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1 **In-Situ observation of New Particle Formation in the upper troposphere / lower**
2 **stratosphere of the Asian Monsoon Anticyclone**

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19 **Abstract**

20 During the monsoon season of the year 2017 the airborne StratoClim mission took place in
21 Kathmandu, Nepal with eight mission flights of the M-55 *Geophysica* in the upper troposphere /
22 lower stratosphere (UT/LS) of the Asian Monsoon Anticyclone (AMA) over northern India, Nepal
23 and Bangladesh. More than hundred events of New Particle Formation (NPF) were observed. In
24 total, more than two hours of flight time were spent under NPF conditions as indicated by the
25 abundant presence of ultrafine aerosols, i.e. with particle diameters d_p smaller than 15 nm,
26 which were *in-situ* detected by means of condensation nuclei counting techniques. Mixing ratios
27 of ultrafine particles (n_{uf}) of up to $\sim 50000 \text{ mg}^{-1}$ were measured at heights of 15 – 16 km
28 ($\theta \approx 370 \text{ K}$). NPF was most frequently observed at $\sim 12 - 16 \text{ km}$ altitude ($\theta \approx 355 - 380 \text{ K}$) and
29 mainly below the tropopause, but n_{uf} remained elevated ($\sim 300 - 2000 \text{ mg}^{-1}$) up to altitudes of
30 $\sim 17.5 \text{ km}$ ($\theta \approx 400 \text{ K}$) while under NPF conditions the fraction (f) of submicrometre-sized non-
31 volatile particle residues ($d_p > 10 \text{ nm}$) remained below 50 %. At $\sim 12 - 14 \text{ km}$ ($\theta \approx 355 - 365 \text{ K}$)
32 the minimum of f ($< 15 \%$) was found, and underneath the median f generally remains below
33 25 %. The persistence of particles at ultrafine sizes is limited to a few hours, mainly due to
34 coagulation, as demonstrated by a numerical simulation. Thus, NPF is detectable only for a
35 limited period of time and the frequency of NPF events observed during StratoClim 2017
36 underlines the importance of the UT/LS within the AMA as a source region for aerosols. The
37 effective *in-situ* production of aerosol in the tropopause region and subsequent coagulation
38 and/or condensation likely contribute to the formation and maintenance of the Asian
39 Tropopause Aerosol Layer (ATAL). The observed abundance of NPF-produced ultrafine particles



40 within the AMA is not unambiguously attributable to (a) specific source regions in the boundary
41 layer (according to backward trajectory analyses), or (b) the direct supply with precursor
42 material by convective updraught (from correlations of NPF with carbon monoxide), or (c) the
43 recent release of NPF-capable material from the convective outflow (according to air mass
44 transport times in the TTL). Temperature anomalies of more than one Kelvin, as observed with a
45 wavelength of ~ 90 km during a level flight over several hours, could be associated with the NPF
46 process as a possible cause for the increasing supersaturation of the NPF precursor system. The
47 frequency of NPF observed during StratoClim 2017 exceeds all previous NPF detections with
48 COPAS at TTL levels over Brazil, Northern Australia, or West Africa. The observed NPF
49 abundance and productivity of fresh aerosols during StratoClim 2017 indicates that NPF is
50 capable of directly affecting the extent and persistence of the ATAL.

51 **1. Introduction**

52 Aerosol particles in the upper troposphere / lower stratosphere (UT/LS) influence the radiative
53 balance of the Earth's atmosphere, stratospheric ozone chemistry, and properties of cirrus
54 clouds near the tropopause (Kremser et al., 2016). UT/LS aerosols are mainly composed of
55 sulphuric acid (H_2SO_4), nitric acid (HNO_3), water (H_2O), and organic compounds. Additionally,
56 the particles include fractions of non-volatile (or refractory) material (e.g. Murphy et al. (1998);
57 Murphy et al. (2006); Curtius et al. (2001); Heald et al. (2005); Froyd et al. (2010); Borrmann et
58 al. (2010); Murphy et al. (2014); Schneider et al. (2020)). Non-volatile components of
59 stratospheric aerosol particles originate from (1) natural tropospheric sources, e.g., volcanoes,
60 biomass burning, or pyro-cumulonimbus, (2) from meteoric ablation, or (3) they are
61 anthropogenic, as, for instance, space debris, rocket exhaust fumes, and products from
62 combustion (Kremser et al. (2016)). Chemical and microphysical processes, which involve the
63 stratospheric aerosol, could be influenced by solutes that, e.g., had previously been constituents
64 of the refractory aerosol compounds. Soot, mineral dust, aerosol from biomass combustion,
65 meteoric ablation material, inorganic salts, and other species probably make up the largest share
66 of the non-volatile components of aerosol particles in the UT/LS. In the tropics, underneath the
67 tropopause, the number of non-volatile fine-mode particles (i.e. smaller than $1 \mu\text{m}$ and larger
68 than 10 nm in diameter d_p) typically exhibits a characteristic minimum, resulting in a fraction of
69 $\sim 20 \%$ (and less) of non-volatile aerosol particles (cf. Borrmann et al. (2010); Weigel et al.
70 (2011)). Above the tropopause, at potential temperatures greater than $390 - 400 \text{ K}$, a maximum
71 contribution of non-volatile aerosol constituents seldom exceeds 50% (*ibid.*). Schneider et al.



72 (2020) recently provided laser ablation mass spectrometric analyses of refractory particles in
73 the LS region between the equator and the Arctic, which indicate detectable signatures of
74 meteoric ablation material at all sample locations in the LS. They assume that the meteoric
75 ablation material is partly present as solute or as insoluble inclusion within stratospheric H₂SO₄-
76 H₂O-droplets.

77 In general, the typical particle size distribution of the stratospheric aerosol is characterised by
78 processes such as formation of new particles and their coagulation, the condensation of
79 saturated vapours, and the removal from the stratosphere into the troposphere. In the tropics,
80 above the level of zero net radiative heating where scavenging is lacking in the absence of
81 clouds, aerosol particles are available for isentropic dispersion or upward transport into the
82 stratosphere. Sedimentation or isentropic transport and mixing remove particles from the
83 stratosphere (Thomason and Peter (2006); Kremser et al. (2016)). Moreover, the aerosol
84 removal from the stratosphere occurs with particular efficiency via large-scale air mass
85 subsidence in the polar winter vortex in both, the Arctic (Weigel et al., 2014) and the Antarctic
86 (Campbell and Deshler, 2014).

87 The process of homogeneous nucleation (also known as gas-to-particle-conversion), herein
88 referred to as New Particle Formation (NPF), is considered as one of the most important sources
89 of the H₂SO₄-H₂O solution droplets prevailing in the UT and Tropical Tropopause Layer (TTL).
90 The reservoir of stratospheric H₂SO₄ is maintained by oxidation of gaseous precursors like
91 sulphur dioxide (SO₂), carbonyl sulphide (OCS), and carbon disulphide (CS₂), or Dimethyl
92 sulphide (Thomason and Peter (2006); Kremser et al. (2016)) from sea surface emissions, from
93 volcanism or from anthropogenic pollution, which often undergoes long range transport before
94 reaching the TTL (e.g. Law et al., 2010). Sporadically, explosive volcanism injects large quantities
95 of SO₂ directly into the stratosphere (Vernier et al. (2011b); Kremser et al. (2016)). Within the
96 planetary boundary layer, SO₂ is found in mixing ratios from 20 pmol mol⁻¹ to more than
97 1 nmol mol⁻¹. SO₂ mixing ratios of up to several hundreds of nmol mol⁻¹ are found in the vicinity
98 of cities and highly polluted areas (Seinfeld and Pandis, 2016). From the boundary layer, SO₂ can
99 be transported very efficiently by deep convection within cumulonimbus (Cb) clouds to UT
100 heights. Although SO₂ is efficiently bound within clouds and dissolved in cloud hydrometeors,
101 cloud-resolving model calculations suggest that a proportion of 40-90 % of SO₂ may reach the



102 outflow region of deep convection (Barth et al., 2001), and these calculations are largely
103 consistent with estimates by Crutzen and Lawrence (2000). However, other model studies
104 (Ekman et al., 2006) show that only 30 % of SO₂ from the boundary layer reaches the cloud top.
105 Laboratory investigations by Jost et al. (2017) yielded a comparatively moderate retention
106 coefficient (0.2 – 0.5) of SO₂ in the ice phase of clouds, compared to a retention of 100 % for
107 hydrochloric acid (HCl) and for nitric acid (HNO₃) (*ibid.*). Hence, large fractions of the in-cloud
108 dissolved SO₂ leave the cloud ice composite as soon as the cloud particles freeze or riming
109 occurs. Alternatively, the SO₂, which remains in the cloud ice composite, is entirely released
110 when the ice particles sublimate in the convective outflow region, or below, while the ice
111 particles sediment. Crutzen and Lawrence (2000), as well as Barth et al. (2001), however,
112 clarified that cloud's acidity determines its capacity to remove a soluble gas (such as SO₂).
113 Results from airborne *in-situ* measurements of SO₂ at altitudes between 8 and 15 km were
114 compiled by Thornton et al. (1999). Remote MIPAS observations were compared by Höpfner et
115 al. (2015) with SO₂ data from *in-situ* measurements between 8 and 12 km altitude, which were
116 carried out before the year 2001. At altitudes between 8 and 15 km, the mean values of SO₂
117 mixing ratio vary between 5 and 800 pmol mol⁻¹ in the northern hemisphere, between 8 and
118 120 pmol mol⁻¹ in the tropics, and between 5 and 20 pmol mol⁻¹ in the southern hemisphere
119 (Kremser et al., 2016). Enhanced SO₂ mixing ratios in the vicinity of the tropopause are often
120 observed in connection with the uplift of polluted air masses by Warm Conveyor Belts (WBC)
121 (*ibid.*). Apart from sulphuric acid, potentially also other species contribute to particle nucleation
122 and growth, such as organics (Metzger et al. (2010); Kerminen et al. (2010)), amines (Kürten et
123 al. (2018)) or ammonia (e.g. Kirkby et al. (2011); Kürten (2019)). Given the amount of organics
124 (Murphy et al. (2006)) and ammonia species (Höpfner et al. (2019); Stroh et al. (2020)), which
125 were found in aerosol particles at UT/TTL heights in the AMA during the StratoClim 2017
126 mission, such compounds very likely act as agents promoting NPF in the UT and TTL region.

127 **1.1 New particle formation**

128 New Particle Formation (NPF), comprises (1) the initial combination of molecules into clusters
129 (of ~ 1 nm diameter) and (2) their subsequent growth to larger diameters (Kulmala et al., 2013).
130 Nucleation mode (ultrafine) aerosol particles with diameter (d_p) of at least 3 nm frequently form
131 in considerable quantities from gaseous precursors. Once formed, the particles are subject to



132 altering processes (e.g. coagulation, growth by condensation of water vapour and other gases,
133 evaporation, scavenging). Within the entire atmosphere, NPF seems ubiquitous as was
134 demonstrated by various studies and observations of NPF's occurrence:

- 135 • at or close to the surface (Kulmala et al. (2004); Nieminen et al. (2018)),
- 136 • at elevated altitudes within the boundary layer (e.g. Sellegri et al. (2019); Wehner et al.
137 (2015), Crumeyrolle et al. (2010); Venzac et al. (2008)),
- 138 • in the boundary layer and in the free troposphere under the direct influence by volcanic
139 activity (e.g. Sahyoun et al. (2019)),
- 140 • up to tropopause altitudes and the TTL region (Kerminen et al. (2018); Williamson et al.
141 (2018); Williamson et al. (2019).

142 Modelling studies suggest that the NPF process constitutes one of the most important
143 contributions (up to 45 %) to global mean tropospheric concentrations of Cloud Condensation
144 Nuclei (CCN) activated at 0.2 % supersaturation (Merikanto et al., 2009). Uncertainties remain
145 concerning the effectiveness of NPF, which complicates the implementation of the NPF
146 mechanism in global scale simulations of aerosol number densities (Yu et al. (2010), Zhang et al.
147 (2010)). Chamber experiments, conducted at temperatures similar to those prevailing in the UT,
148 and numerical simulations also confirm that the UT constitutes an important source region for
149 atmospheric particles (Kürten et al. (2016), Dunne et al. (2016)).

150 Based on airborne in-situ observations of high particle number concentrations together with
151 high levels of particle volatility in the cloud-free tropical UT, the conditions of NPF occurrence
152 were described for the first time by Brock et al. (1995). Between 7 and 20 km altitude, fields of
153 recent NPF events were encountered in about 20 % of the probed flight segments (Lee et al.
154 (2004)). NPF of largest intensity was observed particularly at the bottom TTL, as shown by
155 airborne measurements during missions over Brazil and over North Australia (Weigel et al.
156 (2011)). Recently, a survey of NPF occurrence in the free troposphere (~ 0.2 - 12 km altitude)
157 suggests that the NPF-produced particles persist (zonally almost invariant) as a globally
158 extending band within the tropical UT, thereby covering 40 % of the Earth's surface (Williamson
159 et al., 2019).



160 All NPF observations, which were made during the StratoClim 2017 mission in the UT and TTL
161 region at altitudes of up to 20 km in the Asian Monsoon Anticyclone, are discussed in their
162 entirety within this study. The specific differentiation of NPF data is provided in Weigel et al.
163 (2020b) where NPF encounters in the presence of cloud ice particles are separately studied and
164 where it is shown that NPF proceeds largely unaffected by the faint ice clouds typically occurring
165 in the TTL.

166 **1.2 The StratoClim 2017 field campaigns in the Asian Monsoon Anticyclone**

167 Between 27 July and 10 August 2017, during the Asian monsoon season, a total of eight scientific
168 flights with the high-altitude research aircraft M-55 *Geophysica* over parts of the Indian
169 subcontinent were performed from Kathmandu, Nepal (27° 42' 3" N, 85° 21' 42" E) during the
170 StratoClim 2017 mission (see Figure 1, and see also Stroh et al. (2020)). Some of these flights
171 partly spanned out of Nepalese airspace, to East India, Bangladesh, and to the northern part of
172 the Bay of Bengal.

173 The Asian Monsoon Anticyclone (AMA) represents one of the most important circulation
174 systems, mostly associated with deep convection, which mainly determines the circulation in the
175 UT/LS during the monsoon season over the Indian subcontinent. From the beginning of June
176 until about the end of August, the large-scale anticyclone persists at altitudes from the UT to the
177 LS regions (e.g. Randel and Park (2006), Park et al. (2007)), extending over longitudes from East
178 Asia to the Middle East/ East Africa (e.g. Vogel et al. (2014), Vogel et al. (2019)). The anticyclonic
179 rotation of the system induces a horizontal transport barrier inside the UT/LS (Ploeger et al.
180 (2015)), which abates the isentropic exchange between the AMA's interior and its surrounding.
181 Air masses in the region of the Asian monsoon are rapidly lifted by convection up to the
182 maximum level of convective outflow (~ 360 K, corresponding to ~ 13 km) followed by a slow
183 diabatic lift superimposed on the anticyclonic motion (e.g. Vogel et al. (2019)). This mechanism
184 transports young air to UT/LS altitudes during boreal summer and in this way various
185 pollutants and other gaseous material (Glatthor et al. (2015); Chirkov et al. (2016); Pan et al.
186 (2016); Santee et al. (2017)) and in particular water vapour (Ploeger et al. (2013)) are lifted into
187 the UT/LS region within the AMA. Based on satellite studies, the existence of the aerosol layer at
188 tropopause altitudes within the AMA region (ATAL – Asian Tropopause Aerosol Layer) was
189 proven and investigated (Vernier et al. (2011a); Thomason and Vernier (2013)). The existence



190 of the ATAL is further confirmed by in situ balloon-borne backscatter measurements (Vernier et
191 al. (2015); Vernier et al. (2018); Brunamonti et al. (2018); Hanumanthu et al. (2020)) in Lhasa
192 (August 2013), Saudi Arabia (August 2015) and India (August 2016, 2017) as well as recently by
193 aircraft measurements of Mahnke et al. (2020).

194 Hence, the constituents of the rising young air may also include precursor material from
195 anthropogenic (Vernier et al. (2015), Yu et al. (2015)) and other sources, which maintain the
196 observed ATAL. The NPF process in the TTL region could contribute significantly to the
197 formation and persistence of ATAL as a source of additional aerosol material (He et al., 2019).
198 Once the boundary layer material has reached UT/LS levels within the AMA, the elevated
199 tropopause potential temperature during the monsoon season allows the material's isentropic
200 dispersion into the "overworld" stratosphere (Pan et al. (2016)). Thereby, it is under debate
201 whether the upward transport is best described with the model of a draughting "chimney" or of
202 a pushing "blower" (Pan et al. (2016)). However, three-dimensional simulations with the
203 Chemical Lagrangian Model of the Stratosphere (CLaMS) and backward trajectory analyses show
204 that by end of August, during the 2008 monsoon season, comparatively young air masses
205 (younger than 6 months) reach the top of the AMA at about 460 K potential temperature
206 (corresponding to ~ 60 hPa). According to these simulations (Vogel et al. (2019)), air masses are
207 lifted due to diabatic (mainly radiative) heating in an anticyclonic large-scale upward spiral with
208 ascent rates of about 1 K potential temperature per day. The anticyclonic lift of air in the AMA
209 occurs across the tropopause while elsewhere, in the extra-tropics, the tropopause typically acts
210 as an obstacle for the immediate vertical transport (Vogel et al. (2019)). This capability of the
211 tropopause to cap the troposphere seems largely diminished in connection with the AMA (*ibid.*),
212 and is consistent with the conclusions of previous works (Bergman et al. (2012), Garny and
213 Randel (2016), Ploeger et al. (2017)).

214 **2 *In-situ* instrumentation**

215 **2.1 Total number concentration of sub-micrometre sized particles**

216 Particle number concentrations were *in-situ* measured by means of a 4-channel condensation
217 nuclei (CN) counter COPAS (COndensation PArTicle counting System, cf. Weigel et al. (2009))
218 with continuous flow, using the chlorofluorocarbon FC-43 as working fluid. COPAS



219 measurements and data storage are performed at a frequency of 1 Hz. To reduce the statistical
220 noise of the directly recorded raw signal of the scattered-light-detectors integrated in COPAS,
221 the 1 Hz-raw data are preprocessed by applying a 15-second running average. Three of the four
222 COPAS channels operate with different 50 % detection particle diameters d_{p50} (i.e. 6 nm, 10 nm
223 and 15 nm). The fourth COPAS channel (with $d_{p50} = 10$ nm) detects particles downstream of a
224 heated (270°C) sample flow line, resulting in measured particle mixing ratios of non-volatile
225 (nv) or refractory particles (e.g. soot, mineral dust, metallic aerosol material, etc.).

226 2.1.1 COPAS operation during StratoClim 2017

227 The forward facing aerosol inlet of COPAS is located well outside the boundary layer of the
228 aircraft. The inlet consists of two serial diffusers, which slow down the ambient air velocity to
229 the flow speed of the instruments' sampling. For stratospheric particle concentrations, the
230 COPAS measurement uncertainty of the StratoClim 2017 data discussed herein is about 15%,
231 which is due to particle counting statistics and uncertainties in the volume flow. The
232 measurement properties of COPAS are described in detail by Weigel et al. (2009), and its
233 performance has been demonstrated by several studies (Curtius et al. (2005); de Reus et al.
234 (2009); Borrmann et al. (2010); Frey et al. (2011); Weigel et al. (2011), and Weigel et al. (2014)).
235 Compared to previous missions (*ibid.*), during StratoClim 2017 a new inlet configuration was
236 required, which caused both COPAS instrument units to sample via a single aerosol inlet.

