure dropped with the ambient pressure on each flight but did not reach values below 0.025 mbar during flights when the SSQ was at 2 mbar. The depicted pressures are coloured by height in m AGL. Panel (c) depicts the total ion count (ions s^{-1}), also coloured by height.

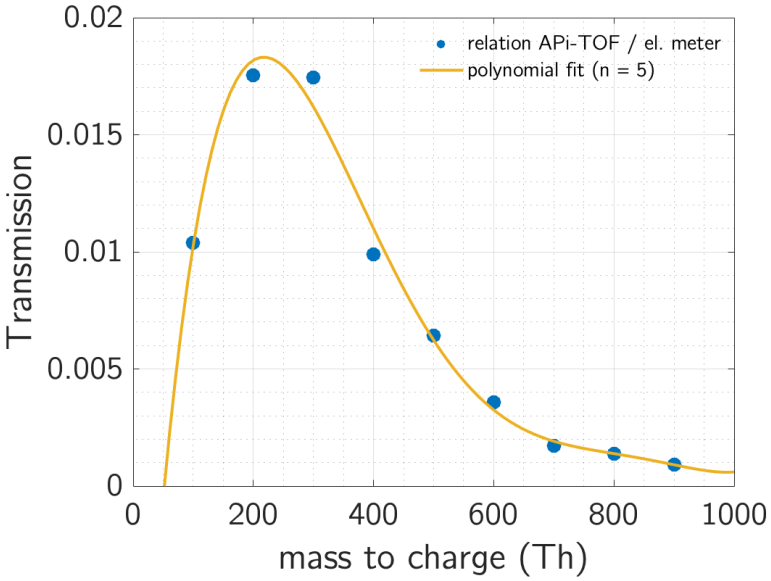

Figure S3 Transmission curve of the flying APi-TOF. The transmission was measured with a high resolution differential mobility analyser (HDMA) setup as described in Junninen et al. (2010), introducing charged particles from a nickel-chrome wire into the air sample. The sample with the charged particles was split in two: one towards the HDMA for selecting the size range and a electrometer to count the particles; the second sample flow was lead to the APi-TOF. The transmission of ions around 100 Th is 1 %, around 200 to 300 Th ~1.8 %, around 400 Th 1 % and for ions bigger than 500 Th, the transmission is below 0.5%.

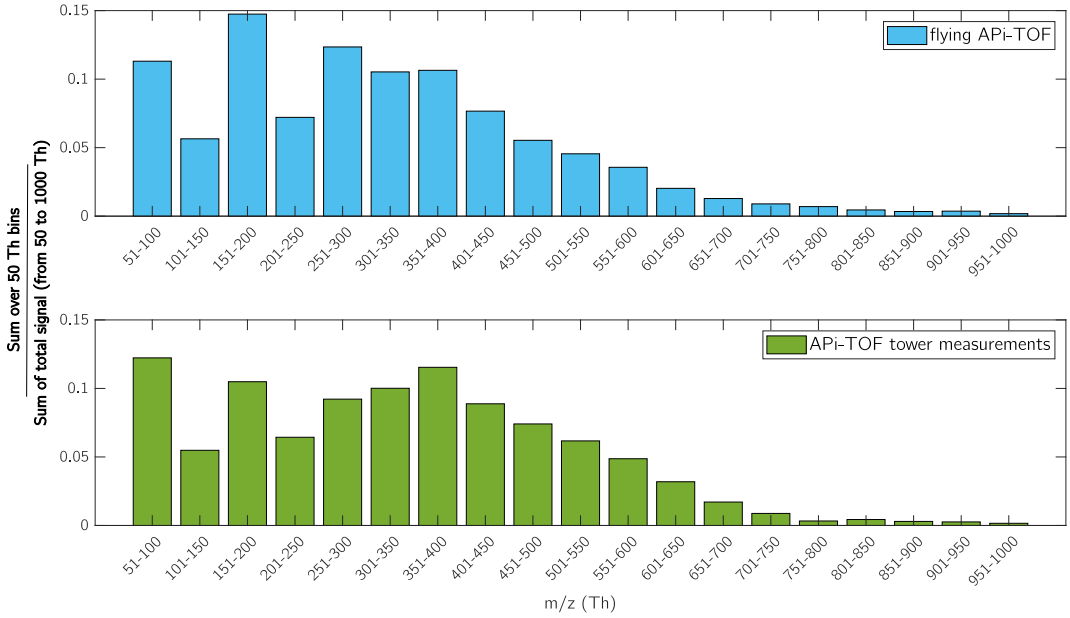

Figure S4 Comparison between both APi-TOF measuring ambient ions on 24 May 2017, 8 – 18 EET. The APi-TOF in the top panel was the instrument used for airborne measurements. The APi-TOF in the lower panel was stationary located at the tower (where the comparison of both instruments was also conducted) at the SMEAR II station.

