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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol particle formation in the upper residual layer" *by* Janne Lampilahti et al.

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We thank the Referee for the comments. It is suggested that the nucleation mode particle layers we observed might have originated from elevated upwind pollution sources, such as power station flue stacks.

As an example air mass back trajectories for the May 2, 2017 case study are shown. It is noted that the air masses arriving at 1800 m altitude above Hyytiälä at 12 UTC (this is where the aerosol particle layer was observed) traveled over a power station few hours prior to arriving in Hyytiälä.

We tested the emission hypothesis by checking if the entrained particle layer was associated with increased SO2/CO concentrations. We observed no increase in the pollu-

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tant concentrations during the day or when the particles mixed down (Fig. 1). Therefore we believe it is unlikely that these particles originated from the power station emissions.

We also checked the pollutant concentration for the second case study (May 19, 2018) we added to the manuscript (see our answer to Referee #1) and no increase in pollutant concentrations was observed when the particles mixed down at around 9:20 (Fig. 1).

Junkermann and Hacker (2018) explains that the flue stack emissions are usually released to altitudes below 400 m. In Finland the tallest chimneys are well below 200 m agl. The particle layers we observed from the Cessna were on average between 2300-2700 m above Hyytiälä. During daytime when the BL is mixing flue stack emissions would be mixed throughout the mixed layer and then stay in the residual layer the following night. One would not expect a distinct layer at the top of the RL to form. If the emissions were released into the residual layer during night, they would remain at roughly the same altitude due to lack of vertical transport during night and not be transported to the top of the RL. We think that in this case the better explanation is that the nanoparticles were formed aloft.

Comment: What are the ambient conditions relevant to particle formation or aging in the residual layer, temperature, humidity, water vapor concentration, wind speed and direction, potential condensation sink? Are aging nano-particles in this layer growing or shrinking (Kerminen et al, 2018 and references cited therein)?

Answer: According to Alonso-Blanco et al. (2017) conditions in the residual layer that would favor particle shrinkage are lack of sunlight during night and dilution because the air is cleaner. After sunrise the increased solar radiation at higher altitudes would not favor particle shrinkage. Also the lower temperature would not favor particle shrinkage. After sunrise increased solar radiation, low pre-existing aerosol particle surface area and cold temperatures would favor NPF. NPF would probably not be taking place during the night due to lack of solar radiation

Comment: 40 km is a wide range, see Fig. 1. Where is the GPS-location of the

measurements with respect to well known locations of major precursor molecule and/or primary nanoparti- cle emissions upwind? What is the flight pattern during ascents and descents? Can this be used to derive wind data from example from GPS when the Lidar is not sensitive enough?

Answer: The majority of flights were centered over Hyytiälä. We modified Figure 1 in the manuscript to also show the horizontal distribution of measurements (Fig. 2). Notable emission sources close to this area would be the city of Tampere ∼60 km SW (population ∼250000) from Hyytiälä and the Korkeakoski sawmill ∼6km SE from Hyytiälä (Eerdekens et al., 2009). When we flew over Tampere the effect on particle number concentrations was always clear. Usually the >3 nm number concentrations increased to about 5000 cm-3 from the background 2000 cm-3 at couple hundred meters above the ground.

Also we noticed the 2011-2018 dataset was not restricted to this 40 km radius from Hyytiälä. So we remade the Figures 1 and 6 in the manuscript with the 40 km boundary condition. The average vertical number-size distribution in Figure 6 did not change much but the 3-10 nm bin showed slightly negative values above the ML. For the updated figure we only used the SMPS data (Fig. 3). Also for the temperature profile we only considered profiles when there was an increased (larger than 75th percentile) sub-25 nm number concentration in the RL (2000-3000 m).

The flight patterns were straight legs perpendicular to the mean wind direction while ascending or descending. So at least the wind direction can be inferred from the direction of the flight legs.

Comment: Why are the measurements in the ascend beginning at 200 m, the descend ends at 600 m agl? Teisko, âĹij 15 km (alt 158 m) from Hyytiälä (alt 178 m) would be a location for missed approaches and legal low altitude flying. O'Dowd et al (2009) presented profiles nearly to the ground from QUEST 2003.