237 The coupled sampling flow through one common inlet bears the advantage of an increased
238 super-isokinetic flow ratio at the inlet's entry as compared to previous operations. This way the
239 flow speed through commonly used parts of the aerosol line setup is almost doubled, which
240 effectively reduces diffusional particle loss (cf. Weigel et al. (2009)). However, this new
241 instrument setup required a reanalysis of the corrections to account for particle loss as
242 compared to the previous values documented in (Weigel et al. (2009)). Equivalently to the
243 previously described procedure (*ibid.*), the pressure-dependent corrections for the used aerosol
244 lines and given volume flows were re-calculated (Table 1) using the method introduced by von
245 der Weiden et al. (2009) with modifications for low-pressure application. These corrections are
246 applied to the number densities of particles in the ultrafine size mode (cf. Section 2.1.3).

247 The particle densities are typically measured by COPAS in particle number concentrations N (in
248 cm^{-3}), but are also presented here as mixing ratio n in units of particles per milligram of ambient



249 air (mg^{-1}). This way the measurements from different pressure levels are consistently
250 comparable and allow direct correlations between the mixing ratios of particles and of gaseous
251 tracers. Hereafter, the notation n_{10} refers to the mixing ratio of sub-micrometre sized particles
252 with diameters greater than 10 nm. The measurement of n_6 (of particles with $d_p > 6$ nm) and n_{15}
253 ($d_p > 15$ nm) aims at the identification of recent NPF (cf. Section 2.1.3). The notation $n_{10\text{nv}}$ refers
254 to the mixing ratio of non-volatile particles with identical size range as specified for n_{10} . The
255 proportion f of non-volatile particles is given as the ratio of $n_{10\text{nv}}$ and n_{10} in percent
256 (equivalently determined from the ratio of the measured number concentrations $N_{10\text{nv}}$ and N_{10}),
257 since only non-volatile particles with sizes $d_p > 10$ nm are detected (Section 2.1.2).

258 2.1.2 COPAS detection of non-volatile (refractory) aerosol particles

259 COPAS includes a denuder-type device based on an established and commonly used technique
260 for exposing atmospheric aerosol samples to heat in order to obtain indications concerning the
261 chemical properties a) of the volatile compounds of aerosol particles (when analysed e.g. by gas
262 chromatography) or b) of the remnants, which survive the heat exposure. One of the four COPAS
263 channels is equipped with a heated stainless steel tube, which is used to vaporise volatile
264 compounds upstream of one of the particle detectors. In this way, the preheated COPAS channel
265 detects the residual aerosol component by number per sample volume. The particles that
266 remain after passing through the heated tube (at $\sim 270^\circ\text{C}$) are considered and designated
267 hereafter as non-volatile (or refractory) at given temperature (see Curtius et al. (2005), Weigel
268 et al. (2009), and Borrmann et al. (2010)). The specific heating temperature is chosen with the
269 aim to vaporise mainly stratospheric particle species, which typically consist of aqueous
270 solutions of sulfuric acid ($\text{H}_2\text{SO}_4\text{-H}_2\text{O}$) and/or nitric acid ($\text{HNO}_3\text{-H}_2\text{O}$), which reportedly volatilise
271 at 180°C (Rosen, 1971). In addition, most of volatile and several semi-volatile organic
272 compounds can evaporate at temperatures below 270°C .

273 The working principle of the COPAS aerosol vaporiser was demonstrated by means of laboratory
274 experiments with pure $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ particles of several sizes and at pressure conditions between
275 70 – 300 hPa (Weigel et al., 2009); more than 98 % of the sub-micrometre sized $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$
276 particles were volatilised. As refractory material, which could be detectable with COPAS, is
277 unlikely to be generated by the heater itself, instrumental artefacts may be excluded as a
278 potential cause of false observation. To avoid artefacts as a result, e.g., of re-suspension of



279 aerosol material, which had been deposited on the tube's inner walls during previous operations,
280 the sample lines were flush-cleaned with ethanol and distilled water, at least before every
281 second mission flight. Inefficiencies of the vaporiser, e.g. due to diminished heat transfer from
282 the tube's inner wall to the passing aerosol particles, particularly at low atmospheric pressures,
283 would cause the number (fraction) of detected refractory particles to be unexpectedly high
284 ($f \approx 100\%$) over extended measurement periods, which was not observed throughout the field
285 missions (cf. Borrmann et al. (2010); Weigel et al. (2011)). Conversely, instrumental artefacts
286 inherent with the vaporiser's tube length, e.g. particle loss, would lead to comparatively low
287 number concentrations of detected refractory particles. Diffusional loss effects increase with
288 decreasing pressure, but thermophoresis should counteract the particles' diffusion towards the
289 hot tube walls. With the same vaporiser system, Weigel et al. (2014) observed rising mixing
290 ratios of refractory aerosol, most likely from meteoric ablation, with altitude at stratospheric
291 levels inside the polar vortex, while outside the vortex the amount of refractory aerosols nearly
292 stagnated over the corresponding altitude range. This may additionally confirm the principle
293 function of the vaporiser.

294 2.1.3 NPF criterion and event definition

295 To serve as an indication of recent NPF (within hours prior to the observation), the number
296 concentration of ultrafine aerosol particles (hereafter referred to as N_{uf}) results from the
297 difference $N_6 - N_{15} = N_{6-15}$, which moreover requires meeting the NPF criterion:

$$298 \quad 0.8 \cdot N_6 - 1.2 \cdot N_{15} > 0. \quad (1)$$

299 This criterion was reassessed for the StratoClim 2017 data set based on the definition used by
300 Weigel et al. (2011) to account for the COPAS detectors' signal-to-noise ratio and the counting
301 statistics. The NPF criterion therefore sets a conservative threshold (*ibid.*) to take an overall
302 uncertainty of up to 15 % of the individual COPAS channels into account. Resulting N_{6-15} are then
303 subject to corrections concerning particles' diffusional loss inside the aerosol lines as described
304 in Section 2.1.1 (cf. also Table 1). The calculated number concentrations N_{6-15} were multiplied by
305 the correction factor κ_L (Table 1) being a function of static pressure during the measurements.

306 Provided that the NPF criterion is met, a series of measurement points is denoted as an event if
307 the measured number concentration (or mixing ratio) of ultrafine particles remains



308 continuously greater than zero for at least 5 measurement seconds. Limitations of this event
309 definition concern observations, which lasted over time periods between one and five seconds.
310 Overall, 25 cases of a total of 130 individually encountered events are affected by this instance.
311 Specifications such as the number of newly formed particles and the duration of such short
312 events is uncertain: too short signal features (e.g. over one second) and thus fine spatial
313 structures are smoothed by the data pre-processing or filtered out by applying the NPF criterion.
314 Note, during one second the measurement platform has moved a horizontal distance of ~ 150 m
315 and a vertical distance of up to ~ 10 m assuming cruising speed and a maximum ascent/descent
316 rate of 10 m s^{-1} . Based on the mean flight speed of the M-55 *Geophysica* ($\sim 154 \pm 39 \text{ m s}^{-1}$), this
317 definition implies that an event (i.e. elevated N_{uf} for more than 5 seconds) covers a horizontal
318 distance of ~ 770 m (into flight direction of a straight heading). The corresponding event
319 definition applies also for investigations concerning the occurrence of NPF in the presence of
320 cloud ice elements during the StratoClim 2017 mission (see Weigel et al. (2020b)).

321 The time period during which the event criterion (Equation 1) is fulfilled, i.e. during which the
322 number of ultrafine particles remains at significantly elevated levels, is referred to hereafter as
323 the NPF event duration. From this primary information of measured data, the horizontal extent
324 of NPF fields is derivable with caveats. On the one hand, such estimates are limited by the
325 assumption that an encounter of elevated N_{uf} (over tens of seconds and minutes) is actually due
326 to a single NPF event and does not consist of a series of possibly overlapping events. On the
327 other hand, the determined horizontal distances refer to the average flight speed
328 ($\sim 154 \pm 39 \text{ m s}^{-1}$) and the flight attitude is assumed as unchanged during the event duration.

329 Hereafter, an NPF event is denoted as *elevated* if detected aerosol densities of ultrafine particles
330 exceed 10000 mg^{-1} . The terms *intermediate* or *weak* NPF are used in connection with number
331 densities of particles in the ultrafine size mode in the range of $1000 \text{ mg}^{-1} < n_{\text{uf}} < 10000 \text{ mg}^{-1}$ or
332 $< 1000 \text{ mg}^{-1}$, respectively. This classification refers to laboratory studies by (Kirkby et al. (2011),
333 Kürten et al. (2016)); according to these the NPF-rate and, hence, the NPF intensity (i.e. its new
334 particle productivity) varies with the degree of supersaturation of the vapour from which the
335 new particles form. However, it should be noted that hereafter the notation *most intense* NPF, i.e.
336 events of particularly increased ultrafine particle production, is often used synonymously with
337 *most recent* NPF. Due to the short persistence of the freshly formed particles in the ultrafine size



338 mode (cf. Section 4.5), an intense NPF event is still proceeding when observed, or it had phased-
339 out very recently (within hours) before the detection. For NPF encounters with low or
340 intermediate n_{uf} , the conclusions concerning the event's age remain ambiguous.

341 2.2 The Ultra-High Sensitive Aerosol Spectrometer UHSAS-A

342 The measurements of the aerosol particle size distributions during the StratoClim 2017 field
343 campaign (Höpfner et al. (2019); Stroh et al. (2020)) were performed with an in-house modified
344 airborne version of the Ultra High Sensitive Aerosol Spectrometer (UHSAS-A; manufacturer DMT
345 Inc., Longmont, CO, USA). The modifications made to the flow and pumping system of the
346 UHSAS-A enabled maintaining constant system-flows (sample-, sheath-, purge-flow) through the
347 instrument even under ambient (stratospheric) pressures as low as 50 hPa. The airflow system
348 of the UHSAS-A was characterised and calibrated in the laboratory prior to the StratoClim 2017
349 field campaign using a controlled low-pressure chamber. The size binning of the aerosol particle
350 size distributions results from instrument's laboratory characterisations with size-classified
351 particles (by means of a Differential Mobility Analyzer; DMA), such as Polystyrene Latex (PSL),
352 Sodium Chloride, Ammonium Nitrate and Ammonium Sulfate. The particle sizing performance of
353 the UHSAS-A throughout the field campaign was monitored by means of calibrations prior to
354 each mission flight. For these calibrations, exclusively PSL particle standards were used. The
355 uncertainty of the number concentration measured by the UHSAS-A with 1-Hz resolution was
356 determined to be approximately 10 % for the particle diameter range of $65 \text{ nm} < d_p < 1000 \text{ nm}$.
357 This uncertainty is based on laboratory characterisations of the sample-flow measurement and
358 of the counting efficiency of the instrument (Mahnke et al., 2020). Some of the results from the
359 measured particle size distributions and a comparison with other instruments and the Cloud-
360 Aerosol Lidar with Orthogonal Polarization (CALIOP) are also presented by Mahnke et al.
361 (2020).

362 2.3 Carbon monoxide measurements

363 At tropospheric altitudes, the role of carbon monoxide (CO) is understood as a pollutant (Park et
364 al. (2009)), thus, CO is often used as a representative pollution tracer (e.g. Pan et al. (2016)). The
365 main sources of CO are natural as well as anthropogenic, including combustion, and the
366 conversion from hydrocarbons due to oxidation. The entire tropospheric CO budget is assumed



367 to be maintained almost equivalently by: (1) its photochemical production and (2) its emission
368 from ground sources. The atmosphere's CO content is mainly depleted via oxidation with
369 hydroxyl radical (OH) (Logan et al. (1981), Seinfeld and Pandis (2016)). The CO's atmospheric
370 lifetime is comparatively short and ranges at about two to three months (Zahn et al. (2002);
371 Hoor et al. (2004); Hoor et al. (2005)). CO is frequently used as tropospheric tracer for
372 investigations concerning the air mass transport in the troposphere, across the tropopause, and
373 in the lowermost stratosphere. In the free troposphere, CO mixing ratios typically range between
374 50 nmol mol^{-1} (unpolluted) and values of up to $700 \text{ nmol mol}^{-1}$ (polluted) next to emission
375 sources (Clerbaux et al. (2008), Park et al. (2009)). CO mixing ratios remain comparatively high
376 ($\geq 100 \text{ nmol mol}^{-1}$) within the AMA and up to altitudes of $\sim 15 \text{ km}$. Between 15 km and 20 km
377 altitude, CO mixing ratios gradually decrease down to $\sim 40 \text{ nmol mol}^{-1}$ (Park et al. (2009)).

378 During the StratoClim 2017 mission, CO mixing ratios were determined by means of the tunable
379 diode laser (TDL) detection principle, which the analyser Carbon Oxide Laser Detector-2 (COLD-
380 2) spectrometer is based on. According to comprehensive comparisons to the previous
381 instrument version COLD (Cryogenically Operated Laser Diode, 4 s temporal resolution, (Viciani
382 et al., 2008)), the new system implies several improvements (Viciani et al., 2018). The laser
383 source is now a room temperature Quantum Cascade Laser (QCL) that no longer requires a
384 liquid nitrogen cooling, which also reduces size, weight and operational complexity. The
385 measurement's temporal resolution is improved by a factor of four, the in-flight sensitivity of the
386 COLD-2 spectrometer ranges at about 2 nmol mol^{-1} at integration times of 1 s , and an accuracy of
387 3% is specified for the CO measurement with COLD-2 (Viciani et al., 2018).

388 **2.4 Meteorological measurements**

389 Atmospheric temperature and pressure data were taken from the Unit for Connection with the
390 Scientific Equipment (UCSE, Sokolov and Lepuchov (1998)), which is a part of the navigational
391 system of the M-55 *Geophysica*. UCSE data are available as 1-Hz -resolved ambient pressure
392 (accuracy: $\pm 1 \text{ hPa}$) and temperature ($\pm 2 \text{ K}$ accuracy). Based on these UCSE data, the potential
393 temperature θ along the mission flight tracks is calculated in compliance with the definition by
394 the World Meteorological Organization (WMO (1966)).



395 **3 Analytical methods**

396 **3.1 The height of the lapse-rate tropopause and the equivalent latitude**

397 Meteorological data were also taken from ERA-Interim reanalyses by the European Centre of
398 Medium-Range Weather Forecasts (ECMWF) (Dee et al., 2011). Hybrid reanalysis levels in the
399 TTL are located at various pressure heights (i.e. around 177, 154, 133, 113, 96, 80, 67, 55 hPa,
400 respectively) representing a vertical resolution of about one kilometre in this region.

401 The aircraft data are analysed in coordinates relative to the tropopause height and to the
402 monsoon anticyclone center, respectively. The height of the lapse-rate based thermal
403 tropopause was determined based on ERA-Interim data and following the WMO criterion (WMO,
404 1957) as the lowest altitude (z_0) where the temperature lapse-rate falls below 2 K km^{-1} , if the
405 average lapse-rate within an overlying layer of 2 km thickness (i.e. $z_0+2 \text{ km}$) remains below
406 2 K km^{-1} . The cold point tropopause definition often yielded ambiguous results for the
407 tropopause heights within the AMA for the StratoClim 2017 period (cf. von Hobe et al. (2020)).
408 The potential temperature at tropopause level was interpolated to the 1-Hz-resolved position
409 along the flight track of the M-55 *Geophysica*, and the measurement data were sorted as a
410 function of potential temperature distance to the local tropopause as vertical coordinate.

411 The centre of the AMA was determined based on the anomalous potential vorticity distribution
412 within the monsoon region at the 380 K potential temperature level, where lowest values of the
413 potential vorticity (PV) are found in the AMA centre. For that reason, the AMA-centred
414 equivalent latitude was calculated for a given closed PV contour as a projection onto polar
415 coordinates (Ploeger et al., 2015). Therefore, an equivalent latitude of 90° North corresponds to
416 the center of the anticyclone (lowest PV), and the equivalent latitude decreases with increasing
417 distance from the centre, or rather, towards the anticyclone's edge. Note, that the calculation of
418 AMA-centred equivalent latitude is rigorously valid within a layer of about $\pm 10 \text{ K}$ around 380 K
419 potential temperature, where a clear negative PV anomaly occurs. The uncertainties of
420 calculated equivalent latitude become significant at levels beyond the $\pm 30 \text{ K}$ range above/below
421 380 K.



422 **3.2 The Coagulation Model for investigating the particles' persistence in the ultrafine**
423 **size mode**

424 Particle coagulation comprises the processes of particle collisions and their subsequent
425 coalescence. More specifically, if two particles with masses m_i and m_j collide and coalesce, a new
426 particle with mass $m_i + m_j$ is formed. The coagulation rate of particles with masses m_i and m_j is
427 described by $\beta_{ij} n_i n_j$, where n_i and n_j are the number concentrations of particles with masses m_i
428 and m_j , respectively. The coagulation kernel β_{ij} characterises the coagulation rate. The choice of
429 the coagulation kernel depends on the type of coagulating particles, in particular, on their size.
430 Coagulation of ultrafine aerosol particles is sufficiently well described by a Brownian
431 coagulation kernel (Jacobson (2005), Equation 15.33 therein). The kernel includes a correction
432 term to account also for particles in the transition regime, i.e. the transition between the free-
433 molecular regime, where the particles are small compared to their mean free path, and the
434 continuum regime, where the particles are large compared to their mean free path.

435 The model employed in this study numerically solves the discretised coagulation equation (cf.,
436 e.g., Jacobson (2005) and Equation 15.2 therein) as formulated in the numerical chemistry-
437 climate model SOCOL (Solar Climate Ozone Links; Stenke et al. (2013)). The particles are
438 assumed as spherical, and the model is based on a discretisation of the volume space, wherein
439 the ratio of two subsequent volume size bins is constant, $\frac{V_{k+1}}{V_k} = 1.4$. The particle size range of
440 the first volume size-bin V_1 corresponds to particle diameters of $7.5 \text{ nm} < d_{p,1} < 8.5 \text{ nm}$. With a
441 total number of 40 size bins, hence the largest particle size included in this investigation is about
442 635 nm ($= d_{p,40} = (1.4)^{\frac{39}{3}} \cdot d_{p,1}$).

443 The coagulation rate and, thus, the persistence of the ultrafine particles, was simulated under
444 given background conditions during observation. As input for the simulation, the aerosol size
445 distribution detected by the UHSAS-A (nominally covering $65 \text{ nm} < d_p < 1000 \text{ nm}$, cf. Section 2.2
446 and Mahnke et al. (2020)) was extended towards smaller diameters by further particle size bins
447 obtained from the measurements with COPAS. For this case study, the NPF event on 04 August
448 2017 (KTM 5) over 26 seconds between 04:04:40 and 04:05:06 UTC (pressure altitude: 110 hPa;
449 ambient air temperature: 196 K) was selected. The detection ranges of three COPAS channels
450 determine two regimes in the ultrafine size mode, i.e. $6 \text{ nm} < d_p < 10 \text{ nm}$ and $10 \text{ nm} < d_p < 15 \text{ nm}$,



451 each of which are divided into three sub-bins, to exploit a higher particle size resolution of the
452 coagulation model. The three sub-bins within the size classes 6 – 10 nm and 10 – 15 nm were
453 uniformly set to one third of the respective concentration N_{6-10} ($\sim 10000 \text{ cm}^{-3}$) and N_{10-15}
454 ($\sim 3600 \text{ cm}^{-3}$). The difference between the total number concentrations N_{15} (COPAS) and N_{65}
455 (UHSAS-A) yields the number concentration of N_{15-65} . The number concentration N_{15-65}
456 ($\sim 5000 \text{ cm}^{-3}$) was interpolated over 13 sub-bins (with exponential degradation on increasing
457 particles size) to achieve a continued transition of the size distribution towards the detection
458 size range of the UHSAS-A. The size-segregated aerosol concentrations measured with the
459 UHSAS-A were interpolated (with respect to particle size) to the resolution of the remaining 21
460 sub-bins of the coagulation simulation. The particle concentrations $N(d_p)$ over the entire particle
461 size range from the ultrafine sizes to up to $d_p = 1 \mu\text{m}$ were converted into an aerosol size
462 distribution $dN/d \log d_p$ in cm^{-3} as a representation of an initial state and input for the
463 coagulation simulation (for more details see the results in Section 4.5).