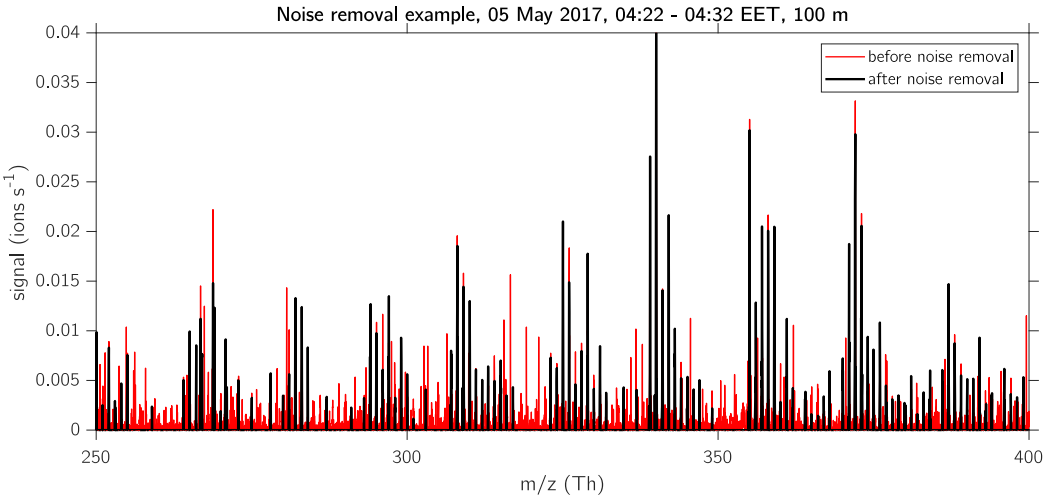

Figure S5 Measurements from APi-TOF on 5 May 2017, 04:22 – 04:32 EET at 100 m height. The red line shows the raw data averaged over 10 min, the black line is the result of averaging after implementing the noise removal function.

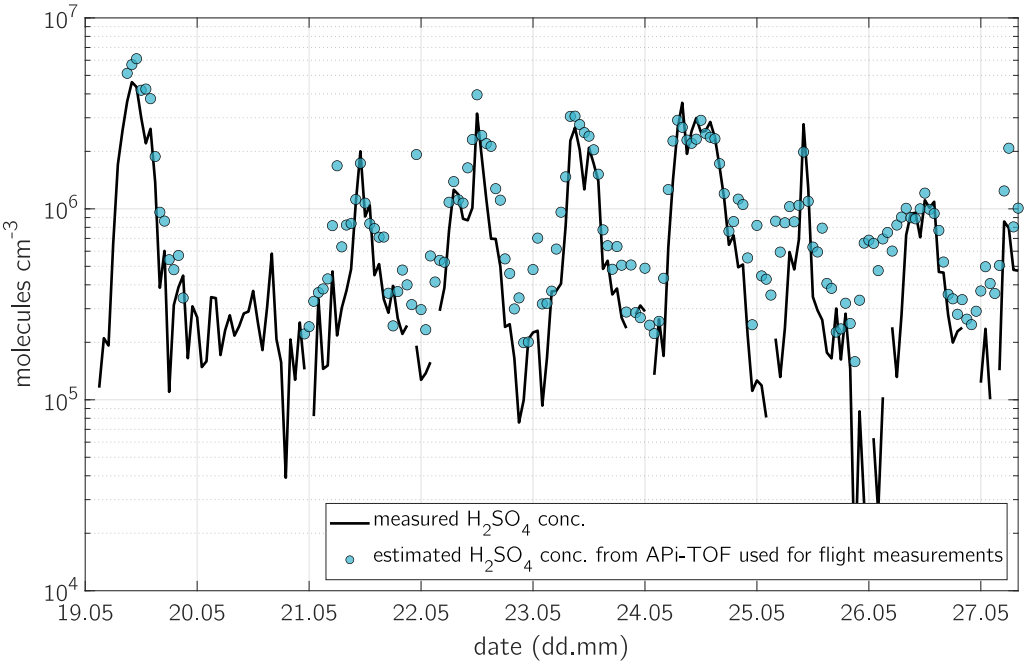

Figure S6 Measured and estimated sulfuric acid concentration at SMEAR II station from 19 May to 27 May 2017. The figure has been modified from Beck et al., 2021b.