Answer: We extended the lowest altitude bin to 200-400 m asl (Fig. 4). The descend

ended at 500 m asl during that flight.

Comment: Are there any indicators for example from the Hyytiälä meteorological tower that can be related to vertical mixing intensity? Potential parameters could be surface temperature or temperatures in the vertical profile. Glider pilots use ground based temperature measurements for a decision when to take off.

Answer: There is a 3d anemometer close to canopy, so in principle turbulence intensity above the canopy could be calculated.

Comment: Although an SMPS is onboard there is no size distribution presented for the case study. A complete size distribution would be a mandatory information for the interpretation as it carries information about the age of the particles (and potential distance to the source location).

Answer: We added the size distribution (Fig. 4)

Comment: For comparison of airborne and ground based data the same parameters, particle size distributions and not particles on the aircraft and air ions on the ground, should be used.

Answer: We added the particle size distribution from SMEAR II (Fig. 4). The downside is that the time resolution is not as good (10 min instead of 4 min)

Comment: Whether the vertical profiles within 2 1/2 hours in the early afternoon and another flight in the morning are comparable at all remains open, see the HYSPLIT trajectories above. The vertical profiles of the morning flight including air mass history and trajectory need to be included as well.

Answer: We changed the text to say: "During this flight no elevated particle layer was observed and the number concentrations were quite uniform with altitude in the different size ranges, staying below 1500 cm-3." The profiles are included in Fig. 4.

Comment: To investigate, whether the 1.5 nm particles grow into the size range of

3-20 nm and to disentangle between NPF in a sulphur rich environment and primary emissions a better size resolution is necessary (Junkermann et al, 2011a). See there and in subsequent papers size distributions with a finer resolution in the range below 10 nm. It needs a lagrangian flight pattern to confirm that airmass change is excluded, see Junkermann and Hacker (2015)

Answer: The SMPS measurements onboard (going down to 10 nm) and the ground-based measurements at the SMEAR II station (going down to 4 nm in the Fig. 4, the smallest size channel was noisy) do not show multiple nucleation modes. The gas measurements at the field station do not suggest sulphur rich environment.

One interpretation is that the particles were horizontally advected to the site in another air mass. However the particle layer was observed aloft first and then $\sim$15 min later at the field station coupled with a downward peak in particle flux suggesting that the particles were mixed down from aloft.

Comment: The observations in the 3-20 nm range are well in agreement with the patchy structure of particle number and size distributions from previous studies in the area (O'Dowd et al, 2009, Schobesberger et al, 2013, Väänänen, et al, 2016, Leino et al, 2019) as well as the patchwork blanket of power station plumes shown by Junkermann et al (2016). All these studies point towards a significant contribution from local emission hotspots. Chemical analysis from 20 years of particle research at Hyytiälä reveals that sulphur molecules and likely ammonia are among the key substances required for the production of nanoparticle clusters. A recent publication by Hao et al (2018) about measurements at Hyytiälä even requires particulate sulphate in the residual layer mixed downward to explain the observations on the ground.

Answer: In the studies mentioned the patchiness was observed inside the mixed layer but not above. It seems there are few sub-25 nm particles above the mixed layer in Hyytiälä, except for the top of the RL (Fig. 3).

The patchiness of nucleation mode particles can have other explanations such as vari-

able cloud cover (Wehner et al., 2007), land features (O'Dowd et al., 2009), and organized convection like roll vortices (Lampilahti et al., 2020).

Comment: The sources of such sulfate particles in the atmosphere are well known and typically linked to burning processes (Bigg and Turvey, 1978, Ayers et al, 1979, Whitby et al, 1978). In Finland these sources are mostly located along the coastline, about 200 km or approximately 5-6 hours upwind of Hyytiälä, (Fig. 1, www.endcoal.org). Further examples for primary nanoparticle size, aging and emission rates are shown in the papers of Junkermann et al.

Transport via the residual layer is not an exclusive pattern, veering plumes from wind direction changes in the planetary boundary layer can explain as well the observations without additional aerosols in the residual layer (examples: O'Dowd et al, 2009, for Hyytiälä under conditions with snow covered ground or Laaksonen et al, 2005 (SPC, Italy) Junkermann and Hacker (2018)). In all cases 3D-meteorology is the key for analysis of these observations.