464 It is worth noting, that for the coagulation simulation, the NPF event is considered as expired, i.e.
465 any fresh supply of ultrafine particles due to continuous or renascent NPF is excluded for the
466 simulated runtime of the coagulation process over 24 hours. Generally, constant conditions of
467 atmospheric pressure (p) and temperature (T) are assumed over the 24-hours period for the
468 simulation, as the air is lifted very slowly at TTL levels within the AMA (by $\sim 1 \text{ K}$ potential
469 temperature per day, cf. Vogel et al. (2019), corresponding to $\Delta p \approx 1\text{-}1.5 \text{ hPa}$ and $\Delta T < 1 \text{ K}$ per
470 day). However, for atmospheric layers where convective dynamics and air mass mixing are still
471 effective, the assumption of constant ambient conditions may not hold for such simulations.

472 **3.3 Analyses of trajectories and the air mass transport history**

473 Fifty – days backward trajectories were calculated for each sample collected during the
474 StratoClim 2017 mission using the trajectory module of the Chemical Lagrangian Model of the
475 Stratosphere (CLaMS; McKenna et al. (2002), Konopka et al. (2012), Pommrich et al. (2014)).
476 The CLaMS backward trajectory calculations are driven by horizontal winds and are based on
477 the new high-resolution ERA-5 reanalysis (Hersbach and Dee (2016)) which was recently
478 released by the ECMWF. The improved resolution of the ERA-5 data compared to the ERA
479 interim data set should increase the reliability of tropospheric transport processes along the
480 backward trajectory analysis and may strengthen the assignment to possible source regions.



481 ERA-5 data are given on a horizontal grid of about $0.3^\circ \times 0.3^\circ$, in 1-hour temporal resolution, in
482 137 hybrid levels from the surface to the 0.01 hPa pressure altitude. Hence, a much better
483 representation of convective updraught and tropical cyclones is realised with the ERA-5 dataset
484 (Hoffmann et al. (2019)) compared to earlier reanalyses (Dee et al. (2011)), in particular, in the
485 region of the Asian summer monsoon (Li et al. (2020)). However, further validation of the ERA-5
486 products is required, thus ERA-Interim reanalyses still represent the state-of-the-art.

487 For vertical air mass transport velocities, the diabatic approach was applied using the total
488 diabatic heating rate to extract the vertical velocity, thereby including the release of latent heat
489 (for details, see (Ploeger et al., 2010)). The model boundary layer is set at $\sim 2 - 3$ km above the
490 surface following orography (cf. Pommrich et al. (2014), Vogel et al. (2015)).

491 In general, trajectory calculations have limitations due to trajectory dispersion depending on the
492 trajectory length. However, the frequently employed trajectory length to study transport
493 processes in the Asian monsoon region is ranging from a couple of weeks to a few months (e.g.
494 Chen et al. (2012); Bergman et al. (2013); Garny and Randel (2016); Müller et al. (2016); Li et al.
495 (2017) and Li et al. (2018)). The CLaMS trajectory products based on the ERA-5 dataset were
496 extensively investigated concerning their spatial and temporal resolution in connection with
497 strong vertical transport (e.g. Hoffmann et al. (2019)). However, Li et al. (2020) demonstrated,
498 by means of satellite-borne (FY-2D) brightness temperature data and balloon measurements in
499 China, that convective events over the Pacific Ocean associated with tropical cyclones are
500 resolved by CLaMS trajectory calculations with high accuracy.

501 The CLaMS backward trajectory calculations, which were initialized from each sampling position
502 along the flight track in 1-Hz resolution, were used to allocate the air's latest contact with the
503 model boundary layer at 2 - 3 km above the ground. This allows for investigating the location of
504 the sources influencing the mixing ratios in the air samples taken aboard the M-55 *Geophysica*.
505 To include the uncertainty of a certain backward trajectory, ERA-5 backward trajectories were
506 calculated for each second of air sampling during the flight.

507 **3.4 The age of air since release from convective outflow**

508 This approach aims at investigating the possible influence of recent convection on NPF, filtering
509 out small contributions from matured air, which may be mixed in an air parcel but have only a



510 minor influence on the overall air mass composition. The history of a convective air mass is
511 analysed by making use of the TRACZILLA Lagrangian model (Pisso and Legras, 2008), which is
512 a variation of FLEXPART (Stohl et al., 2005). In its recent version the model interpolates
513 velocities and heating rates directly from the hybrid grid to the position of the parcel using
514 logarithmic pressure or potential temperature as vertical coordinate. The simulations were
515 based on the release of a cluster of 1000 back-trajectories, representative of a generic aerosol
516 tracer, which is launched at respective, each trajectory cluster from a 1-second resolved time
517 step along the flight path. The trajectories were traced back over a period of 30 days in the
518 geographical domain, spanning a longitude and latitude range of 10°W-160°E and 0-50°N,
519 respectively. The meteorological fields are taken from the ECMWF reanalysis ERA-5 with 1-
520 hour-resolution, assuming diabatic vertical motion. The convective influence is then
521 distinguished from uninfluenced cases by the high-frequency images (one image per 10 –
522 15 minutes) of cloud top altitudes from the geostationary satellites MSG1 and Himawari. For
523 computational reasons Himawari images were analysed in time steps of 20 minutes. The cloud
524 top height of convective clouds is derived from the cloud top temperature and height (CTTH)
525 product, developed within the European Organisation for the Exploitation of Meteorological
526 Satellites (EUMETSAT) Satellite Application Facility (SAF) by support of Nowcasting Very Short
527 Range Forecasting (NWC) products (Derrien et al. (2010); Schulz et al. (2009)).

528 Convective sources are identified as such if the course of a trajectory within a certain
529 geographical area coincides with the cloud top level, as similarly done by Tzella and Legras
530 (2011) and Tissier and Legras (2016). The possible convective sources are classified into major
531 source region categories. It is noteworthy that, while the adopted trajectory method bypasses
532 the uncertainties related to the convective representation in the reanalysis by using
533 observation-based information on the convective events, uncertainties still remain. Those arise
534 mainly from uncertainties in the identification of the cloud top from image data of geostationary
535 satellites, the impossibility to account for the entrainment – detrainment – processes, and
536 reanalysis-related uncertainties concerning advection. For more details on the trajectory-
537 convective clouds coupling methods see Bucci et al. (2020). In the presented analysis, the air
538 mass age is computed as the difference between the time of release of the cluster and the
539 convective cloud crossing. Since the trajectory cluster can spread in space and bring different
540 contributions from different regions, only the mean age from the dominant convective source



541 (i.e. the mean age from the regions with the highest percentage of convective clouds crossings) is
542 considered in this analysis.

543 **4 Observations and results**

544 Figure 1 shows the flight tracks of the eight mission flights conducted during StratoClim 2017.
545 The vertical indices (Panel b) highlight the flight sections where significantly increased mixing
546 ratios of ultrafine particles n_{uf} were encountered, which are most likely attributed to NPF. NPF of
547 varying intensity occurred near or above the southern flank of Himalayan mountain chain
548 (features over Nepal and towards Northeast India) and in certain distance from the mountains
549 (near the coastline of Bangladesh or the Northeast Indian coast towards the Sea of Bengal). Of
550 the entire COPAS measurement time (~ 22.5 hours) at altitudes above 10 km (≥ 350 K potential
551 temperature) over almost one third (i.e. ~ 9 hours) of the air samples were taken north of 26° N,
552 i.e. mainly in the immediate vicinity of the Himalayan Mountains, over Nepal and neighbouring
553 areas of northeast India. Hence, during the monsoon season of the year 2017, the main transport
554 of NPF precursor material into the UT/LS was apparently by convection above the foothills of
555 the Himalayas. The present study aims at a classification of encountered NPF events with regard
556 to:

- 557 • the height intervals and geographical positions of NPF observations,
- 558 • the time limits (event duration and day time of occurrence),
- 559 • spatial dependencies with regard to tropopause height and AMA geometry.

560 Moreover, the relationship between NPF and the air's origin and age is investigated. It is
561 noteworthy that, during StratoClim 2017, NPF was frequently observed in the presence of ice
562 cloud particles at the bottom TTL of the AMA. The particular occurrence of in-cloud NPF is
563 discussed in Weigel et al. (2020b). Since the NPF turned out to be almost undisturbed by the
564 presence of cloud elements (until a certain density and size of the ice particles are reached), for
565 the present study the NPF encounters remain unseparated concerning clear-air or in-cloud
566 conditions and are instead discussed in their entirety.



567 **4.1 Vertical distribution of particle number concentrations with respect to**
568 **observations in the tropics**

569 Vertical profiles of the total particle number concentration obtained from various field
570 campaigns in the tropics are shown as median with percentiles in Figure 2. The vertical CN
571 profiles from tropical regions of South America and West Africa (TROCCINOX, 2005 and SCOUT-
572 AMMA, 2006, Figure 2, Panels a and b) exhibit merged data of two independent CN-detectors
573 with individual d_{p50} (i.e. N_6 for $\theta > 350$ K and N_4 for $\theta < 350$ K), which were deployed on
574 individual aircraft, the M-55 *Geophysica* and the DLR Falcon-20 (cf. Borrmann et al. (2010) and
575 Weigel et al. (2011)). The observations from measurements within the AMA over the Indian
576 subcontinent (StratoClim 2017) at altitudes of about $320 \text{ K} < \theta < 475 \text{ K}$ potential temperature
577 exclusively result from COPAS measurements aboard the M-55 *Geophysica*. The dark shaded
578 areas of the vertical profiles illustrate the scatter of number concentrations between the 90th
579 and 99th percentiles. At tropopause altitudes around 380 K (indicated by vertical bars), or rather
580 at the bottom TTL, the variability of detected concentration reaches a maximum between 90th
581 and 99th percentile (note the logarithmic scale of N). The increased data scatter indicates that the
582 population of sub-micrometre sized particles is subject to NPF occurring at these TTL levels,
583 resulting in elevated particle number concentrations, which are highly variable, as is the
584 production-rate of particles due to NPF (cf. Section 2.1.3). Exclusively above the tropopause
585 within the AMA (Figure 2 c), the scatter of the concentration values of sub-micrometre sized
586 particles remains elevated at up to ~ 400 K potential temperature. Up to this point, the dark
587 shaded area (90th to 99th percentile range) of the AMA profile is visibly increased compared to
588 median values, while aerosol concentrations measured above the tropopause in other regions
589 (Figure 2 a and b) exhibit a smoother transition into the stratosphere.

590 For comparison, in Panel d of Figure 2, particle number concentrations $N_{5,3}$ are compiled as a
591 vertical median profile (with percentiles) obtained from airborne measurements with the Nuclei
592 Mode Aerosol Spectrometer (NMASS; Brock et al. (2000)) during several years (2004 – 2007,
593 including winter and summer season) over Central America. These observations additionally
594 differentiate the bottom TTL (here 350 – 379 K) as the region where NPF predominantly occurs
595 with the largest impact on the fine-mode (sub-micrometre sized) aerosol particle concentration
596 (e.g. Borrmann et al. (2010) or Weigel et al. (2011)). However, this vertical profile (Figure 2 d)



597 implies additional features at altitudes above the mean tropopause altitude (assumedly at
598 ~ 380 K). The locally increased concentrations with respect to the median become apparent at
599 $\sim 380 - 390$ K and at $\sim 400 - 410$ K, respectively. Above tropopause levels, significantly
600 increased concentrations of fine-mode particles, potentially caused by local NPF, were observed
601 over both, Central America (Figure 2 d) and the Indian subcontinent within the AMA (Figure
602 2 c).

603 **4.2 Mixing ratios of submicron particles, abundance and fraction of refractory particles** 604 **from StratoClim 2017 observations**

605 The entire StratoClim 2017 data set of measured (1-Hz-resolved) particle mixing ratios n_6 and
606 n_{10} is summarized in Figure 3 a as function of potential temperature. The resulting median
607 profile n_6 of the StratoClim 2017 measurements is shown with 25th and 75th percentile (blue
608 profile). This allows for a direct comparison with the corresponding median profiles from earlier
609 COPAS measurements at tropical regions (in red: TROCCINOX, Brazil, 2005 and in dark green:
610 SCOUT-AMMA, West Africa, 2006, cf. Borrmann et al. (2010) and Weigel et al. (2011)). Figure 3 a
611 includes also the median vertical profile of the mixing ratios of fine-mode particles (bright green
612 line), which was obtained from measurements over Central Pacific, at tropical latitudes (Brock et
613 al., 1995).

614 The profiles (n_6 , n_{10} , and n_{ref}) appear to be structured as:

- 615 1) $\sim 350 - 380$ K: characterised by the largest scatter of the particle mixing ratios and the
616 highest values of up to $5 \cdot 10^4 \text{ mg}^{-1}$, thus, representing the height level of the profile's
617 maximum.
- 618 2) $\sim 380 - 400$ K: the scatter of the particle mixing ratios is still increased though less
619 expressed, which may also be a result of recent NPF.
- 620 3) $\sim 400 - 415$ K: characterised by a comparatively weak but extant scatter level of particle
621 mixing ratios, which also includes features of the median n_6 profile at $410 - 415$ K within
622 the AMA.

623 The general course of the median profiles exhibits largely similar characteristics. The common
624 feature of all median profiles from the tropics is their almost consistently located maximum at
625 about $350 - 360$ K, while the AMA observations indicate a corresponding maximum at slightly



626 higher altitudes (i.e. 355 – 365 K). Further up, the particle mixing ratios obtained from different
627 locations decrease with altitude with an almost corresponding gradient. In the altitude range
628 between 360 K and 400 K, the tropical data obtained over South America (red) constitute the
629 lowest particle mixing ratios (by median values), whereas all other profiles are almost in line
630 with each other up to 400 K. The vertical median profile of particle mixing ratios determined in
631 the AMA (blue) during StratoClim 2017 exhibit, however, the highest mixing ratios at each
632 height level up to ~ 415 K. Additionally, the AMA profile features a substantial increase of the
633 median mixing ratio at altitudes of ~ 410 - 415 K, where the values exceed those from the
634 tropical regions by almost 35 %. Above 415 K, the continuation of the tropical profiles from west
635 Africa and Central America (coloured green) with altitude is largely consistent with the particle
636 mixing ratios measured throughout StratoClim 2017, while at these altitudes the measurements
637 from South America (red) show comparatively increased values. Above 440 K, the particle
638 mixing ratio over West Africa (dark green) significantly deviates from those of all other vertical
639 profiles, as it is expressed by a gradual increase of the particle mixing ratio with altitude, which
640 was attributed to the influence of the high-reaching volcanic injections of Soufriere Hills
641 (Borrmann et al., 2010). The 1-Hz-resolved StratoClim 2017 data (grey dots in Figure 3 a)
642 additionally illustrate how the scatter of measured particle mixing ratios relates to
643 corresponding median profiles.

644 Figure 3 b shows the vertical distribution of the mixing ratio of the ultrafine particles n_{uf} (cf.
645 Subsection 2.1.1). The flight-by-flight colouration of the data points indicates that increased n_{uf}
646 values were observed during each of the eight StratoClim 2017 mission flights. In addition,
647 Figure 3 b shows the wide range of altitudes over which the layers of increased n_{uf} were
648 observed during the individual flights. Remarkably high values of n_{uf} were detected up to
649 altitudes as high as 400K.

650 Figure 3 c exhibits the 1-Hz-resolved mixing ratios of the non-volatile particles n_{10NV} (cf.
651 Subsection 2.1.2) as a function of the potential temperature. The graphic also includes the
652 resulting median profile of n_{10NV} with 25th and 75th percentiles. Figure 3 c additionally depicts
653 the median profile of n_6 as in Figure 3 a, to evaluate the vertical course of n_{10NV} in direct
654 relationship to the total particle mixing ratio. In Figure 3 d, the vertical distribution of the



655 fraction f is shown in 1-Hz-resolution as well as the resulting median profile with 25th and 75th
656 percentiles.

657 At lower altitudes (< 350 K), the mixing ratio of non-volatile particles appears predominantly
658 low with a relatively large scatter. At altitudes where n_6 exhibits the maximum particle mixing
659 ratio (i.e. $\sim 355 - 365$ K), the n_{10nv} profile almost stagnates or even decreases slightly. The local
660 minimum in the fraction f is reached at about the same height ($355 - 375$ K), as result of the
661 significantly increased total particle mixing ratio (likely due to NPF) with simultaneously
662 declining n_{10nv} . On transition to 370 K, the mixing ratio n_{10nv} is again slightly elevated, and
663 above 370 K, the n_{10nv} profile follows the general decline with height. Nevertheless, up to 380 K,
664 the decrease of n_6 with altitude is steeper compared to that of n_{10nv} . On transition to the 390 K
665 level, a sharp drop in the median f profile mainly results from the sudden change of the n_6
666 gradient at this altitude, whereas the n_{10nv} profile exhibits no obvious feature at the same
667 height. Above 390 K, both mixing ratios (n_6 and n_{10nv}) decrease uniformly and the fraction f
668 remains almost constant at $\sim 45 - 50$ % for the altitude range up to 430 K. Towards 435 K, the
669 total mixing ratio n_6 almost stagnates whereas n_{10nv} exhibits slightly dropping mixing ratios.
670 Thus, at this point, the shape of the median f profile is mainly determined by a decrease of the
671 non-volatile proportion of the particle population. Further above, in transition to 440 K potential
672 temperature, both mixing ratios (n_6 and n_{10nv}) commonly exhibit a steep decrease.

673 In essence, the vertical profiles of the total particle mixing ratio n_6 and those of the non-volatile
674 particles n_{10nv} are divided into three ranges:

675 A) At the bottom TTL region ($\theta < 375$ K), both n_6 and n_{10nv} seem to be mainly characterised
676 by NPF as indicated by the high mixing ratios of ultrafine particles n_{uf} . NPF causes a
677 significant addition to the scatter of the total mixing ratios towards high values, which
678 exceed the median by more than one order of magnitude. In this altitude range, a local
679 deficit of the non-volatile particle compounds is a favourable precondition for NPF to
680 occur.

681 B) Further above, i.e. ~ 375 K $< \theta < 415$ K, continued albeit attenuated NPF is identified at
682 tropopause levels within the AMA. The non-volatile particle compounds are slightly
683 elevated compared to levels below 375 K. The fraction f however rises towards 40 %.



684 Nevertheless, n_{uf} of 400 - 2000 mg^{-1} at heights of up to ~ 400 K indicate unimpededly
685 proceeding NPF.

686 C) Above 415 K, the values of the total mixing ratio n_6 approaches a course that
687 corresponds to previous observations (e.g. Brock et al. (1995)). The scatter of n_6 and
688 n_{10nv} is considerably decreased at these altitudes. NPF appears to have abated entirely,
689 since at these heights sufficiently high n_{uf} were not observed at all. The median
690 proportion f of non-volatile particles of $\sim 40 - 50$ % remains up to the highest altitude.