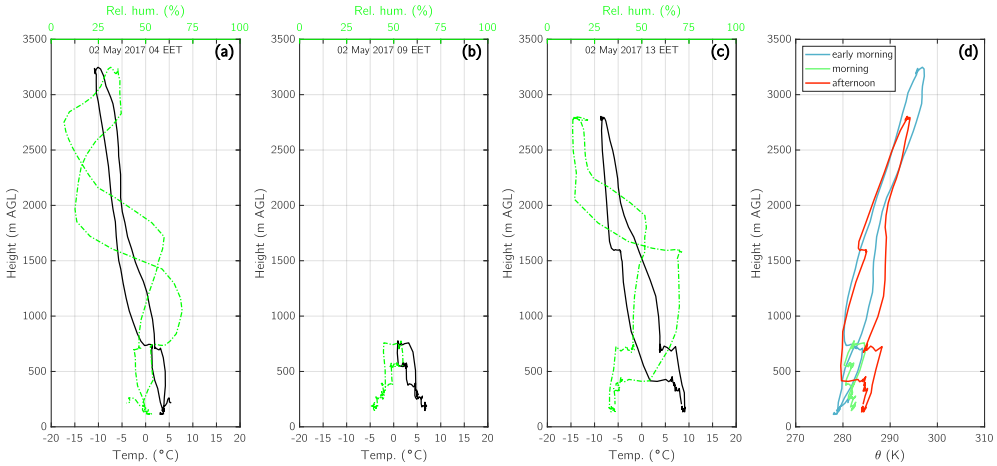

Figure S7 Temperature and relative humidity profile on 2 May 2017: Temperature (black, solid line) and relative humidity (green, dashed line) of the measurements conducted during early morning (a), morning (b) and afternoon (c). The exact times of measurements can be determined from Table S1. (d) Potential temperature (θ) profile of all flights in Kelvin.

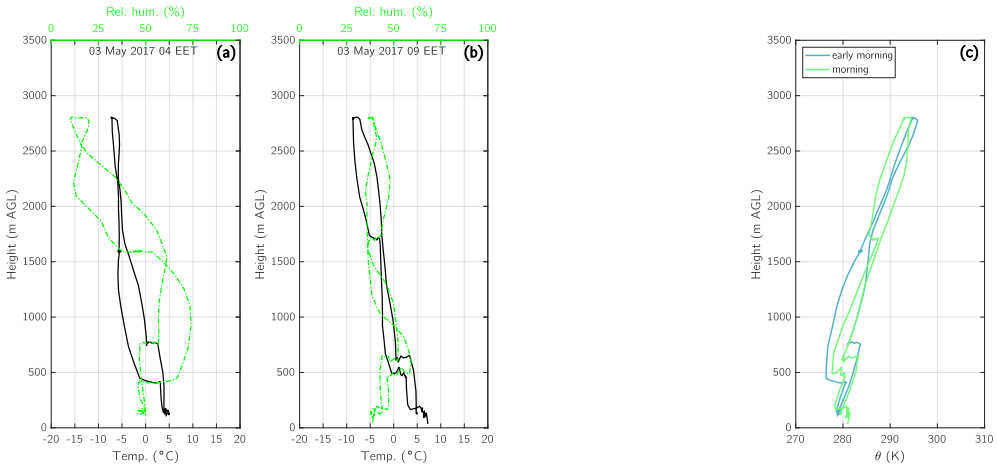

Figure S8 Temperature and relative humidity profile on 3 May 2017: Temperature (black, solid line) and relative humidity (green, dashed line) of the measurements conducted during early morning (a) and morning (b). The exact times of measurements can be determined from Table S1. (c) Potential temperature (θ) profile of all flights in Kelvin.

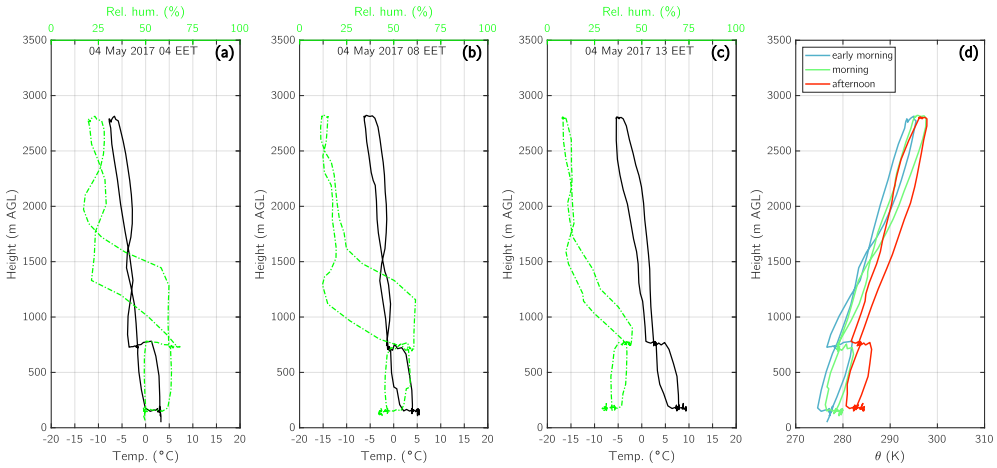

Figure S9 Temperature and relative humidity profile on 4 May 2017: Temperature (black, solid line) and relative humidity (green, dashed line) of the measurements conducted during early morning (a), morning (b) and afternoon (c). The exact times of measurements can be determined from Table S1. (d) Potential temperature (θ) profile of all flights in Kelvin.