Answer: We do observe increased aerosol particle concentrations in the top parts of the RL. The case studies (May 2, 2017 and May 19, 2018) and ground-based observations from the BAECC campaign fit the idea that particles are mixing down from the top of the RL.

With such moving emission plumes we would expect to see changes in SO2/CO concentrations but for example in the case studies this was not observed.

Comment: Recent and historic literature is not always taken into account. There are not many airborne studies of nanoparticles, but they should be included.

Answer: We extended the Introduction, see the answer to Referee #1.

Comment: Time within the manuscript is mixed between UTC and Eastern European Summer Time (EEST) in figures and text

Answer: All time should be fixed to UTC now.

Comment: Fig. 5: Scales for Theta and water vapor are missing. The figure is not really sup- portive, it suggests a high mixed layer at night although the upper rim of the Lidar data reflect only the vertical range of the measurements. Significant TKE for vertical mixing is restricted only to daylight hours.

Answer: We added temperature soundings on top of the lidar data (Fig. 4). The point of showing the temperature soundings was to show where the temperature inversion was and it agrees with the mixed layer height based on TKE dissipation rate. During the night the vertical mixing reduces but the temperature inversion remains present and shows where the top of the night time residual layer was. In all the figures where we show the lidar data and the soundings we added the temperature scale to the top.

Comment: please take into account: a few hours upwind of Hyytiälä one of Finland's largest pollution source âĹij150 kg sulphur dioxide / his located, emitting both a large amount of primary particles and a mixture of substances relevant to nanoparticle formation independent on the time of the day

Answer: We added the following paragraph to the case study

"The air masses came from the Arctic Ocean over northern Scandinavia. They went over the west coast of Finland where there are known pollution sources (most notably the Vaskiluoto coal-fired power plant), however in Hyytiälä the SO2 and CO levels remained low all day ($\sim$0.025 ppb and $\sim$115 ppb for SO2 and CO, respectively). Even when the particles were observed at the surface no increase in pollutant concentrations was observed. Pollution released into the night time RL from elevated sources such as flue gas stacks would be expected to form layers at roughly the altitudes where the emissions occured below few hundred meters. This is because of the lack of vertical mixing. If the pollution was released during daytime into a ML, it would be uniformly mixed into the ML and stay like that in the RL during night (Junkermann and Hacker, 2018). The likely explanation for sub-10 and sub-3 nm particles at this altitude is NPF."

Comment: Fig. 9 should be discussed in terms of the annual variability of meteorology,

for example the intensity of convection under typical weather conditions in Finland. The intensity of the vertical mixing process described in the manuscript is dependent on surface conditions (snow until the end of March?) and surface and vertical profile temperatures throughout the year.

Answer: The days were sunny spring days without snow (and one sunny day in July). On such days the ML is expected to be well-mixed and the particles should reach the surface in less than an hour or so (Stull, 1988). We looked at the soundings released at ∼11:20 and ∼17:20 from Hyytiälä during these days. The approximately constant potential temperature profiles suggest a well-mixed layer (Fig. 5).

Comment: Fig. 10 is outdated and needs severe revision. Sources are not always on the ground they can be elevated as well

Answer: We will add some trees to represent biogenic emissions and smokestacks to represent anthropogenic emissions of precursors. However relative to the ∼2.5 km asl altitude where the particle layers were on average observed (Fig. 3) we find it does not make much difference to distinguish between sources that are at ∼150m altitude or at surface.

Comment: Platis et al 2015 should be Platis et al, 2016

Answer: 2015 should be correct (see: https://link.springer.com/article/10.1007/s10546-015-0084-y)

Comment: Junkermann and Hacker (2018) is cited in the text but missing in the reference list.