691 The steeply dropping vertical profile of the total mixing ratio of the sub-micrometre sized
692 aerosols above ~ 415 K may subtly indicate the upper limit of the AMA's influence on the
693 vertical mixing of the UT/LS. From the CO, ozone, and nitrous oxide content in air samples taken
694 throughout StratoClim 2017, von Hobe et al. (2020) concluded that the AMA's interior was
695 largely isolated from stratospheric in-mixing up to altitudes of 10 to 20 K above the tropopause
696 (i.e. $\theta \approx 400$ K). Moreover, they found that mixing processes with stratospheric air are of
697 increasing significance at levels between 400 K and 420 K (*ibid.*). At altitudes above $\theta \approx 440$ K,
698 the median mixing ratios n_6 exhibit a vertically stable continuation after another sharp drop
699 between 435 K and 440 K (Figure 3 a and b). Brunamonti et al. (2018) specified the 440 K level
700 as the top of confinement (TOC) of the AMA for the 2017 monsoon season, thus, above 440 K
701 potential temperature ($\gtrsim 18.5 - 19$ km), the median n_6 (Figure 3 a and b) may represent
702 stratospheric background values.

703 The ATAL (Vernier et al. (2011a), and see also Höpfner et al. (2019); Mahnke et al. (2020)) is
704 mainly attributed to the uplift of pollution from the boundary layer as concluded from balloon-
705 borne and satellite-based observations (Vernier et al., 2018). The described drop in the aerosol
706 concentration (*ibid.*) at potential temperatures of ~ 400 -420 K (well above tropopause levels)
707 coincides with the uppermost altitude limit of main NPF activity at ~ 400 K (~ 17.5 km)
708 observed during StratoClim 2017 (cf. Figure 3). Here, the most substantial decrease of both
709 mixing ratios n_6 and n_{10nv} was observed on height change from ~ 410 K to ~ 415 K (at ~ 18 km).

710 **4.3 Occurrence frequency of NPF events**

711 In compliance with the event definition (cf. Subsection 2.1.3), all observed NPF events (130
712 individual events) are sorted by their duration and the result is displayed in Figure 4. Based on
713 the average flight speed (Section 2.1.3), and assuming a constant heading during flight, the mean



714 horizontal distance per 10 seconds flight time ranges at about 1.5 km. The spatially most
715 extended uninterrupted NPF signature throughout StratoClim 2017 spanned a mean horizontal
716 distance of ~ 110 km. The majority of observed NPF events cover durations of several tens of
717 seconds or less. About 50 NPF events had very short durations of less than 10 s (Figure 4 a and
718 b), while an almost equal number of NPF events was observed over a continuous period of 10 –
719 55 s (~ 1.5 – 8.5 km; Figure 4 b). Longer lasting NPF events of about 40 – 80 s (~ 6 – 13 km)
720 occurred less than six times during the entire campaign period. All NPF events of even longer
721 duration (up to ~ 12 minutes) occurred mostly once, but never more than two times in total
722 throughout the mission period. The hitherto most extended NPF event observed with COPAS at
723 TTL level over South America (Weigel et al. (2011)) lasted over a continuous duration of
724 262 seconds (~ 35.5 km of covered flight distance). Another three individual NPF events were
725 observed above West Africa (*ibid.*) over 20, 83, and 98 seconds (~ 3 km, ~ 12 km, and ~ 13 km)
726 respectively. Approximately 45 % of 130 NPF events observed throughout StratoClim 2017
727 were of less than 20 seconds duration (~ 3 km), while the majority (~ 75 %) of NPF
728 observations above the Indian subcontinent extended over less than 80 seconds (~ 12 km).

729 The diurnal distribution of observed NPF events is exhibited in Figure 5. Initially, the frequency
730 of NPF event observations is analysed as a function of the local daytime (LT) at Kathmandu,
731 Nepal (Figure 5 a). Apart from one exception, the occurrence frequency of the NPF events seems
732 evenly distributed over the course of a day. The exception is a time window at about 10:00 a.m.
733 to 10:30 a.m. (LT) when recent particle formation was observed up to 2.5 times more often than
734 at other times of the day. In this time window, about one third of all NPF events (31 of 105
735 events with durations of more than 5 seconds) was observed, most of which (25 of 31 events)
736 lasted for less than 80 seconds (< 12 km mean horizontal distance). The measurements in this
737 time window occurred at two distinct altitude layers, ~ 360 - 370K and ~ 390 – 400 K. The
738 majority of the NPF events in this period (20 of 31 events) were from altitudes above 390 K
739 while ascertained mixing ratios $\overline{n_{uf}}$ never ranged outside ~ 500 - 5000 mg^{-1} during this day
740 time. Throughout the StratoClim 2017 mission, no further NPF event was observed above 390 K
741 at any earlier day time and only two single events were encountered at these heights during
742 different flights at a later day time ($\sim 12:20$ and $\sim 17:30$ LT, respectively). Hence, it is very likely
743 that this pronounced frequency of NPF occurrence at a certain time of day results from biasing
744 effects and that instead the diurnal distribution of NPF events is in fact more evenly distributed.



745 In addition, however, a preferred daytime was not identified at which NPF observations would
746 have occurred with particular frequency. This would be expected if H_2SO_4 is assumed to be the
747 main nucleating compound whose production maximum (from the reaction $\text{SO}_2 + \text{OH}$) at local
748 noon-time correlates with the solar zenith (cf. Weigel et al. (2011)).

749 According to the flight-by-flight distribution of NPF events over the day (Figure 5 b), the
750 impression could arise that NPF events were observed more frequently in the morning than at
751 local noontime. The observations of events in the afternoon are widely distributed over several
752 hours, compared to the morning. NPF was predominantly observed before local noontime
753 during the mission flights KTM 2, KTM 3, KTM 5 and KTM 7, while all other observations were
754 made mainly during the afternoon. The diurnal distribution of the events as a function of event
755 duration (Figure 5 c) indicates that the number of very short events (25 events) over less than
756 five seconds (i.e. $< 10^{0.7}$ s on the logarithmic scale, blue data points) remains below 20 % of all
757 encountered events (130 events) and predominantly occurred in the morning hours. The
758 majority of data points stem from event encounters, which lasted from 5 to 60 seconds ($\sim 10^{0.7}$ -
759 $10^{1.8}$ s, greenish colours). Such durations of NPF encounters correspond to mean horizontal
760 distances of about one to ten kilometres. These events, as also all longer lasting events, were
761 almost homogeneously distributed over the day. Furthermore, Figure 5 c may also indicate that
762 the longest NPF events are not necessarily associated with highest mean mixing ratios $\overline{n_{\text{nr}}}$. The
763 duration of an event is therefore primarily an indicator of the spatial extent of a region where
764 NPF takes place. The derivation of the spatial extent from the duration of individual events,
765 however, bears significant uncertainties, since changes in flight attitude, such as curve
766 manoeuvres or changing flight levels during an event, are not taken into account.

767 However, the NPF events observed during StratoClim 2017 are among the most frequent and
768 spatially most extended of all those, which have been identified by means of COPAS
769 measurements during previous missions (cf. Borrmann et al. (2010); Weigel et al. (2011)). Only
770 a few events were observed during StratoClim 2017, which lasted more than 100 seconds, but it
771 cannot be excluded that they were actually composed of individual events of smaller extent.
772 Very short events (< 10 s) make almost 40 % of all NPF events observed. Consequently,
773 hereafter, all events shorter than five seconds (i.e. 25 out of 130 events) are discarded from
774 further analyses for the reasons described in Subsection 2.1.3 and for avoiding biasing effects.



775 4.4 The occurrence of NPF relative to the tropopause height and the AMA's centre

776 ERA-interim reanalysis data were used to determine the altitude of the lapse-rate tropopause in
777 accordance with the definition by the WMO (1957) for each measurement point along the flight
778 path (cf. Section 3.1). For the individual NPF events, a mean tropopause height was obtained
779 together with mean values of detected $\overline{n_{uf}}$. The relationship between the measurement height
780 and the lapse-rate tropopause height is expressed as difference $\Delta\theta$ in K. In addition to the
781 vertical position of NPF events (i.e. in terms of absolute height or distance from the tropopause),
782 the individual NPF events were examined with respect to their position within the AMA by
783 means of the equivalent latitudes ϕ_{equ} (cf. Section 3.1).

784 Figure 6 illustrates the mean n_{uf} measured during the individual NPF events as a function of (1)
785 the distance $\overline{\Delta\theta}$ to the lapse rate tropopause (Figure 6, Panels a and c) and (2) the mean
786 equivalent latitude $\overline{\phi_{equ}}$ (Figure 6, Panels b, d and e). NPF events above the lapse-rate
787 tropopause (Figure 6 a, positive $\overline{\Delta\theta}$) were mainly observed during the first half of the
788 StratoClim 2017 mission (KTM 2, KTM 3, and KTM 5; on 29 July, 31 July, and on 04 August 2017,
789 respectively) or during the last mission flight (KTM 8, on 10 August 2017). All further
790 observations were located below the lapse-rate tropopause (negative $\overline{\Delta\theta}$) or in its close vicinity
791 ($\overline{\Delta\theta} \approx 0$ K, e.g. KTM 6, 06 August 2017). As indicated by Stroh et al. (2020), the first half of the
792 StratoClim 2017 mission was characterised by weak convection, while in succession of the
793 campaign the convective activity increased. NPF event observations throughout StratoClim 2017
794 were limited to an altitude interval between about -35 K and $+30$ K potential temperature
795 around tropopause heights, corresponding to a pressure range of 70 – 340 hPa and ambient
796 temperatures between 187 K and 257 K according to observational data. With respect to the
797 AMA centre, most NPF events were encountered north of 60° equivalent latitude (Figure 6 b). An
798 exception is a flight segment of flight KTM 3 (on 31 July 2017), where NPF with mixing ratios $\overline{n_{uf}}$
799 of $\sim 500 - 1300 \text{ mg}^{-1}$ were detected at the farthest distance from the AMA centre (near the
800 turning point at about 21.5° N and 80° E geographic coordinates, see Figure 1).

801 These measurements (at $\overline{\phi_{equ}} < 60^\circ$ N) were made well above the tropopause since mean CO
802 mixing ratios of $45 - 50 \text{ nmol mol}^{-1}$ (Figure 6, Panels c and d) are commonly found at $\overline{\Delta\theta}$
803 between $+5$ K and $+10$ K. Towards the AMA centre ($\overline{\phi_{equ}} > 60^\circ$ N), the NPF events are
804 distributed over the entire range of $\overline{\Delta\theta}$. Here, NPF with several hundreds of ultrafine particles



805 per milligram were observed at $\overline{\Delta\theta}$ of up to about + 28 K above the lapse-rate tropopause. The
806 vertical distribution of the NPF events indicates that those events with the highest $\overline{n_{\text{uf}}}$ and
807 mainly elevated CO mixing ratios (mean values of more than 65 nmol mol⁻¹ and up to
808 ~ 137 nmol mol⁻¹) were encountered exclusively below the lapse-rate tropopause (to minimum
809 $\overline{\Delta\theta}$ of - 35 K). However, none of these NPF events with elevated n_{uf} was detected at equivalent
810 latitudes $\overline{\phi_{\text{equ}}} < 60^\circ\text{N}$. Between 60°N and 90°N equivalent latitude, however, no indication is
811 apparent that the mixing ratios of $\overline{n_{\text{uf}}}$ and CO depend on the position with respect to the AMA
812 centre.

813 Panel e of Figure 6 shows the NPF event distribution in the combined coordinate space of the
814 equivalent latitude and the vertical distance from the lapse-rate tropopause. NPF events with
815 highest $\overline{n_{\text{uf}}}$ (≥ 1300 mg⁻¹) were found exclusively between 60° N and 90° N with respect to the
816 AMA centre and often immediately underneath the lapse-rate tropopause ($\overline{\Delta\theta} \lesssim - 3$ K, colour-
817 range from blue to yellow). NPF events with low $\overline{n_{\text{uf}}}$ ($\lesssim 1300$ mg⁻¹) were exclusively found at
818 tropopause levels ($\overline{\Delta\theta} \approx 0 \pm 5$ K, orange colours) or well above the lapse-rate tropopause
819 ($\overline{\Delta\theta} > 10$ K, red data points). Close to the AMA centre (60°N - 90° N) and in an altitude range of
820 almost ± 30 K around tropopause heights, both the distribution of CO-enriched air masses and
821 the occurrence of NPF appear as largely independent from $\overline{\phi_{\text{equ}}}$.

822 **4.5 Persistence of particles in the ultrafine size mode**

823 Coagulation constitutes one of the main processes, which limits the persistence of ultrafine
824 particles. Due to the high diffusivity of the ultrafine particles, especially at elevated number
825 densities, the particles collide and coagulate amongst each other and with the present
826 background aerosol particles on short time scales. Gaseous precursors, which are highly
827 saturated under NPF conditions, may condense and additionally contribute to the growth of
828 particles out of the ultrafine size range, which is considered hereafter, however, as a secondary
829 process.

830 The aerosol size distribution, which was compiled from the measurements during a NPF event
831 as input for the coagulation simulation (cf. Section 3.2), is depicted in Figure 7 (black circles with
832 horizontal bars indicating the width of their respective particle size bins of the model). The
833 simulated change of the initial aerosol size distribution due to coagulation is shown in 1-hour



834 steps in different colours and line types (Figure 7 a). From this simulation, the temporal decay of
835 N_{uf} was derived (Figure 7 b, solid black line), whereby the gradient of this decay illustrates the
836 coagulation rate. The sequence of the simulated size distributions indicates that the initial
837 amount of ultrafine particles is reduced by coagulation within a few hours. Coagulation is most
838 effective particularly in the particle size range of $d_p < 15$ nm. Within the first hour after an
839 expired NPF event the ultrafine particle mode is no longer predominant in the overall size
840 distribution, as seen from the maximum of the distribution at $d_p > 15$ nm after one hour of
841 simulated coagulation (solid red line in Figure 7 a). Hence, based on number concentrations of
842 particles in the ultrafine size mode, a clear signature of NPF is detectable only when a NPF event
843 is just proceeding or for a very short time right after an expired NPF event.

844 The concentration of ultrafine particles N_{uf} decreases steeply over time (Figure 7 b). From
845 initially ~ 13000 cm⁻³ of ultrafine particles (~ 75 % of N_{total}) at the earliest stage, N_{uf} falls below
846 1000 cm⁻³ (~ 20 % of N_{total}) within about 1 hour (the grey shaded areas may serve for reference).
847 The detection of 1000 cm⁻³ of ultrafine particles, however, could be interpreted as a NPF event of
848 intermediate strength. In addition, coagulation leads to N_{uf} below 100 cm⁻³ (< 5 % of N_{total})
849 during less than four hours, which significantly impedes the identification of NPF based on in-
850 situ detections. N_{uf} of less than 10 cm⁻³ (reached within nine hours) would not be identified as
851 NPF event by means of COPAS measurements.

852 The sensitivity of this simulation was investigated by varying the simulation input. Therefore,
853 exclusively the input in the ultrafine size range was modified while keeping constant
854 background aerosol conditions. In three further simulation runs, the initial N_{uf} was multiplied by
855 the factors 0.1, 10 and 100, respectively ($N_{uf,0.1}$, $N_{uf,10}$, $N_{uf,100}$, dashed lines in Figure 7 b).
856 Increased initial concentrations of ultrafine particles, $N_{uf,10}$ and $N_{uf,100}$, last only for about 15
857 minutes compared to the original N_{uf} (black line in Figure 7 b). The initial values $\sim 10^5$ or
858 $\sim 10^6$ cm⁻³ drop very quickly due to elevated coagulation rates, and in both of these cases, $N_{uf,10}$
859 and $N_{uf,100}$ fall below 1000 cm⁻³ within less than one hour. The threshold of 100 cm⁻³ is crossed
860 after less than 2 hours ($N_{uf,10}$) or after 30 minutes ($N_{uf,100}$). Therefore, NPF events, which
861 produce much higher concentrations of ultrafine particles, require even shorter time periods for
862 a successful detection (e.g. by COPAS) after their expiration. However, for the simulation of
863 decreased concentrations ($N_{uf,0.1}$), the coagulation rates remain nearly constant, as indicated



864 from the almost identical decays of $N_{uf,0.1}$ and N_{uf} (Figure 7 b). Simulated concentration of
865 ultrafine particles fall below 100 cm^{-3} within almost the same time from the initial values $N_{uf,0.1}$
866 or $N_{uf,10}$, respectively.

867 Based on these estimations, the detection of elevated N_{uf} strongly indicates that an event with
868 high NPF-rates is currently proceeding, or a recently expired NPF event was observed. Due to
869 the short persistence of ultrafine particles (a few hours), the observations of events with
870 elevated N_{uf} are considered as made "well in time". Detections of lower values of N_{uf} could
871 indicate a) intermediate or weak (currently proceeding) NPF at low supersaturation of the NPF
872 precursor or b) a NPF event (potentially of high particle productivity) that has phased-out
873 several hours before the observation. NPF is measured *in-situ* while the formation event is
874 currently in progress or at most a few hours later. Therefore, the short periods of time available
875 for a clear NPF detection and the yet frequent NPF encounters on each measurement flight
876 during StratoClim 2017 indicate the prevalence of such events within the AMA.

877 5 NPF's connection to ground sources and vertical transport

878 5.1 NPF in relationship to CO as pollution indicator

879 NPF events with moderate numbers of ultrafine particles ($< 1000 \text{ cm}^{-3}$) in the lower TTL region
880 were previously attributed to CO mixing ratios above $\sim 70 \text{ nmol mol}^{-1}$ (as a reference: $60 -$
881 70 nmol mol^{-1} were assumed as a typical CO background in the pristine marine boundary layer,
882 cf. Weigel et al. (2011)). Elevated amounts of ultrafine particles (of up to $\sim 6000 \text{ cm}^{-3}$) at
883 altitudes of $350 \text{ K} < \theta < 360 \text{ K}$ were associated with significantly increased CO mixing ratios of
884 more than 85 nmol mol^{-1} (*ibid.*). These results, based mainly on two individual NPF events over
885 West Africa (SCOUT AMMA 2008), may suggest a relationship between NPF with high densities
886 of ultrafine particles and high CO mixing ratios and thus, a high level of pollution. However, as
887 already indicated in Section 4.4 and Figure 6 (Panels c and d) by almost a hundred of individual
888 event observations, the relationship between pollution level and NPF-induced n_{uf} is less direct
889 than expected. In Figure 8, the 1-Hz-resolved data of synchronous detections of CO and particle
890 mixing ratio during the entire StratoClim 2017 mission are compared. The total particle mixing
891 ratio n_6 is shown in the background (grey dots) of the mixing ratio of ultrafine particles n_{uf} ,
892 (coloured data points in reference to θ) to illustrate the scatter range of both n_6 and of n_{uf} .



893 At altitudes below the tropopause (below ~ 380 K), where NPF causes the highest n_{uf} , the
894 relationship between the 1-Hz-resolved n_6 or n_{uf} and the CO mixing ratio is highly variable. At CO
895 levels of 80 – 100 nmol mol⁻¹, the scatter of the data covers the n_{uf} range from 700 mg⁻¹ to the
896 absolute maximum of about 50000 mg⁻¹. This maximum n_{uf} is exclusively reached at CO mixing
897 ratios of 100 ± 2.5 nmol mol⁻¹. This is in qualitative agreement with earlier results from NPF
898 detections at the bottom TTL over West Africa (Weigel et al., 2011) with a maximum of ultrafine
899 particles at CO mixing ratios of up to 95 nmol mol⁻¹. However, compared to these results, the
900 StratoClim 2017 data set is significantly extended towards much higher CO mixing ratios (up to
901 150 nmol mol⁻¹) coinciding with elevated but variable n_{uf} . At the maximum of the CO mixing ratio
902 (i.e. ~ 150 nmol mol⁻¹) mixing ratios n_{uf} of about 6000 mg⁻¹ (median value) were detected.
903 Within a range of CO content between 85 and 130 nmol mol⁻¹, the n_{uf} (median) mixing ratios
904 ranged consistently between 2000 and 10000 mg⁻¹, apart from the notable exception at about
905 100 nmol mol⁻¹.