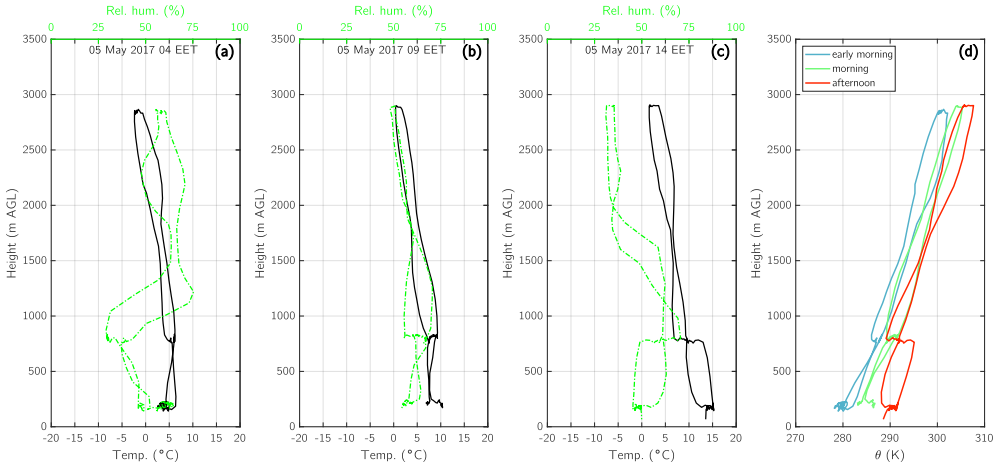

Figure S10 Temperature and relative humidity profile on 5 May 2017: Temperature (black, solid line) and relative humidity (green, dashed line) of the measurements conducted during early morning (a), morning (b) and afternoon (c). The exact times of measurements can be determined from Table S1. (d) Potential temperature (θ) profile of all flights in Kelvin.

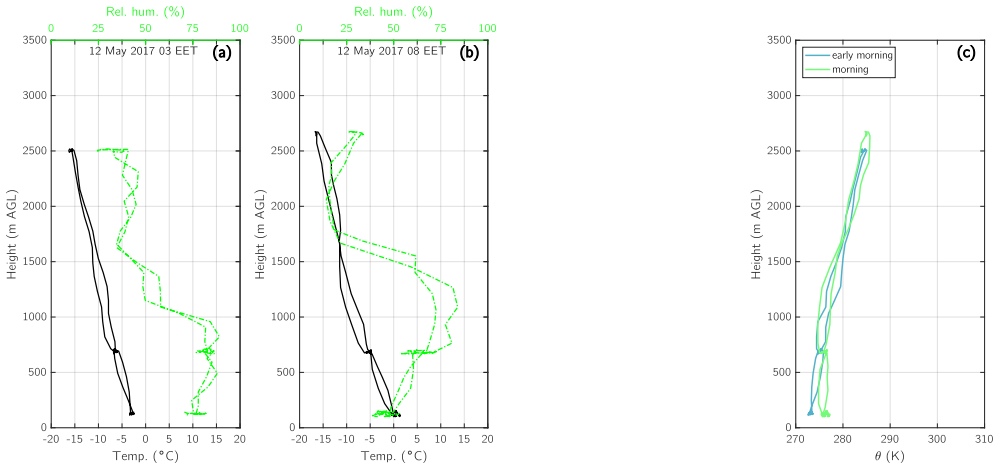

Figure S11 Temperature and relative humidity profile on 12 May 2017: Temperature (black, solid line) and relative humidity (green, dashed line) of the measurements conducted during early morning (a) and morning (b). The exact times of measurements can be determined from Table S1. (c) Potential temperature (θ) profile of all flights in Kelvin.