Answer: It is added to the list

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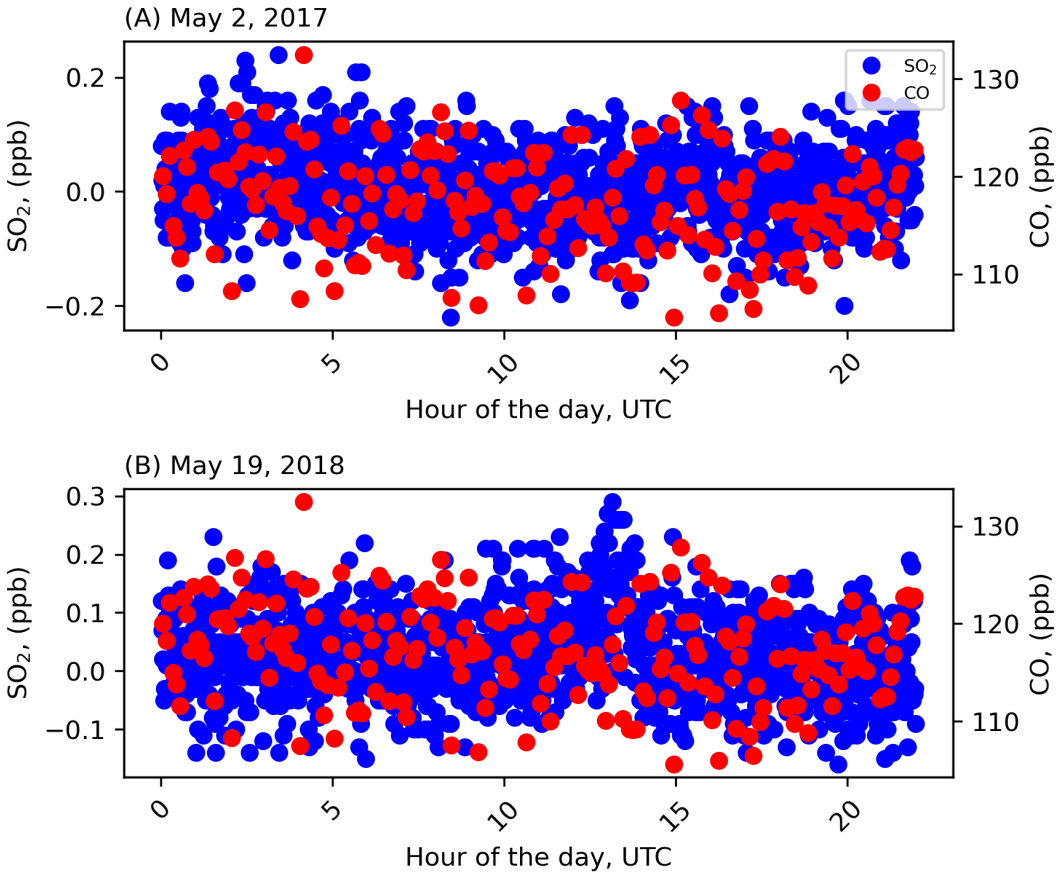

Fig. 1.

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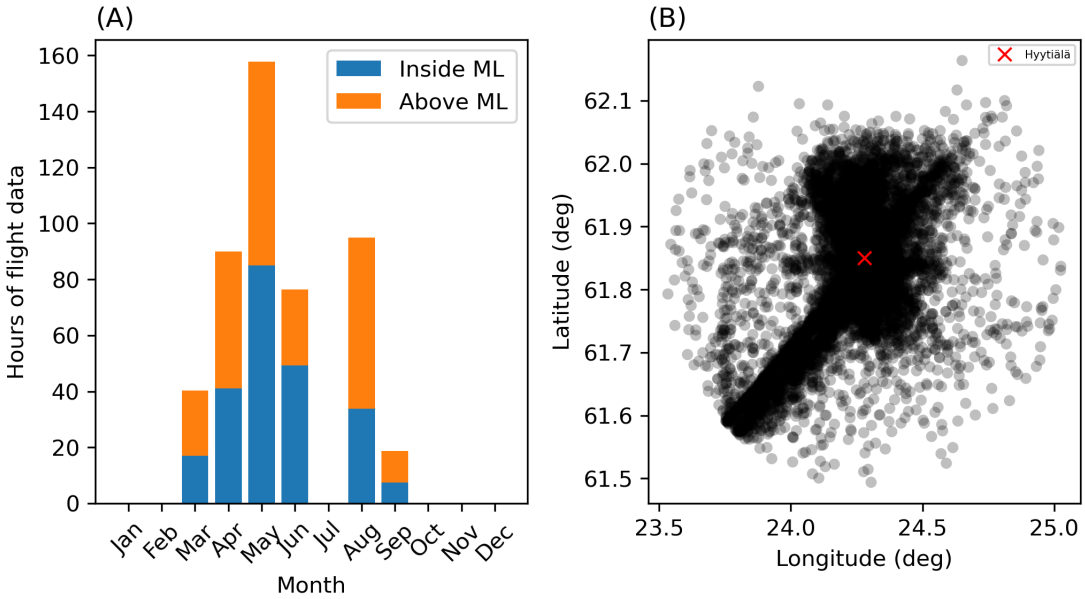