906 At CO mixing ratios below 80 nmol mol⁻¹ and on decrease to about 60 nmol mol⁻¹, the
907 measurements were made just below or at tropopause levels (yellow to orange colours). Here, a
908 decrease of n_{uf} from about 3000 mg⁻¹ to values below 1000 mg⁻¹ was observed. For CO mixing
909 ratios below 60 nmol mol⁻¹, however, n_{uf} almost stagnates between 300 and 1300 mg⁻¹. Towards
910 and across tropopause levels, n_{uf} and CO mixing ratios supposedly follow a systematic trend,
911 which is not necessarily due to a correlation. On the one hand, the convective transport of CO
912 from the ground does not directly reach tropopause levels or altitudes above the tropopause (cf.
913 also Figure 6 c). On the other hand, on uplift further above 370 K potential temperature, CO is
914 increasingly subject to depletion (cf. Section 2.3 and also von Hobe et al. (2020)). The NPF
915 process likewise depends on the supply by NPF precursors, which also hardly reach up to
916 heights above the tropopause by direct transport. Hence, at tropopause levels and aloft, the
917 decreasing CO mixing ratio as well as abating NPF (expressed in decreasing n_{uf} values) very
918 likely result from the lacking supply of precursor material by direct transport. According to von
919 Hobe et al. (2020) any indication is missing that convection penetrated the tropopause during
920 the StratoClim 2017 period. However, Lee et al. (2019) are investigate the TTL-hydrating
921 influence of an overshooting event that occurred in the Sichuan Basin about 1.5 days before the
922 StratoClim measurements southbound of Kathmandu over northeast India (M-55 *Geophysica*,
923 KTM 7 on 8 August, 2017).



924 Hence, there is no clear indication for a direct relationship between CO enriched (polluted) air
925 and the intensity of NPF. Both CO and NPF precursors may commonly be transported by
926 convection and deposited in the lower TTL region. Therefore, both the highest CO and n_{uf} mixing
927 ratios are detectable within the lower TTL, but not coincidentally. However, the comparatively
928 short atmospheric persistence of particles in the ultrafine size mode requires particular
929 consideration for this evaluation. Ultrafine particles in such high n_{uf} mixing ratios ($> 10000 \text{ mg}^{-1}$
930 and up to 50000 mg^{-1}) have a persistence of hours in the atmosphere (cf. Section 4.5). Thus, on
931 observation of such significantly elevated n_{uf} , the NPF must either be in full progress or must
932 have happened within a very few hours prior to the measurement. If CO mixing ratios (as
933 indicator of the recent uplift of polluted air) had a direct impact on the NPF-rate, then the
934 detections of elevated n_{uf} should coincide with correspondingly high CO mixing ratios.
935 Conversely, however, high CO content does not necessarily imply strong NPF, as the limited time
936 the particles persist in the ultrafine size mode does not allow for distinguishing a currently
937 proceeding event of moderate NPF-rate from a strong NPF burst, which has occurred hours ago
938 (cf. Section 4.5).

939 **5.2 NPF and air mass origin in the boundary layer**

940 The apportioning of specific measurement sections, which feature elevated n_{uf} , to the locations
941 on the ground as possible precursor source regions is carried out in two steps:

942 (1) The backward trajectories were traced down to the boundary layer (BL) for each
943 measurement point (cf. Section 3.3) at which NPF was detected according to the passed criterion
944 (Figure 9 a and b).

945 (2) The ERA-5 reanalysis data were examined with regard to the transport time of the
946 trajectories between the position in the BL and the coordinates of the measurement point (Fig
947 9 c and d).

948 The top two panels of Figure 9 (a and c) illustrate the geographic position of the air's last BL
949 contact prior to the observations (1-Hz resolved) of elevated n_{uf} ($\geq 300 \text{ mg}^{-1}$) throughout the
950 entire StratoClim 2017 mission. The bottom panels of Figure 9 (b and d) show the geographical
951 coordinates where the air mass experienced the fastest uplift in its transport history towards
952 each point of n_{uf} detection. Of course, the accuracy of the individual coordinates should not be



953 overestimated, for the reasons described in Section 3.3 and since the local resolutions of the
954 observational data from in-situ measurements and of the reanalysis data are not equivalent.
955 Particularly the spatial resolution of the reanalysis data is vertically variable. However, the
956 panels of Figure 9 convey two aspects: (1) how widespread the distribution of the air masses
957 origins is within the BL, from where an influence on the composition of the air samples could
958 have occurred, and (2) in which geographical region the high-reaching convection has efficiently
959 lifted the material to the level of air sampling. The numerous NPF observations and the currently
960 highest resolution level of the ERA-5 data set should allow for identifying a systematic
961 relationship, if existing, between the observed NPF and the trajectories' contact to the BL.

962 According to the distribution of the trajectories' latest BL contact (Figure 9 a) and the maximum
963 vertical updraught (Figure 9 b) with reference to the n_{uf} mixing ratio, hardly any systematic
964 structure is visible. The possible source regions are distributed over the entire monsoon region
965 almost independently of the NPF intensity. Some of the trajectories' last BL contact point to
966 locations far away from the monsoon region (e.g. in the West: the east coast of Africa and the
967 Gulf of Aden; in the East: Indochina, the South China Sea and as far as the Philippine Sea). The
968 entire possible source area of NPF precursors ranges from the north of India and the Arabian
969 Sea, Pakistan, Afghanistan, Southwest China, Taiwan, the Philippines and the Bay of Bengal.
970 South of $\sim 10^\circ$ N geographic latitude, the number of possible source regions decreases
971 significantly. Locations of strongest vertical updraught are more compactly distributed (Figure
972 9 b) and better reflect the contours of the monsoon region. Hence, for the duration of the
973 StratoClim 2017 mission, the convective uplift may largely have occurred within the AMA. This
974 more compact regional distribution of vertical uplift is possibly related to the occurrence of a
975 *vertical conduit* for upward transport in the monsoon, as conjectured by Bergman et al. (2013).
976 Nevertheless, the almost homogeneous distribution of the n_{uf} mixing ratios within the displayed
977 region of strongest convective uplift does not allow for identifying specific locations as potential
978 source regions of NPF precursors. Furthermore, Figure 9 b indicates air masses of elevated n_{uf} ,
979 which have experienced convective uplift over Tajikistan and northern Afghanistan as well as
980 over regions around the Yellow Sea, the Korean Peninsula or Japan, hence, far away from the
981 AMA system.



982 With regard to the air mass transport time from the BL, the ERA-5 reanalysis data were
983 examined over 50 days prior to the in-situ measurements (Section 3.3). For transport times
984 exceeding 25 days, however, the data points in the Panels c and d of Figure 9 are displayed in
985 grey. According to Figure 9 c, the air masses with the shortest transport times from the BL are
986 found compactly around the region of the Himalayan mountain chain and its foothills. In Figure
987 9 c and d, the contour of the Himalayan chain is clearly reflected by the distribution of the data
988 points (transport times of less than ~ 5 days and fastest vertical updraught). The highest n_{uf}
989 mixing ratios were not detected in air from this region (Figure 9 a). The distribution shown in
990 Figure 9 c also indicates that in air masses from remote locations (Gulf of Aden, Arabian Sea; or
991 Philippine Sea, South China Sea, Bay of Bengal) also strongly elevated n_{uf} ($> 10^4 \text{ mg}^{-1}$) were
992 detected after comparatively long transport times of up to 25 days. Several other cases of
993 elevated n_{uf} are visible in Figure 9 a, for which the transport times from the BL even exceeded 25
994 days (grey points in Figure 9 c). The Figure 9 d finally shows that the region of the air's last BL
995 contact and the location of the fastest vertical uplift do not necessarily coincide. Hence, it may be
996 surmised that, within the free troposphere, the air is subject to loading from various source
997 regions (not exclusively from the location of the last BL contact) prior to its convective uplift. Of
998 course, this finding may complicate an unambiguous apportioning of NPF to specific source
999 regions of precursors in the BL.

1000 The vertical distribution of the n_{uf} mixing ratios as a function of the air mass transport time from
1001 the BL is shown in Figure 10:

1002 1) Above 380 K, almost all observations of enhanced n_{uf} are associated with air mass
1003 transport times of more than 12 days (note that the evaluation of the reanalysis covers up to
1004 50 days of transport time in total). At $380 \pm 3 \text{ K}$, none of the detected n_{uf} is connected to air mass
1005 transport times of less than 12 days. Several times higher n_{uf} (with 10^3 - 10^4 mg^{-1}) were detected
1006 below 380 K in air masses, which had experienced more than 25 days of transport time from the
1007 BL.

1008 2) Below 380 K, the transport times are variably distributed over the altitude range
1009 between 350 K and 380 K. The air masses with shortest transport times are located in the height
1010 interval between 360 K and 370 K. These air masses have presumably reached the $\sim 360 \text{ K}$ level
1011 (altitude of the main convective outflow) very quickly by an effective convective transport and



1012 are then moved further aloft, towards 370 K, with much lower ascent rates (Vogel et al., 2019)
1013 due to the prevailing air mass uplift within the AMA.

1014 3) On occasion, very short transport times were found with maximum n_{uf} at altitudes of
1015 about 367 K and 370 K. However, the highest n_{uf} are mostly not observed in air with such short
1016 transport times. Within 370 ± 3 K, detected n_{uf} reach almost extreme values ($\sim 50000 \text{ mg}^{-1}$) in
1017 air with transport times of up to 15 days. Above 370 K and below 355 K none of the maximum
1018 n_{uf} is associated with transport times of less than 6 days, and here, the highest n_{uf} were detected
1019 in air with transport times of up to 25 days. Therefore, based on the observations and the
1020 trajectories analysed here, the altitude band of the main convective outflow is limited to a range
1021 between 355 – 370 K.

1022 **5.3 The relationship between NPF and convective outflow**

1023 For the following analysis, which is summarised in Figure 11, the vertical distribution of the
1024 mean mixing ratios $\overline{n_{uf}}$ of respective NPF events (cf. Sections 2.1.3 and 4.4) are juxtaposed with

- 1025 a) a measure for the convective contribution to the composition of the probed air mass and
- 1026 b) the mean transport time within the TTL since their release from the top of individual
1027 convective cells (cf. Section 3.4 for both variables).

1028 According to Figure 11 a, two vertical sections may be separated: At altitudes above ~ 380 K, the
1029 $\overline{n_{uf}}$ values remain below 2000 mg^{-1} , while the convective contribution to the probed air mostly
1030 remains below 35 %. At altitudes below ~ 380 K, most of the observed events exceeded $\overline{n_{uf}}$
1031 values of 2000 mg^{-1} . About two thirds of all events that occurred below 380 K are connected to
1032 convective influence by more than 75 %. However, a remarkable proportion of observations
1033 below 380 K indicate convective contributions of less than 60 % and down to 25 %. Below
1034 ~ 375 K, mean mixing ratios $\overline{n_{uf}}$ of $1000\text{-}2000 \text{ mg}^{-1}$ were associated with 100 % convective
1035 contribution, and mixing ratios of more than 10000 mg^{-1} were sometimes observed in air masses
1036 with ~ 30 % convective contribution.

1037 For the observed NPF events, Figure 11 b shows the mean age of the probed air masses since
1038 their release from the top of individual convective cells. Above ~ 380 K, the air escaped at the
1039 convection top mainly 12 days (or more) prior to its probing. Two events at ~ 382 K and at
1040 ~ 385 K, respectively (e.g. likely connected to overshooting convection), indicate a more recent



1041 convective uplift, within 5 days before the air was sampled. Despite the comparatively short
1042 transport times, here, the observed $\overline{n_{uf}}$ remained below 2000 mg⁻¹. At altitudes below ~ 380 K,
1043 the air predominantly resided within the TTL region for less than 5 days prior to the
1044 observation. Nevertheless, some of the comparatively intensive NPF events (with $\overline{n_{uf}} \approx 7000 -$
1045 15000 mg⁻¹ at ~ 373 K, ~ 365 K, and at ~ 360 K) were also observed in air, which has been
1046 released from associated clouds' top more than a week (and up to two weeks) prior to the
1047 measurements. It should be considered, however, that short air mass transport times within the
1048 TTL are indicated also for NPF events with minor convective contribution (< 50 %).

1049 Despite the limited data base, it can be concluded from these analyses that NPF at the lower TTL
1050 (i.e. below tropopause) within the AMA occurs in air masses that have been raised, even if only
1051 partially, by convection within the last 5 days to about two weeks. It remains undetermined,
1052 however, whether in some of the observed events the air samples were taken at a very advanced
1053 stage of NPF or how often the short period of time was missed during which NPF is detectable by
1054 aircraft-based measurements. Potential uncertainties remain to be considered in connection
1055 with the uncertainty of the reanalysis data and the representation of the transport history of the
1056 air masses.

1057 **6 Potential impact of gravity waves on vapors' supersaturation**

1058 The NPF precursor substances may primarily originate from sources on the ground and in the
1059 BL. The convective uplift doubtlessly constitutes one of the most effective transport mechanisms
1060 for lifting the material to altitudes of the lower TTL. As the prerequisite for NPF to take place
1061 within the TTL, however, the gaseous precursor material

1062 a) must survive the convective uplift and must be released in sufficient quantities in the outflow
1063 region and at the lower TTL, and

1064 b) in the TTL region, the NPF precursor material is required to be enriched to sufficient
1065 supersaturation.

1066 If the uplifted precursor material was suitable for NPF immediately after its release from the
1067 convective outflow, the relationship between elevated n_{uf} and convective transport should be
1068 clearer than observed (cf. Section 5). The lack of an unambiguous relationship may indicate that



1069 the recently transported material has yet to be converted into a NPF precursor within the
1070 UT/LS. Such a process would be, for example, the conversion of SO_2 to H_2SO_4 , which occurs at
1071 stratospheric altitudes (Kremser et al., 2016) from tropospheric sulphur species, mainly OCS
1072 and others (cf. Section 1). The presence of ammonium species (Höpfner et al., 2019) or organics
1073 could then promote the NPF of H_2SO_4 in the TTL even at low supersaturations (Metzger et al.
1074 (2010); Kerminen et al. (2010); Kirkby et al. (2011); Kürten (2019)). The *in-situ* measurements
1075 with the aerosol mass spectrometer (AMS), the ERICA-AMS (ERICA, ERc instrument
1076 for the chemical composition of aerosols) during StratoClim 2017 showed that aerosols at the
1077 tropopause levels contain elevated amounts of nitrate (Höpfner et al., 2019). According to these
1078 observations, the nitrate concentration in the particle phase was higher than the particles'
1079 sulphate content at altitudes below 380 K. Nitric acid (HNO_3) is oxidised within a few days from
1080 NO_x , which may be partly transported out of the BL or partly generated *in-situ* by lightning
1081 (Huntrieser et al., 2016). The presence of HNO_3 , combined with ammonia may enhance the gas
1082 to growth of new particles (Wang et al., 2020).

1083 The supersaturation required for initiating NPF could temporally also result from local cooling.
1084 It is conceivable that the NPF process and its intensity is locally triggered, e.g., by gravity waves.
1085 Gravity waves (GWs) represent low-frequency inertial perturbations of the initial atmospheric
1086 state. Such a perturbation is expressed particularly by a change in velocity of the vertical wind
1087 component. The passage of a GW is associated with a change in the vertical displacement of an
1088 air parcel and thus causes locally an adiabatic heating/cooling by a certain absolute value ΔT .

1089 Piani et al. (2000) provided simulations of GWs initiated by deep convections. Their studies
1090 reveal a concentric propagation of GWs at altitudes above 15 km and up to ~ 40 km, which was
1091 effected by convective systems underneath. Horizontal wavelengths of about 40 km and vertical
1092 wavelengths of $\sim 4 - 7$ km were ascertained (*ibid.*). Similar wavelengths were found to be typical
1093 by other simulation studies concerning the propagation of GWs, which have been initiated, e.g.,
1094 by convection at mid-latitudes (Song et al. (2003) and Chun and Kim (2008)) or by tropical
1095 convection (Lane and Moncrieff, 2008). Investigations related to GWs in connection with the
1096 monsoon are sparse, e.g. Wright and Gille (2011) and Ern and Preusse (2012) used satellite
1097 observations (High Resolution Dynamics Limb Sounder) which, however, are limited to
1098 detections of GWs with horizontal wavelengths greater than ~ 300 km. Despite the numerous



1099 observational studies concerning GW properties (Alexander et al., 2010), the indirect retrieval of
1100 GWs' horizontal wavelengths remains uncertain by a factor of two (or more), whereas
1101 instrumental limitations inhibit the GW detection at horizontal wavelengths smaller than
1102 100 km.

1103 Observations using data sampled on commercial aircraft (Fritts and Nastrom, 1992) reported
1104 $\sim 1 \text{ K}^2$ temperature variances (or rather variance enhancements by a factor 6.1 compared to
1105 undisturbed conditions) on passages of convection-induced GWs through the tropopause region.
1106 Based on radiosonde measurements (Vincent and Alexander, 2000), a 6-year averaged
1107 amplitude of 1.5 K is reported as an effect of GWs, with a single-case example of $\sim 4 \text{ K}$ -amplitude
1108 around 20 km altitude in the tropics. Hence, GW-induced temperature anomalies are observable
1109 up to a maximum ΔT of $\sim 4 \text{ K}$, although smaller-scale perturbations occur more frequently. GW-
1110 induced negative temperature anomalies from an initial state T_0 increase a precursor's
1111 saturation ratio and the effect of such an anomaly on the degree of supersaturation applies
1112 qualitatively to any gaseous substance. Also in the homogeneous heteromolecular nucleation of
1113 more complex systems, the respective gas species or gas mixtures convert into particles as soon
1114 as the supersaturation exceeds the level required for the occurrence of NPF. Such more complex
1115 systems of precursor substances, e.g. with ammonia, nitric acid and/or organic components, are
1116 most likely involved in the formation process and promote NPF (Kirkby et al. (2011); Kürten
1117 (2019); Wang et al. (2020)).

1118 Satellite images over the Indian subcontinent (e.g. from MSG-1 or HIMAWARI, cf.
1119 <https://www.eorc.jaxa.jp/ptree/index.html>) indicate quite frequent occurrences of convective
1120 plumes in the sampling areas and during the StratoClim 2017 mission period, which occasionally
1121 even arranged in chains of convective cells along the Himalayan foothills. The StratoClim flight
1122 KTM 6 on 06 August 2017 enabled NPF observations immediately connected to convection,
1123 which shot-through the flight level on passage at constant flight altitude. Corresponding part of
1124 the time series shown in Figure 12 covers the probing period in the air sector over Bangladesh
1125 and the Bay of Bengal (cf. Figure 1). Two phases of NPF observations are highlighted (oblique
1126 hatched areas in Figure 12), immediately before and after the period between 09:20 and 09:30
1127 (UTC), during which the flight altitude changed from 16.2 km to about 13.8 km with subsequent
1128 re-ascent to 16.2 km. The manoeuvre above the northern part of the Bay of Bengal also marks
1129 the turning point of the mission flight path and the two flanking NPF phases were encountered



1130 over the mainland near the coastlines of East India and Bangladesh (cf. Figure 1 b). The
1131 outbound and return sections of the flight passed through the same convectively active region,
1132 and the same convective system was likely probed at opposite positions.