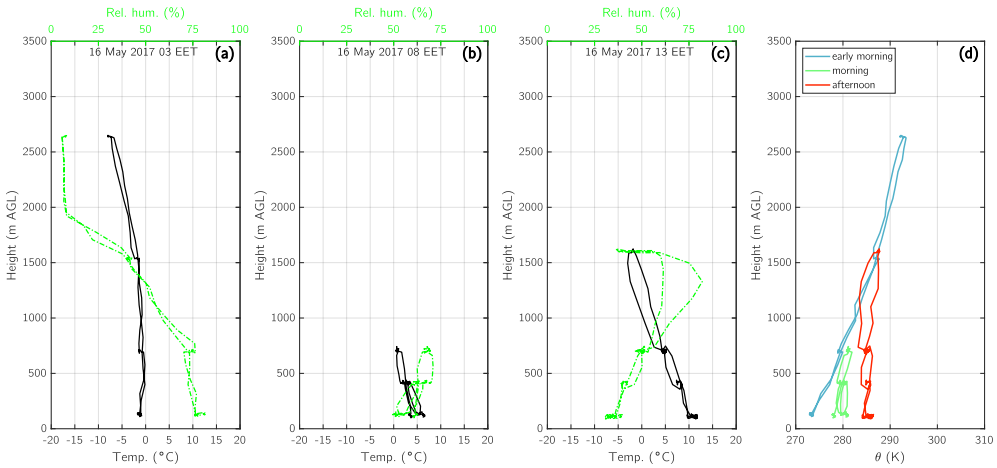

Figure S12 Temperature and relative humidity profile on 16 May 2017: Temperature (black, solid line) and relative humidity (green, dashed line) of the measurements conducted during early morning (a), morning (b) and afternoon (c). The exact times of measurements can be determined from Table S1. (d) Potential temperature (θ) profile of all flights in Kelvin.

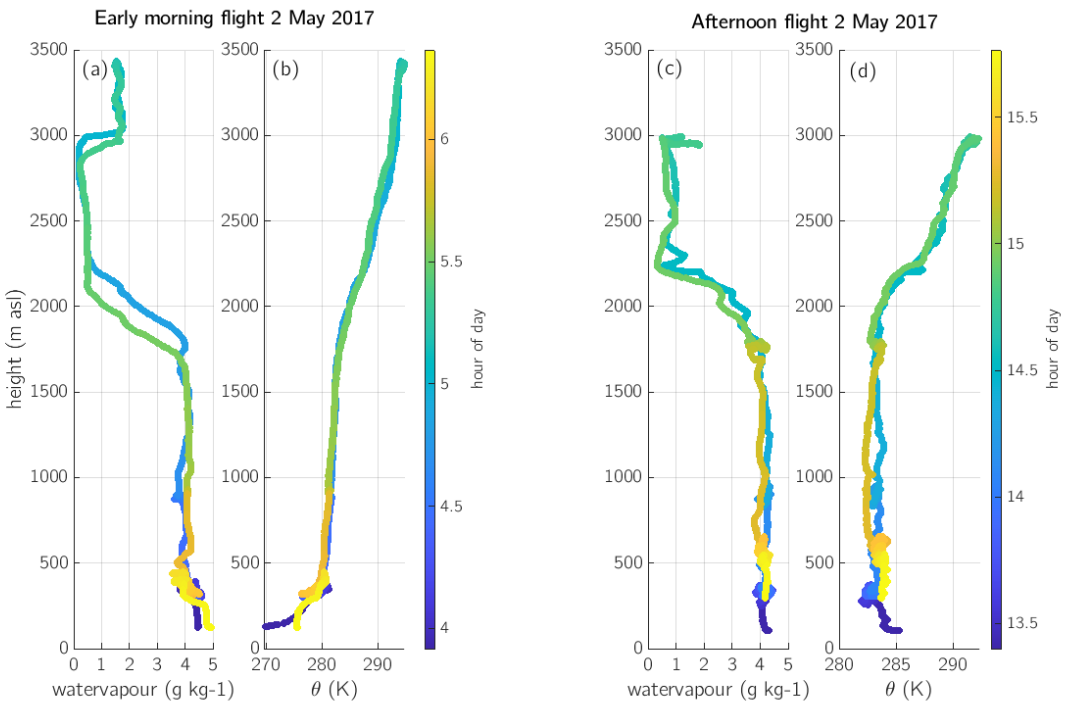

Figure S13 Vertical profile of water vapour and potential temperature on 2 May 2017. (a) Water vapour profile and (b) potential temperature during early morning flight. The colour indicates the hour of the day. (c) Water vapour and (d) potential temperature during the afternoon flight, with coloured hour of the day.