Fig. 2.

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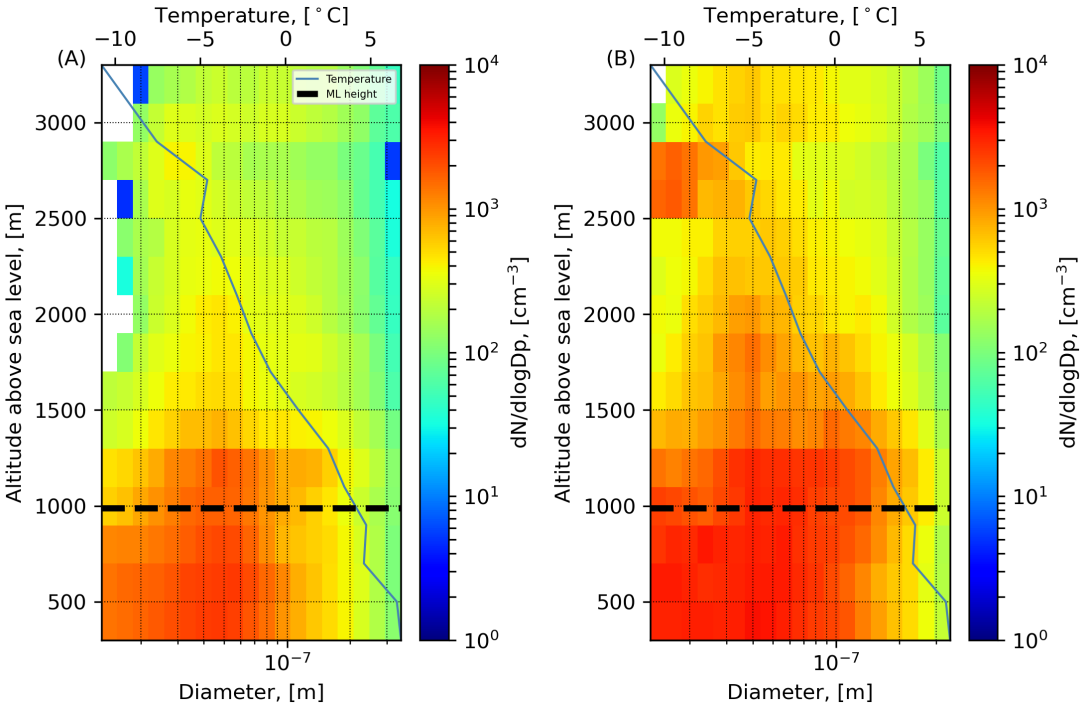

Fig. 3.