1133 Within the limits of the displayed time series (Figure 12 a) constant flight altitude and pressure
1134 level were maintained, except for the turning manoeuvre, which is therefore not subject of
1135 following discussion. The mixing ratios n_6 , n_{10} and n_{15} coincidentally exhibited a clear positive
1136 signal of variable strength (Figure 12 b). The productivity of observed NPF events is derivable
1137 from the mixing ratio of the ultrafine particles n_{uf} (Figure 12 c), whereas during both NPF phases
1138 the particle mixing ratios reached peak values of more than 20000 mg^{-1} or they often remained
1139 elevated ($> 10000 \text{ mg}^{-1}$). The course of n_{uf} is not mirrored at all by the CO signal (Figure 12 d),
1140 e.g. n_{uf} is at maximum values when CO is still at intermediate levels of $\sim 110 \text{ nmol mol}^{-1}$. In
1141 accordance with the results discussed in Section 5.1 and illustrated in Figure 8, high n_{uf} seems
1142 not directly coupled to the supply of pollutants by convective transport (Figure 12 c and d).
1143 Furthermore, in both NPF phases, the peaks of air's CO content (130 - 140 nmol mol^{-1} , Figure
1144 12 d) were accompanied by increasing mixing ratios n_{10nv} by a factor of up to two compared to
1145 the background (Figure 12 b), indicating the passage through the convective outflow plume,
1146 which also contained non-volatile aerosol material that was lifted together with gaseous
1147 pollutants.

1148 During the periods of the NPF observations, however, the ambient air temperature T_{amb} (Figure
1149 12 e) visibly fluctuates in the order of $\pm 1 \text{ K}$ around the respective mean temperature
1150 ($T_{mean} = 193 \text{ K}$ with standard deviation below 1 K). Over the NPF period, the time series of the
1151 temperature fluctuation ($T_{amb} - T_{mean}$, Figure 12 e) exhibits the shape of a wave. In reference to
1152 the first NPF phase and assuming an average airspeed of 170 m s^{-1} throughout this period, the
1153 time series of the temperature fluctuation covers a peak-to-peak duration, which converts into a
1154 horizontal distance of $\sim 90 \text{ km}$. It would go beyond the scope of this study to clearly attribute
1155 this temperature fluctuation to the GW activity initiated by a specific convective system.
1156 However, the amplitude and wavelength of the observed fluctuation correspond qualitatively
1157 and quantitatively to the values typical for GWs. If NPF is initialised by a negative temperature
1158 anomaly under supersaturated conditions, the newly formed ultrafine particles hardly



1159 evaporate at re-rising temperatures (e.g. when the GW-induced temperature anomaly becomes
1160 positive).

1161 The horizontal extent of GW-induced temperature anomalies, which can range from a few to
1162 hundreds of kilometres, is generally comparable with the magnitude of the horizontal extent of
1163 observed NPF fields (cf. Sections 2.1.3 and 4.3 as well as Figure 5 c). Since the time offset
1164 between NPF observation and NPF initiation is not exactly known, it is not straightforward to
1165 connect individual NPF events to specific incidents of GW-induced temperature anomalies.
1166 Moreover, during the monsoon season, several widely distributed, convective systems may
1167 induce GWs at the same time and the resulting, spatially propagating, temperature anomalies
1168 could interfere under each other at TTL heights. The amplification of temperature anomalies
1169 inherent with such interferences is neither locally resolvable nor quantifiable. Hence, GW-
1170 induced temperature anomalies may additionally promote the occurrence of NPF, particularly in
1171 cases, in which the enhancement of the NPF precursor saturation ratio, as prerequisite for NPF
1172 initiation, is not ascribable to direct (convective) uplift from the surface.

1173 7 Summary and Conclusions

1174 Between 27 July and 10 August 2017 the airborne StratoClim 2017 mission took place in
1175 Kathmandu, Nepal, with eight mission flights (~ 22.5 hours of COPAS measurement time above
1176 10 km, $\theta \geq 350$ K) up to altitudes of 20 km ($\theta \approx 475$ K) with the Russian high-altitude research
1177 aircraft M-55 *Geophysica*. The presented analysis comprises the description and discussion of
1178 numerous events of New Particle Formation (NPF), which were observed in the UT/LS region of
1179 the Asian Monsoon Anticyclone (AMA) over northern India, Nepal and Bangladesh.

1180 During the StratoClim 2017 mission, a total duration of 2 hours and 38.5 minutes was spent
1181 under NPF conditions in the region of the Tropical Tropopause Layer (TTL), where enhanced
1182 quantities of ultrafine particles of up to ~ 50000 mg⁻¹ (≈ 11000 cm⁻³) were detected at heights of
1183 15 – 16 km (~ 370 K). The majority of NPF observations with large amounts of ultrafine
1184 particles ($6 \text{ nm} < d_p < 15 \text{ nm}$) were observed at the lower TTL (~ 12-16 km, ~ 355 – 380 K) and
1185 below the tropopause. Nevertheless, NPF with enrichments of intermediate (~ 1000 – 2000 mg-
1186 ¹) or low (~ 300 – 500 mg⁻¹) mixing ratios of ultrafine particles were also observed at levels
1187 around the tropopause (~ 380 K) and up to about 17.5 km altitude (400 K). The frequency of
1188 NPF observations during StratoClim 2017 over durations of ~ 10 seconds (< 1.5 km horizontal



1189 distance) and with n_{uf} peaking up to 50000 mg^{-1} indicates a very effective and spacious source
1190 region of aerosols at TTL levels within the AMA. The numerous encounters of enhanced n_{uf} over
1191 several consecutive minutes (cf. Section 4.3 and Figure 4) indicate the NPF occurrence over
1192 extended fields of approximately 10 to 100 km (at event durations of 60 - 600 seconds).

1193 The persistence of ultrafine particles ($d_p < 15$ nm) in the presence of the background aerosol
1194 population is largely determined by coagulation and limited to few hours only. Within the
1195 supersaturated environment under NPF conditions, co-condensation of gaseous species other
1196 than those involved in NPF may further promote the growth of ultrafine particles. The
1197 comparatively short persistence of the particles in the ultrafine size mode implies:

- 1198 • After an expired NPF event, the number concentration of ultrafine particles decays due to
1199 coagulation by more than one order of magnitude within 2 hours.
- 1200 • About 3-4 hours after a NPF event, the reduced number of ultrafine particles impedes the
1201 identification of NPF events based on aircraft-borne *in-situ* measurements.
- 1202 • The interpretation of low and intermediate amounts of ultrafine particles is limited, as they
1203 may result from either moderate and just proceeding NPF or from an event with elevated
1204 NPF-rate that has phased-out over more than two hours before the measurement.
- 1205 • High amounts of ultrafine particles i.e. ($> 10000 \text{ mg}^{-1}$), however, indicate that NPF had
1206 occurred very shortly (less than one hour) prior to the measurement or was just proceeding
1207 when detected.

1208 The supersaturated conditions, under which NPF occurs, however, may also favour the co-
1209 condensation of gaseous substances (Yu et al., 2017). Whether coagulation or condensation
1210 predominantly contributes to the composition of the background aerosol remains open. Most
1211 likely, both processes impact the formation and persistence of the ATAL (Vernier et al. (2011a),
1212 and see also Höpfner et al. (2019); Mahnke et al. (2020)), which was mainly attributed to the
1213 uplift of pollution from the boundary layer by means of balloon-borne and satellite-based
1214 observations (Vernier et al. (2018); Brunamonti et al. (2018); Hanumanthu et al. (2020)). In
1215 general, a refractory core with diameter greater than 10 nm was detected in almost every
1216 second particle above 395 K up to 475 K. This indicates the supply of other particulate material
1217 due to the updraught within the AMA (cf. also Section 6), although meteoric particles from
1218 higher altitudes (Schneider et al., 2020) may also play a role.



1219 An indirect supply of the stratospheric (Junge) aerosol layer (Junge et al., 1961) at an altitude of
1220 ~ 25 km by freshly formed particles (at altitudes of up to 17.5 km) seems possible if a
1221 sufficiently effective transport mechanism is available. However, whether aerosol material
1222 subsides from TTL levels to mid-tropospheric altitudes and possibly contributes to cloud
1223 formation, as suggested by Andreae et al. (2018) to happen in the Amazon region, depends on
1224 the efficiency of downward transport, and on the aerosol's capability as CCN. Condensation of
1225 gaseous species other than those involved in the NPF process, and internal chemical conversion
1226 of various solutes within a particle could influence the aerosols' CCN capabilities. The transport
1227 times to altitudes far above or below the TTL appear long (several days and weeks) compared to
1228 the short persistence (hours) of ultrafine particles.

1229 Three different approaches were used to correlate the occurrence frequency of most recent NPF
1230 (most enhanced n_{uf}) with possible source regions of precursor material in the BL and to find a
1231 direct connection of NPF to recent convective uplift and air mass transport times. Exceptionally
1232 elevated abundances of ultrafine particles (about ten to hundred times above the level of
1233 background aerosol concentrations) are used as indication for very recent occurrence of NPF.
1234 Intermediate or lower number densities of ultrafine particles could lead to ambiguous
1235 conclusions (cf. Section 4.5):

1236 (1) The measurements indicate that highest n_{uf} values were predominantly found to coincide
1237 with intermediate to elevated CO mixing ratios of ~ 100 nmol mol⁻¹. Beyond that level, the
1238 mixing ratio of ultrafine particles is largely independent of the CO content (between 80 and
1239 145 nmol mol⁻¹) of the air at the lower TTL.

1240 (2) The most intensive uplift of air was confirmed to occur over the Himalayan mountain chain
1241 and its foothills. However, particular source regions of NPF precursors were not ascertainable
1242 within the BL. Furthermore, no indication was found that the most intense NPF was connected
1243 to short durations of air mass transport from the BL into the TTL.

1244 (3) The convective contribution did not immediately determine the intensity of the observed
1245 NPF. The release of the precursor material in the outflow region of the convective top may have
1246 occurred up to 6 days before the NPF observation. Occasionally, however, air mass residence
1247 times of more than 6 days and up to 14 days were found at TTL levels prior to the NPF detection.



1248 Consequently, the observed intensity of NPF is not unambiguously attributable to a) a specific
1249 source region on the ground or in the BL, or b) the effectiveness of the convective vertical
1250 transport, or c) the recent release of NPF-capable material from the convective outflow.

1251 Nevertheless, it should be the convective uplift, which intermittently supplies the lower TTL by
1252 NPF precursor material. At altitudes well above tropopause levels, such an immediate supply by
1253 convection is lacking and could alternatively only proceed by the slow uplift superimposed on
1254 the anticyclonic ascent of the AMA (~ 1 K per day, Vogel et al. (2019); von Hobe et al. (2020) and
1255 Section 1.2). Generally, the question arises whether air mass transport and supply by convective
1256 updraught alone are sufficient to increase precursors' supersaturation such that NPF is
1257 initialised. At TTL levels in AMA, diabatic cooling by emission of infrared (IR) radiation
1258 constitutes a spatially large-scaled process that potentially increases the supersaturation of an
1259 NPF precursor system, but which occurs mainly during night hours, i.e. in the absence of solar
1260 irradiation. Alternatively, adiabatic cooling, however, could induce sufficient supersaturation of
1261 a precursor and thus play a role as a trigger for NPF. Temperature anomalies associated with
1262 gravity waves (GW) could very well increase the supersaturation of a precursor by that crucial
1263 bit above the NPF threshold. Interfering gravity waves, such as those likely initiated during the
1264 convectively very active Asian monsoon season, may increase the probability that occurring
1265 temperature anomalies are adequately large. Furthermore, the vertical propagation of GW-
1266 induced temperature anomalies could initialise NPF above tropopause levels, a) where ambient
1267 air temperatures increase with altitude (from observational data with $\Delta T \approx 1.5$ K per $\Delta \theta = 10$ K),
1268 which principally counteracts the supersaturation of a precursor, and b) where in the absence of
1269 overshooting convection a direct supply of precursor material from below is lacking.

1270 The frequency of NPF observed during StratoClim 2017 exceeds all previous NPF detections
1271 with COPAS in the TTL over Brazil, Australia and West Africa (TROCCINOX 2005, SCOUT-03
1272 2005, SCOUT-AMMA 2006, cf. Borrmann et al. (2010); Weigel et al. (2011)). The maximum of
1273 detected ultrafine particles (~ 50000 mg⁻¹, correspondent to ~ 11000 cm⁻³ under ambient
1274 conditions at 360 K $< \theta < 370$ K) is in comparable orders of magnitude to the earlier COPAS
1275 observations (*ibid*). Moreover, the horizontal extent of the NPF fields during StratoClim 2017,
1276 ranging from a few hundred metres to about one hundred kilometres, well compares to previous
1277 COPAS observations, although caveats inhere in the distinction of individual but closely adjacent



1278 NPF fields due to the COPAS measurement resolution in conjunction with the flight speed of the
1279 M-55 *Geophysica*. The observations made during StratoClim 2017 indicate that frequent NPF
1280 with high production of ultrafine particles is capable of directly affecting the extent and
1281 persistence of the Asian Tropopause Aerosol Layer (ATAL). The continuous supply of freshly
1282 formed aerosol material, which coagulates both, internally and with the background aerosol, and
1283 which itself provides surface for the condensation of supersaturated gaseous substances, may
1284 contribute significantly to the aerosol composition of the ATAL up to altitudes of ~ 17.5 km
1285 (400 K). The chemical composition of the ATAL aerosol may include significant fractions of the
1286 material, which was previously involved in the NPF process and the particles' condensational
1287 growth, but this is subject to further investigation using the StratoClim 2017 data set.

1288 **Data availability:**

1289 *The data shown in this study are available at the StratoClim campaign database at*

1290 <https://stratoclim.icg.kfa-juelich.de/AfcMain/CampaignDataBase>

1291 *or they may be provided by respective PI upon request*

1292 **Author contribution**

1293 *RW evaluated the data, created the figures, and draughted the manuscript with contributions by CM, MB, and*
1294 *AD. SB participated in the data analyses and the manuscript draughting. The code of the coagulation simulation*
1295 *was provided by BPL, and the code was adapted by MB while the calculations were performed by CM. BV, FP*
1296 *contributed with meteorological re-analyses, BV, SiB, and BL performed the air mass trajectory analyses. SV*
1297 *and FD'A took care of the CO data. UCSE data were delivered by GB. The manuscript was critically reviewed*
1298 *by CM, MB, AD, BV, FP, SV, FD'A, SiB, BL, BPL, and SB.*

1299 **Competing interests**

1300 *The authors declare no competing interests.*

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1829 **Figure captions**

1830 Figure 1: (a) Flight patterns conducted throughout the StratoClim 2017 mission over Nepal,
1831 India, and Bangladesh. (b) Regions with elevated number concentrations of ultrafine particles
1832 (N_{uf}) of sizes in the diameter range $6 \text{ nm} < d_p < 15 \text{ nm}$ as observed by means of COPAS
1833 measurements are indicated by the colour-code and by symbol size along the flight tracks.

1834 Figure 2: Synopsis of vertical profiles of the total number concentration (median with 10th, 25th,
1835 75th, 90th, and 99th percentiles) of sub-micrometre sized particles as a function of potential
1836 temperature obtained from condensation nuclei (CN) detections over (a) Brazil (TROCCINOX,
1837 2005), over (b) West Africa (SCOUT-AMMA, 2006) and over (c) the Indian subcontinent
1838 (StratoClim 2017). During TROCCINOX (a) and SCOUT-AMMA (b) the number concentrations N_4
1839 at lower heights ($\theta < 350 \text{ K}$) were measured aboard the DLR Falcon (cf. (Borrmann et al., 2010)
1840 and (Weigel et al., 2011)). All high-altitude measurements ($\theta > 350 \text{ K}$) and the entire
1841 StratoClim 2017 data set result from COPAS measurements. (c) The median profile of $N_{5.3}$ from
1842 repeated measurements (over the years 2004 – 2007) with the NMASS multi-channel CN
1843 counter over Central America (aboard the NASA WB-57F, data courtesy of J. C. Wilson, Denver
1844 University, 2011).

1845 Figure 3: (a) 1-Hz-resolved particle mixing ratios n_6 and n_{10} (grey-shaded COPAS data points)
1846 with n_6 median profile from StratoClim 2017 with COPAS data from tropical regions (over Brazil,
1847 TROCCINOX 2005 and over West Africa, SCOUT-AMMA 2006, cf. (Borrmann et al., 2010)). The
1848 median profile of measurements in the tropics over the Americas (Brock et al., 1995) is added.
1849 (b) The vertical distribution of the mixing ratio of ultrafine particle ($n_{\text{uf}} = n_{6-15}$) in compliance
1850 with the NPF criterion (cf. Section 2.1.3). (c) The 1-Hz-resolved mixing ratio of non-volatile
1851 particles (i.e. thermostable at $\sim 270^\circ\text{C}$) from COPAS measurements throughout StratoClim 2017
1852 with corresponding median profile, including 25th and 75th percentile. Herein, the n_6 median
1853 profile is recalled for comparison from Panel a. (d) The fraction $f (= n_{10\text{nv}}/n_{10} \cdot 100)$ of non-
1854 volatile particles with median, and with 25th and 75th percentiles. Median data points are
1855 connected with lines to guide the reader's eyes.

1856 Figure 4: Frequency distribution of the duration of observed NPF events (cf. Section 2.1.3 for
1857 definition) throughout the StratoClim 2017 mission. (a) The general view, (b) the close up views
1858 of the shortest events, and (c) of the least frequent events.

1859 Figure 5: (a) Diurnal variation of the occurrence frequency of NPF events (cf. Section 2.1.3 for
1860 definition). (b) The diurnal distribution of NPF events' mean particle mixing ratio \bar{n}_{uf} with
1861 standard deviation σ , coloured by flight date, and (c) in colours of the (logarithmic) duration of
1862 respective event. Note, the mean horizontal distance is derived from the event duration based on
1863 a mean flight speed of 154 m s^{-1} ($\sigma = \pm 39 \text{ m s}^{-1}$, variable flight attitude remains unconsidered)
1864 and is understood as equivalent horizontal extension of a NPF event.

1865 Figure 6: Mean particle mixing ratio \bar{n}_{uf} of individual NPF events as function of (left column) the
1866 vertical distance from the mean lapse-rate tropopause ($\Delta\theta$), and of (right column) the equivalent
1867 latitude (90° represents the centre of the AMA as projected to polar coordinates). Data points
1868 are coloured by flight date (Panels a and b) and by CO mixing ratios (Panels c and d). (e) The
1869 mean particle mixing ratio \bar{n}_{uf} as function of the equivalent latitude is colour-coded by the values
1870 $\Delta\theta$ (colour scale on the left of panel (e)).

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1872 Figure 7: Results of a coagulation simulation based on the assumption of a distinct and expired
1873 burst-like event. The simulation's initial particle size distribution (black circles; horizontal bars
1874 indicate the width of each size bin) is merged from data of three COPAS detectors (for N_6 , N_{10} ,
1875 and N_{15}) and of the UHSAS-A ($65 \text{ nm} < d_p < 1 \text{ }\mu\text{m}$) as detected during NPF encountered on 04
1876 August 2017, between 04:04:40 and 04:05:06 UTC. (a): The processing particle size distribution
1877 (coloured lines) over several hours. (b): The concentration of ultrafine particles (N_{uf}) over the
1878 simulation's run time and its fractional contribution to the total particle number concentration
1879 (N_{total}). Furthermore, the simulated decay of variably multiplied N_{uf} (by factors 0.1, 10, and 100)
1880 as initial input of the simulation under constant background conditions (dashed lines).

1881 Figure 8: Particle mixing ratio of fine-mode particles n_6 (grey dots in the background) and of
1882 ultrafine particles n_{uf} (colour-coded with reference to the potential temperature) in relationship
1883 to the CO mixing ratio. The median n_{uf} with the 25th and 75th percentile is shown in bin widths of
1884 $2.5 \text{ nmol mol}^{-1}$ of the CO mixing ratio (black dots), which are connected by lines to guide the eyes
1885 of the reader.