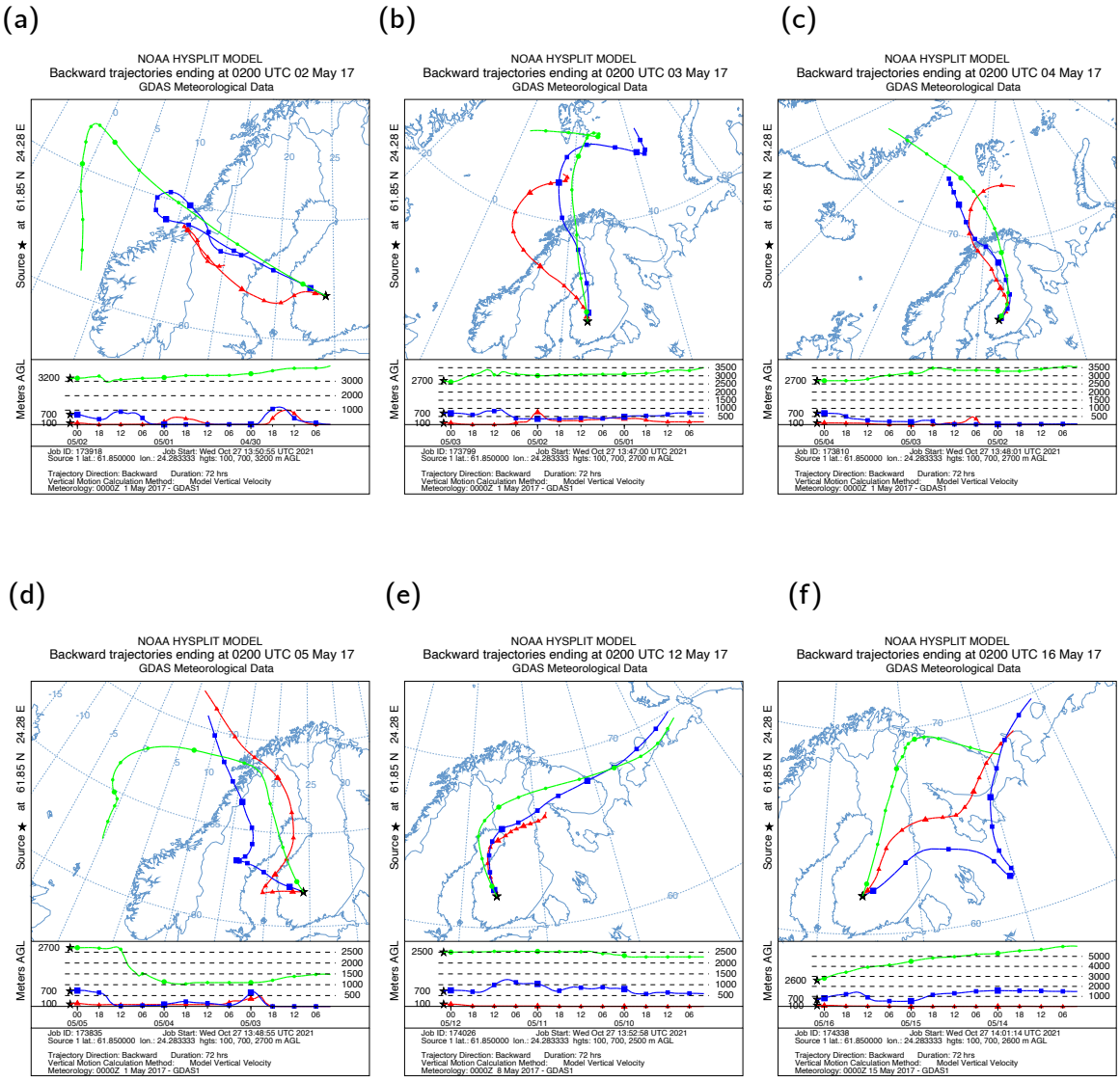

Figure S14 72 hours back trajectories from SMEAR II station calculated with HYSPLIT (NOAA, Stein et al. (2015); Rolph et al. (2017)) for 2 UTC (4 EET) for 2 May (a), 3 May (b), 4 May (c), 5 May (d), 12 May (e) and 16 May 2017 (f). Shown are three different heights, 100 m, 700 m and according to the measurements in the free troposphere (Table S1) on 3200 m (a), 2700 m (b, c, d), 2500 m (e) and 2600 m (f) above ground level.