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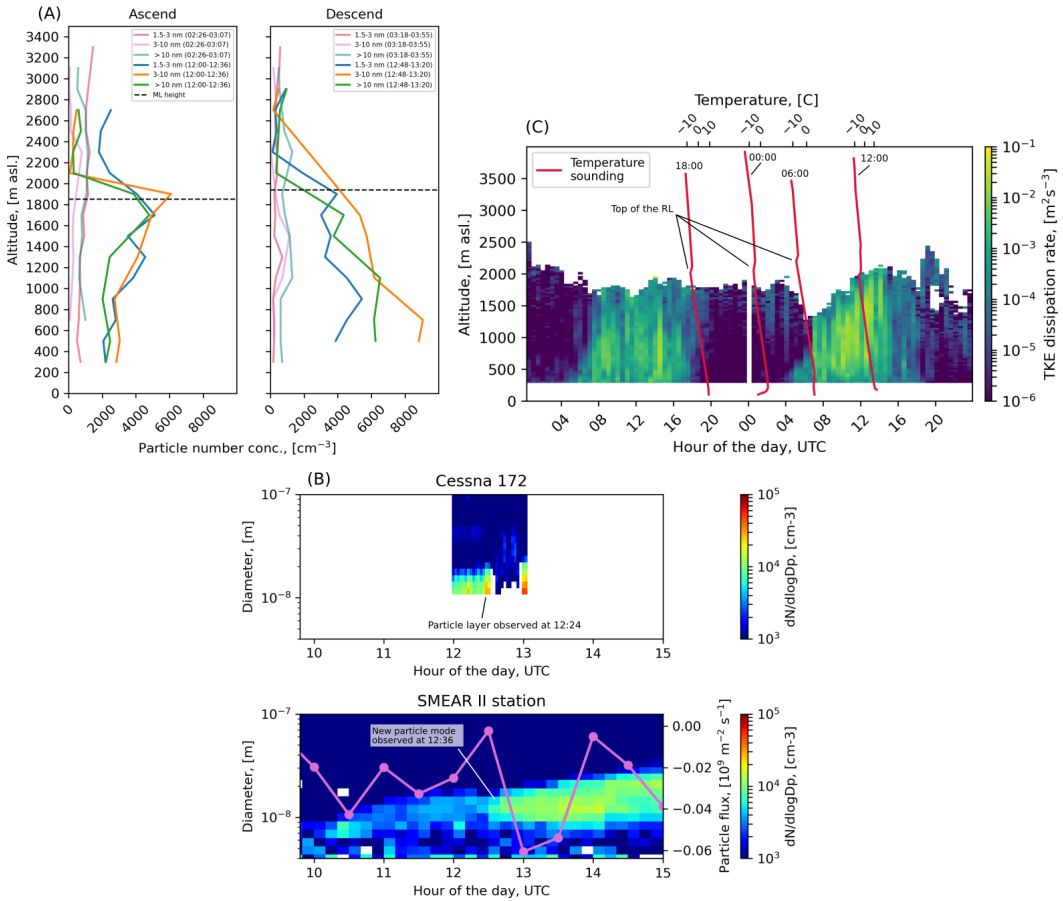

Fig. 4.

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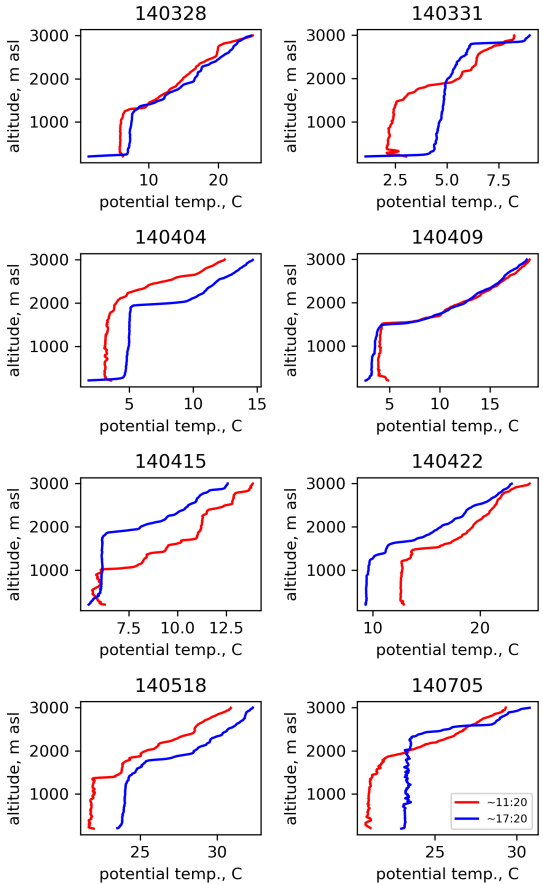
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altitude, m asl
potential temp., C
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Fig. 5.