1886 Figure 9: Upper panels: geographic position of the last boundary layer (BL) contact of the NPF-
1887 connected air mass backward trajectories. Bottom panels: geographic position of the maximum
1888 ascent rate of these trajectories. By means of the chemistry transport model CLaMS and based on
1889 ERA-5 data the backward trajectories were analysed over the last 50 days prior to the NPF
1890 detection as starting point of each trajectory. Points are coloured with reference to the
1891 (logarithmic) mixing ratio n_{uf} of ultrafine particles (Panels a and b) or to the air mass transport
1892 time since the last BL contact (Panels c and d), grey data points indicate transport times > 25
1893 days.

1894 Figure 10: Vertical profile of the 1-Hz-resolved particle mixing ratio of ultrafine particles n_{uf}
1895 colour-coded by the air mass transport time (days) from the boundary layer (BL). By means of
1896 the chemistry transport model CLaMS and based on ERA-5 data the backward trajectories were
1897 analysed over the last 50 days prior to the NPF detection as starting point of each trajectory,
1898 grey data points indicate transport times > 25 days.

1899 Figure 11: Vertical profile of the event-wise mean particle mixing ratio of ultrafine particles $\overline{n_{\text{uf}}}$
1900 with standard deviation σ (bars) as a function of the mean potential temperature ($\pm \sigma$). (a) The
1901 data points are colour-coded by the proportion of convective contribution to the air sample. (b)
1902 The data points are coloured by the time (days) since the release of the air mass at the top of a
1903 convective cell.

1904 Figure 12: Time series of data sampled during a section of a StratoClim 2017 flight (KTM 6) on
1905 06 August 2017. Except the manoeuvre period between 09:20 and 09:30 (UTC), a strictly
1906 constant altitude and pressure level (Panel a) were maintained. Particle mixing ratios n_6 , n_{10} and
1907 n_{15} and $n_{10\text{NV}}$ (Panel b), the mixing ratio of the ultrafine particles n_{uf} (Panel c), the CO mixing
1908 ratio (Panel d), the ambient air temperature (T_{amb}), and the temperature fluctuation ($T_{\text{amb}} - T_{\text{mean}}$)
1909 (Panel e) feature different characteristics and sequence during two NPF phases (oblique hatched
1910 areas).



1911 **Figures**

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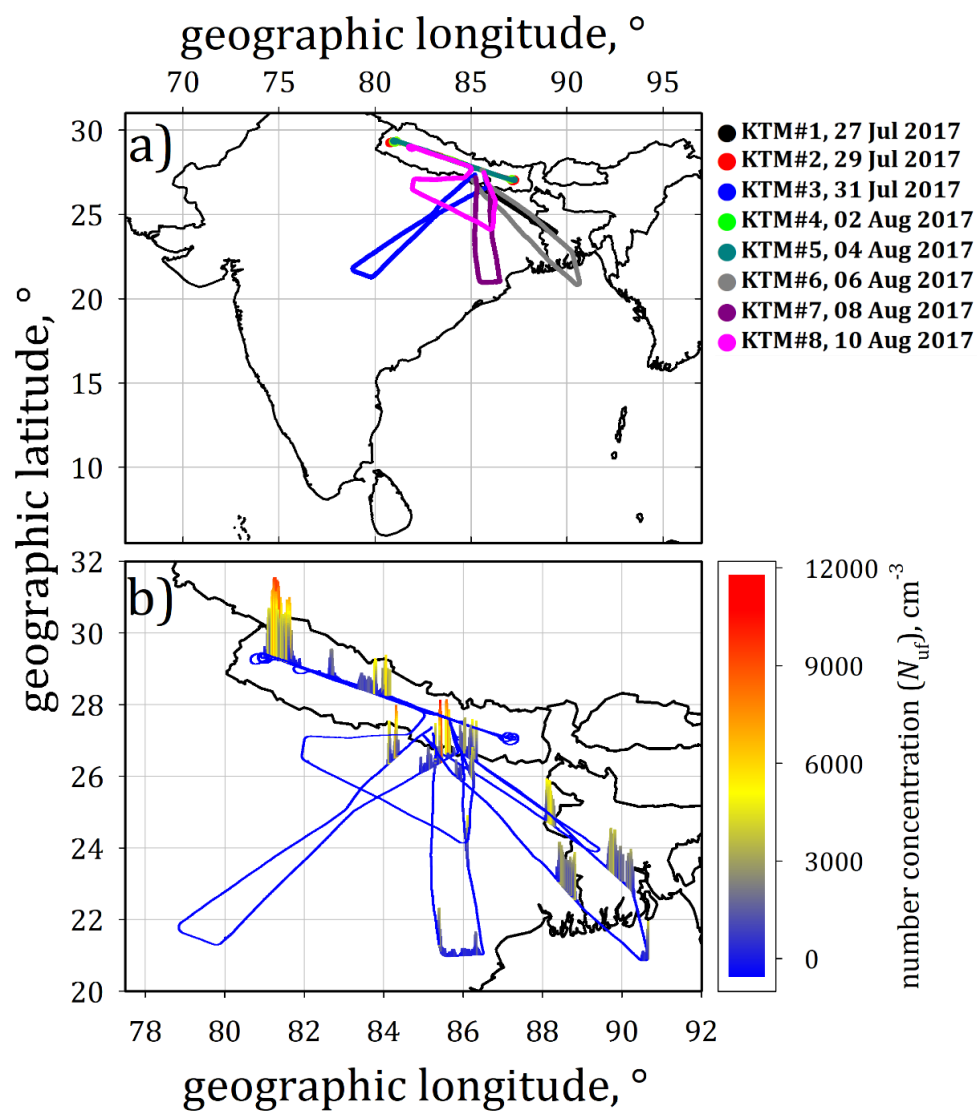


Figure 1

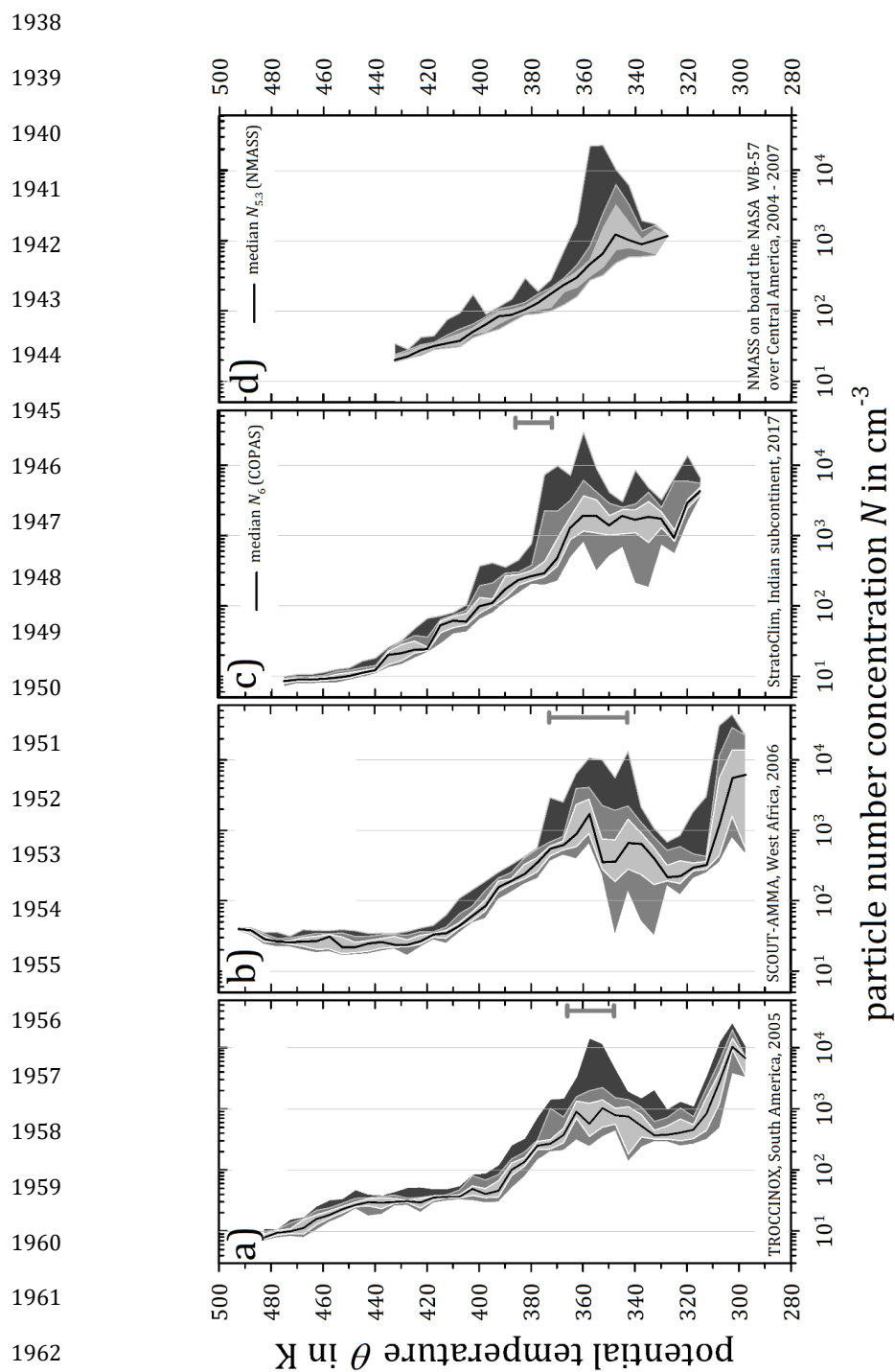


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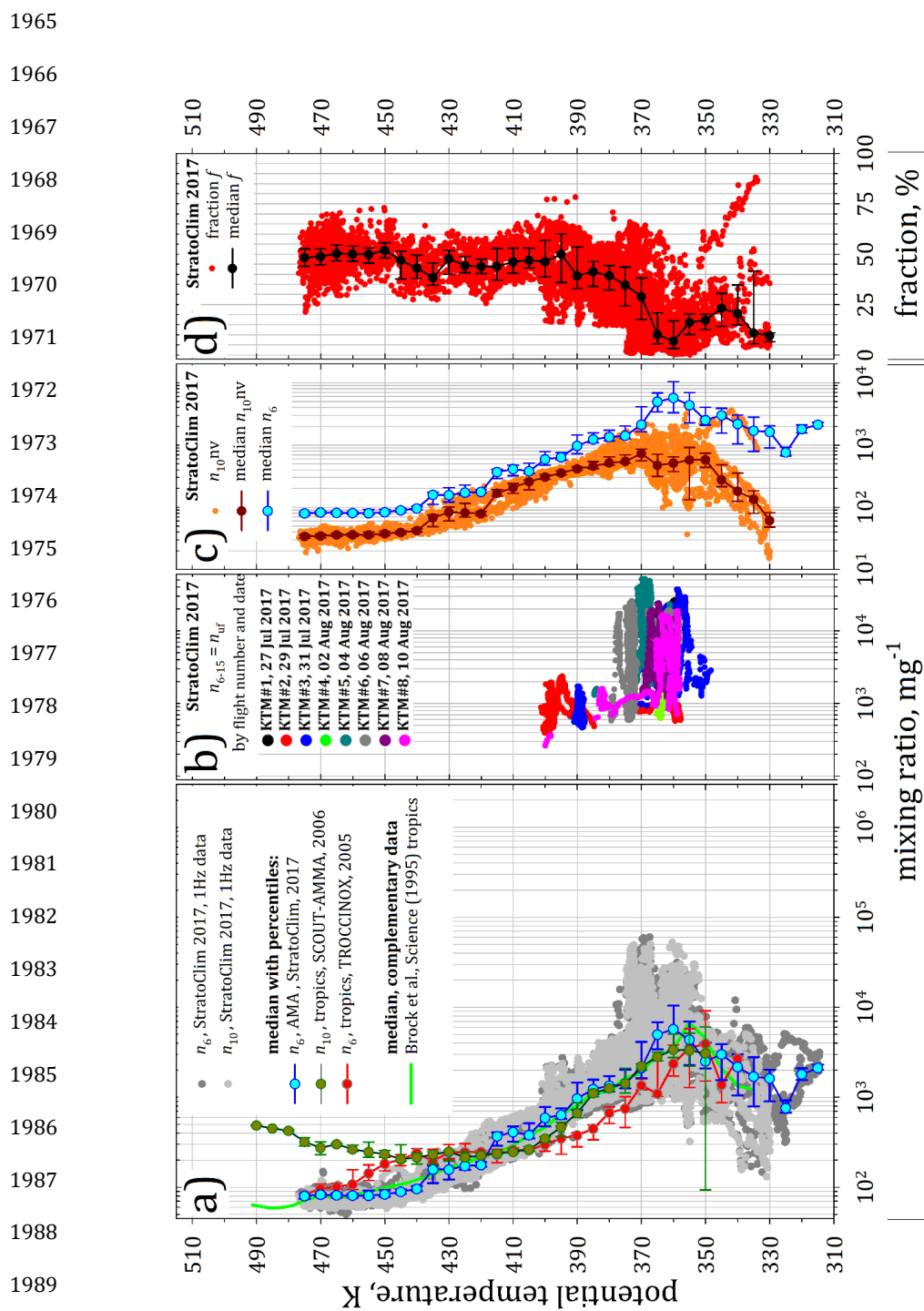


Figure 3



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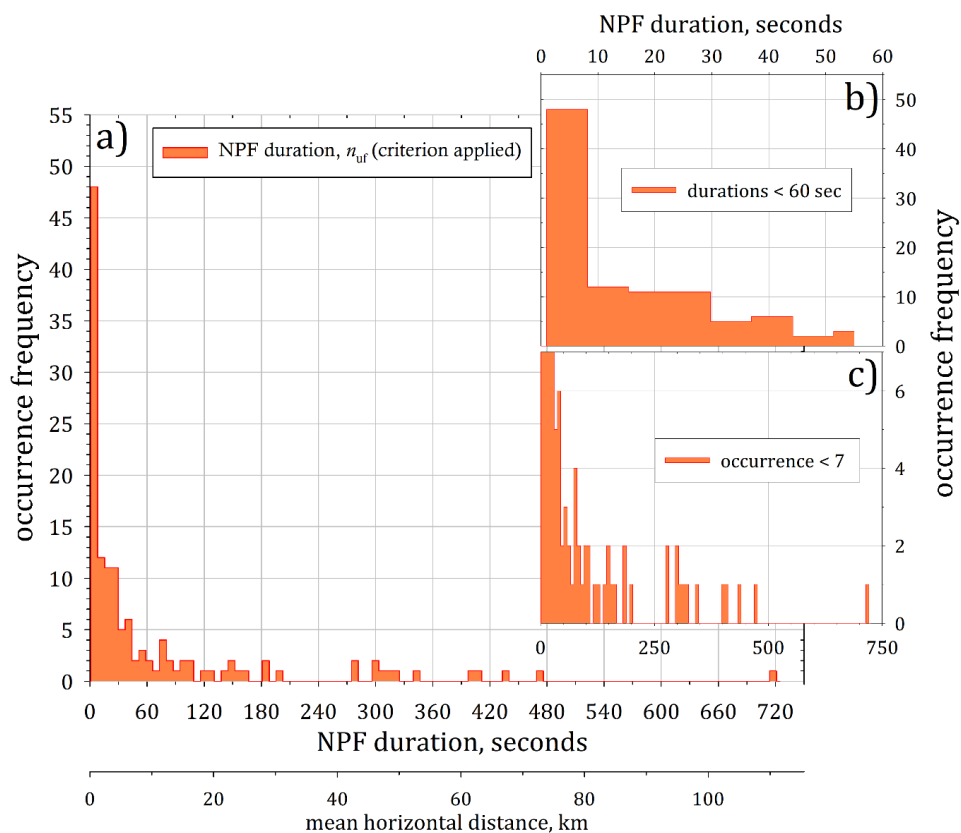


Figure 4



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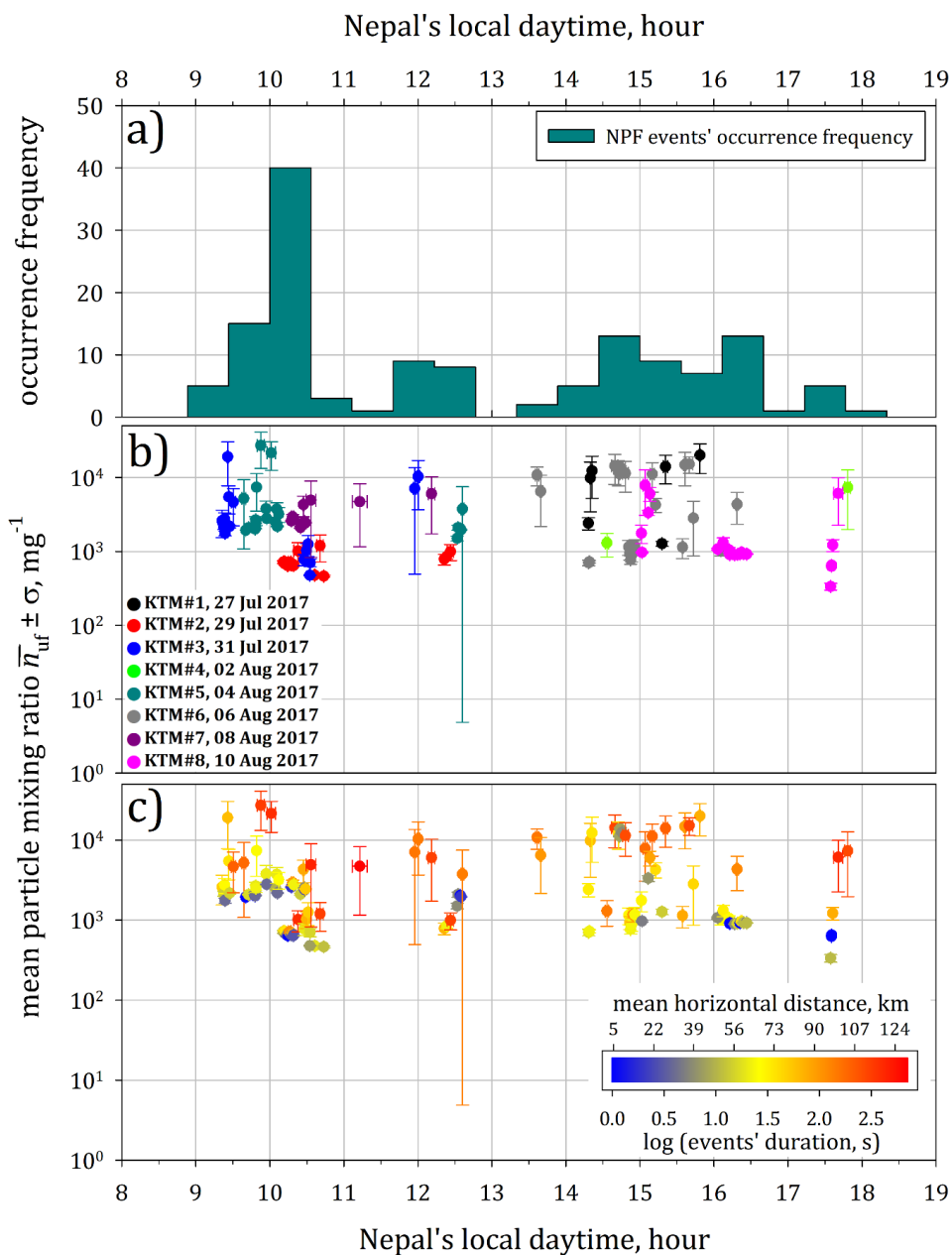