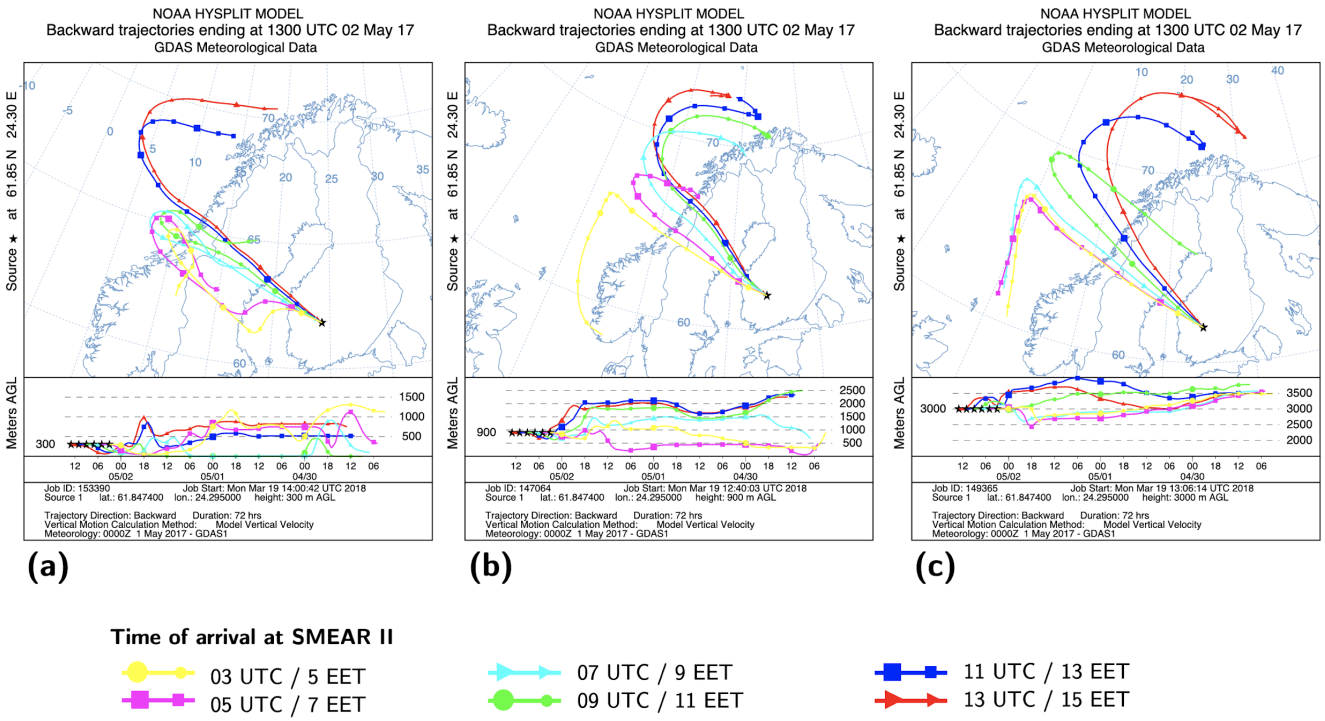

Figure S15 72 hours back trajectories from SMEAR II station calculated with HYSPLIT (NOAA, Stein et al. (2015); Rolph et al. (2017)). (a) for 300 m height ASL, (b) 900 m ASL and (c) 3000 m ASL.

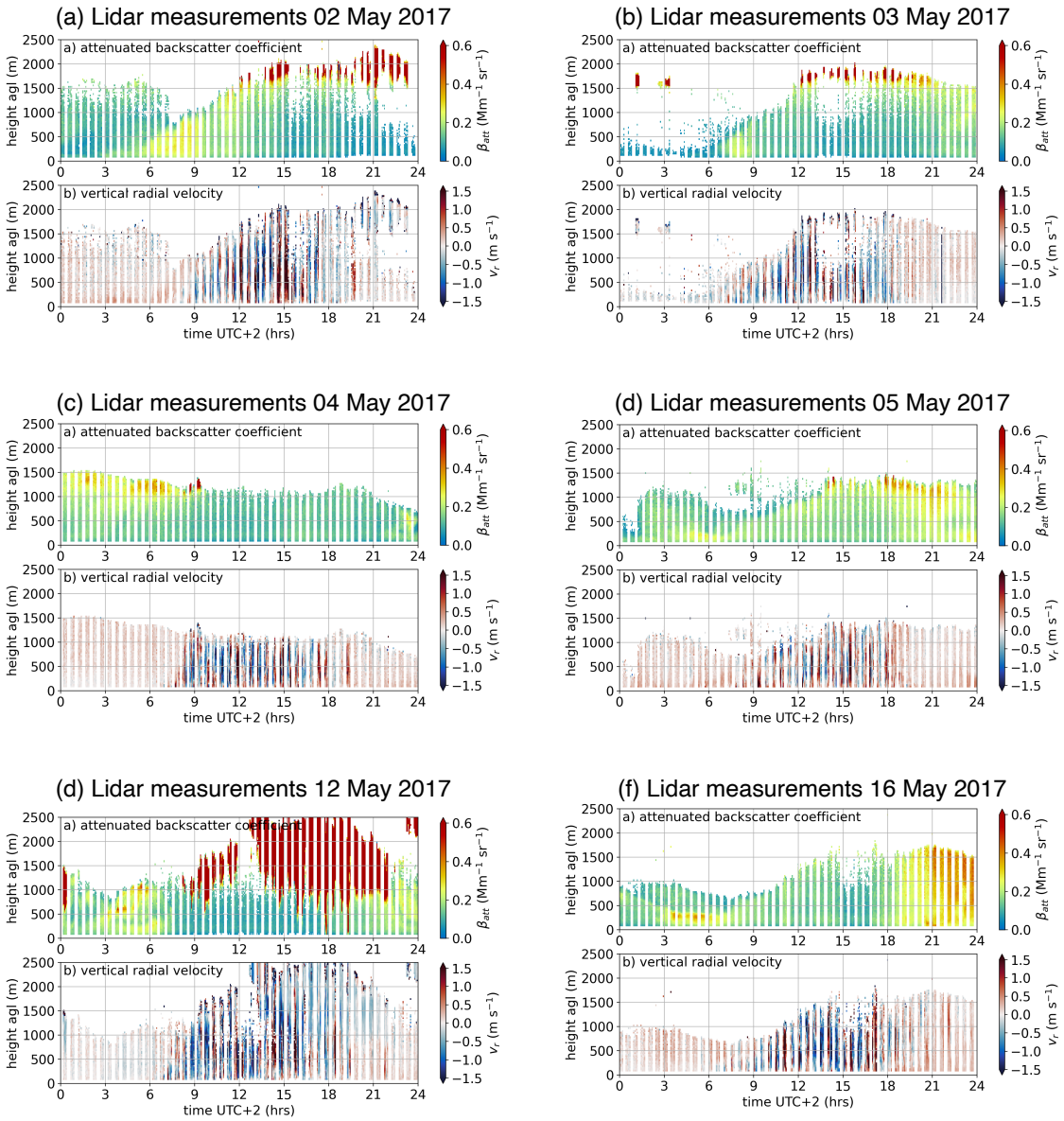

Figure S16 Doppler lidar measurement at Hyytiälä, SMEAR II station.

Each panel is showing one full day when measurements were conducted: (a) 2 May, (b) 3 May, (c) 4 May, (d) 5 May, € 12 May and (f) 16 May 2017. The sub panels show the following: (a) Attenuated backscatter coefficient and **(b)** vertical radial velocity measured on 16 May 2017 at SMEAR II station. The measurements were used to identify the different layers of the boundary layer, such as stable layer, residual layer, mixed layer and free troposphere. The vertical radial velocity (b) is showing the vertical mixing and the resulting increase of the boundary layer height.

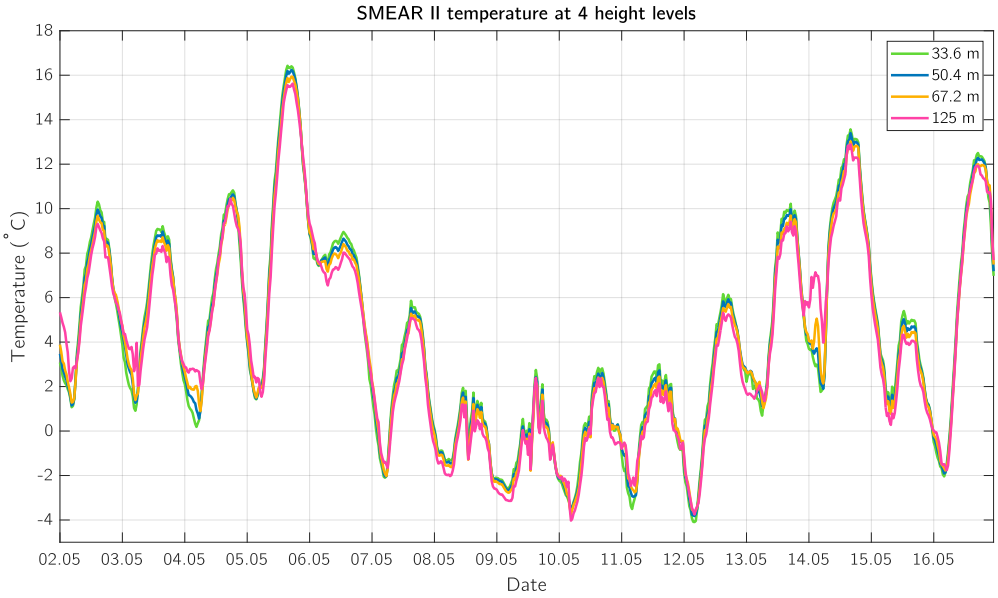

Figure S17 Temperature measured at SMEAR II station at four different heights from 2 May to 16 May 2017. The levels of measurements are 33.6 m, 50.4 m, 67.2 m and 125 m above ground level.