Figure 5



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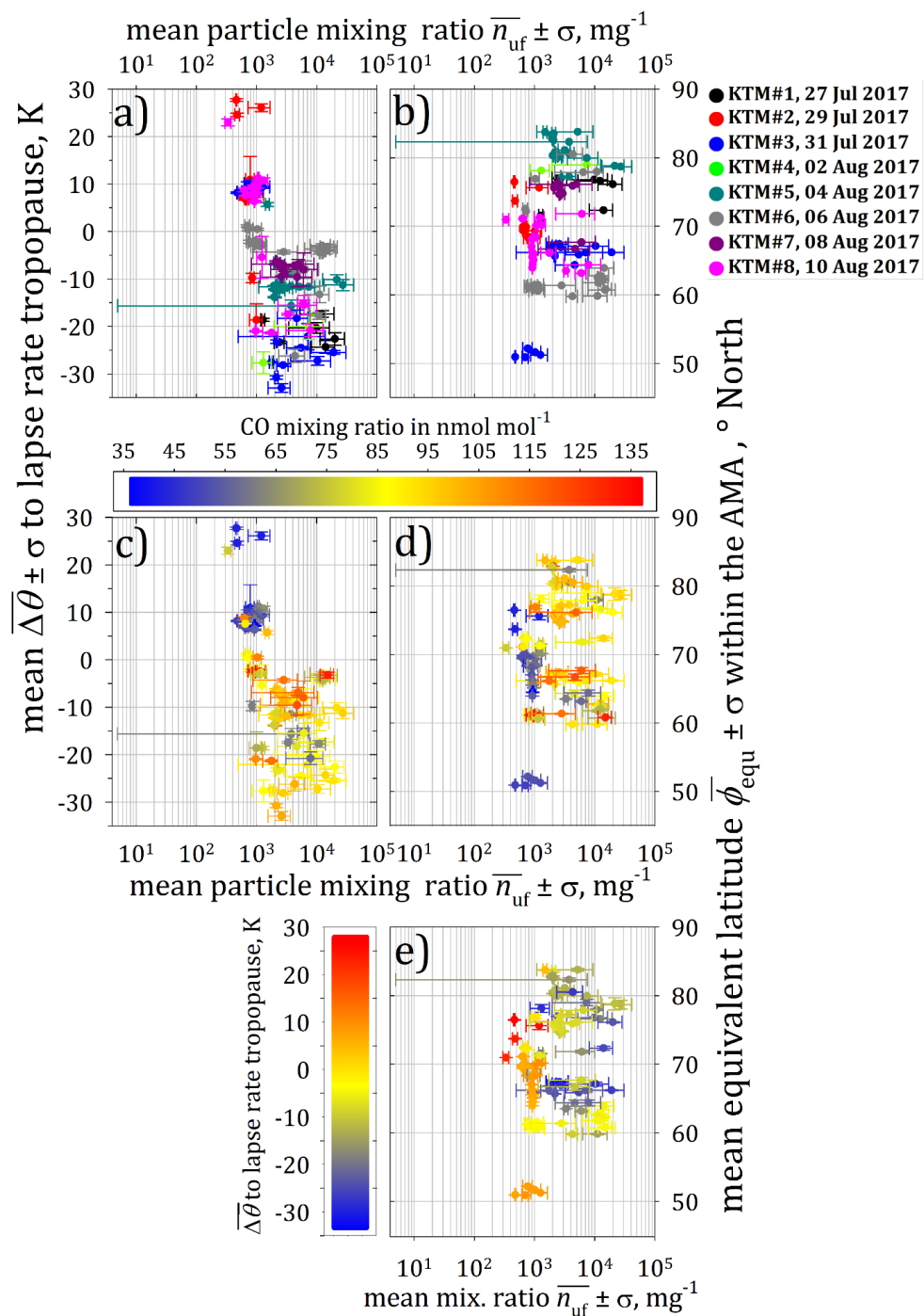


Figure 6



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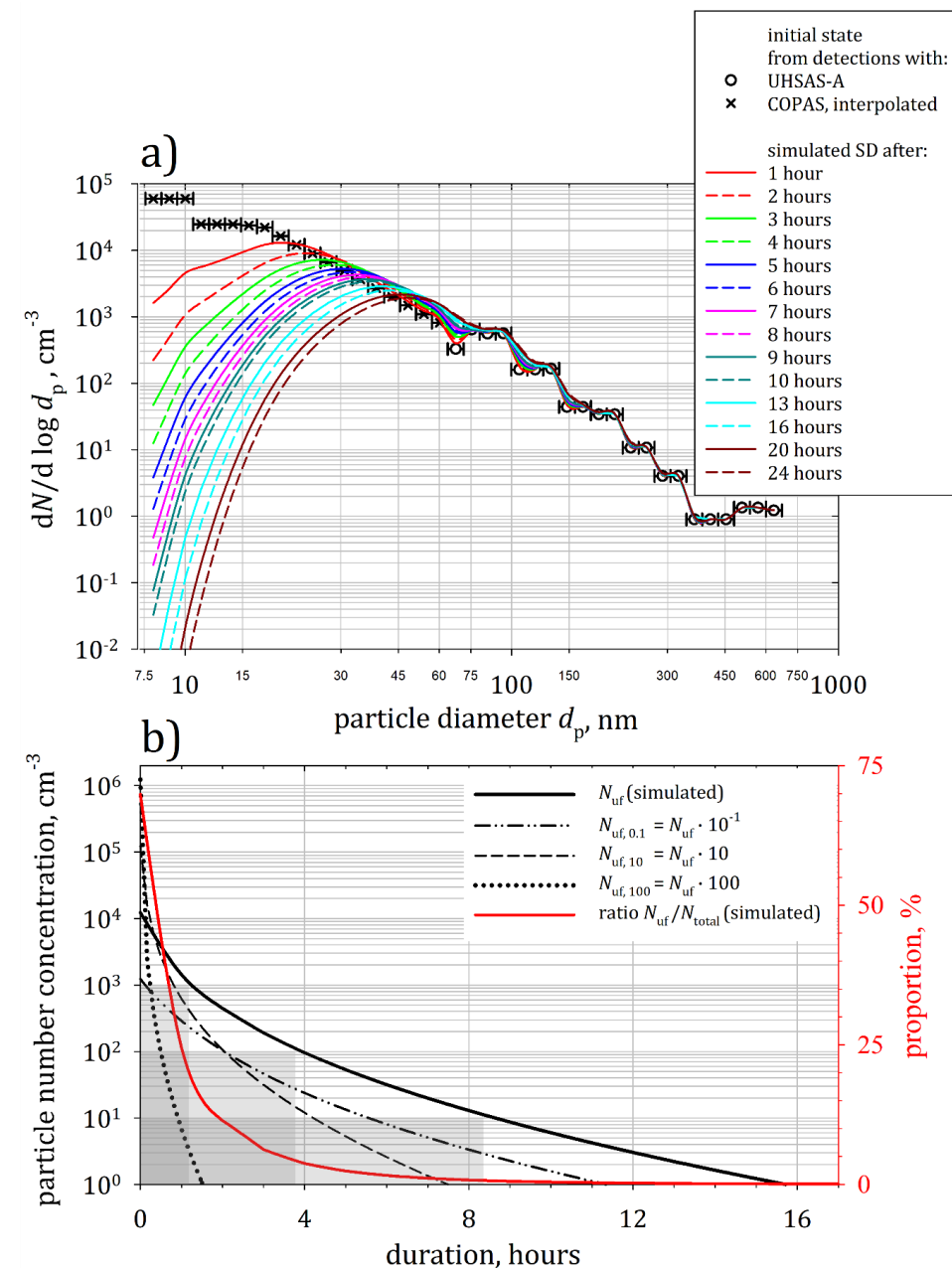


Figure 7



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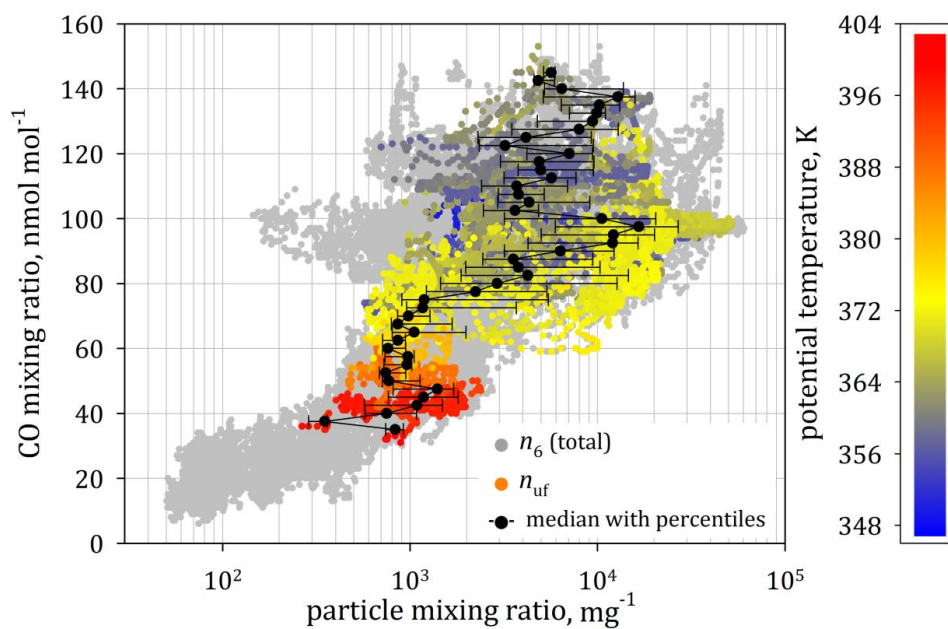
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2124 Figure 8





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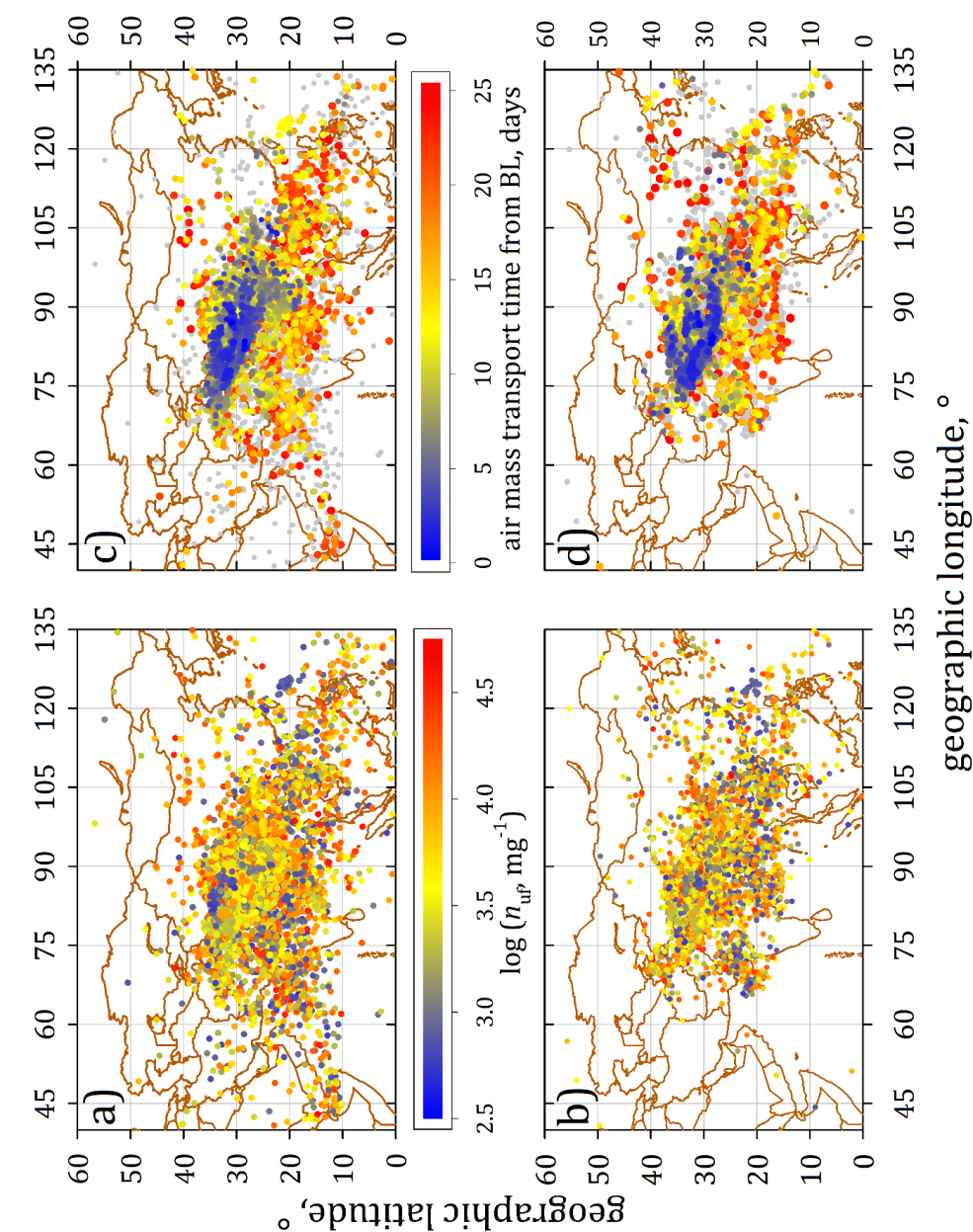


Figure 9



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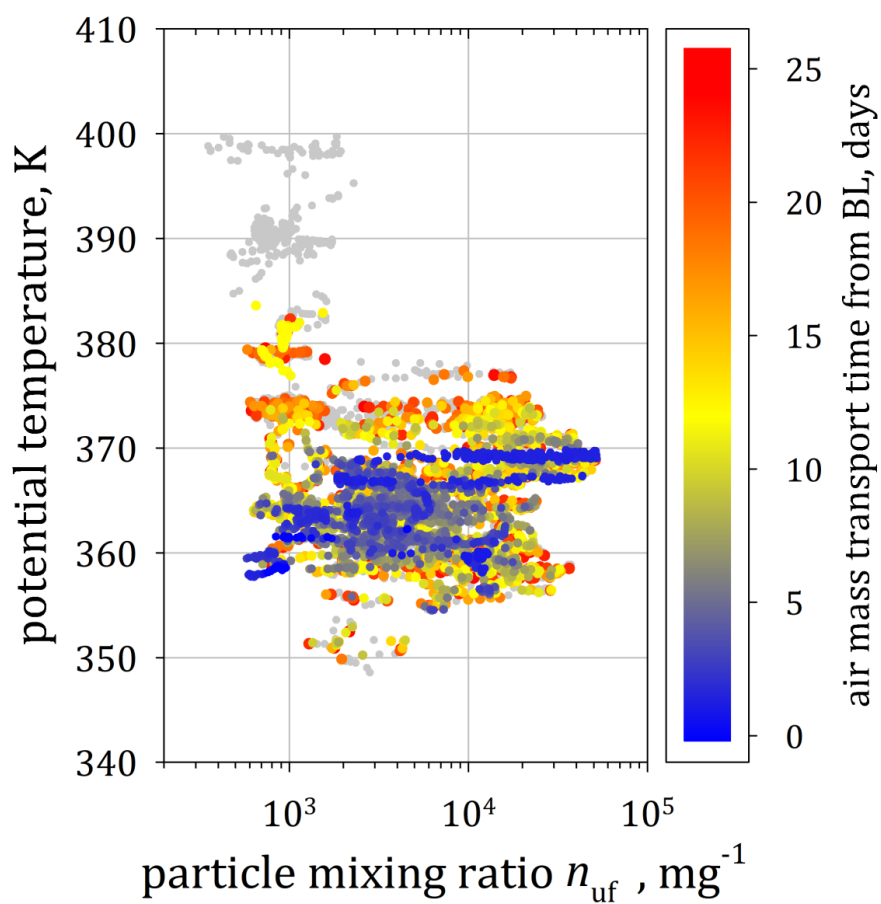
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2175 Figure 10

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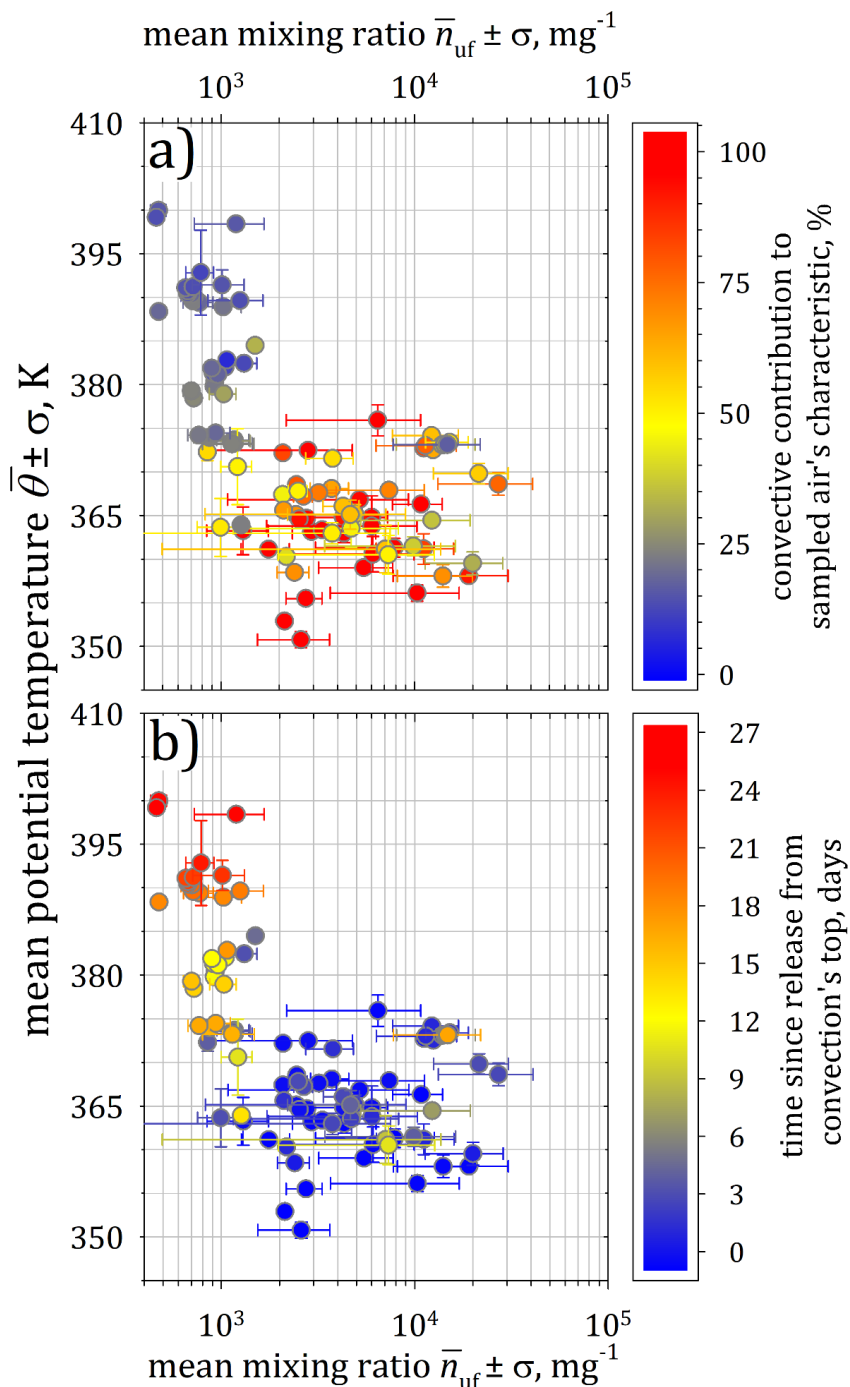


Figure 11



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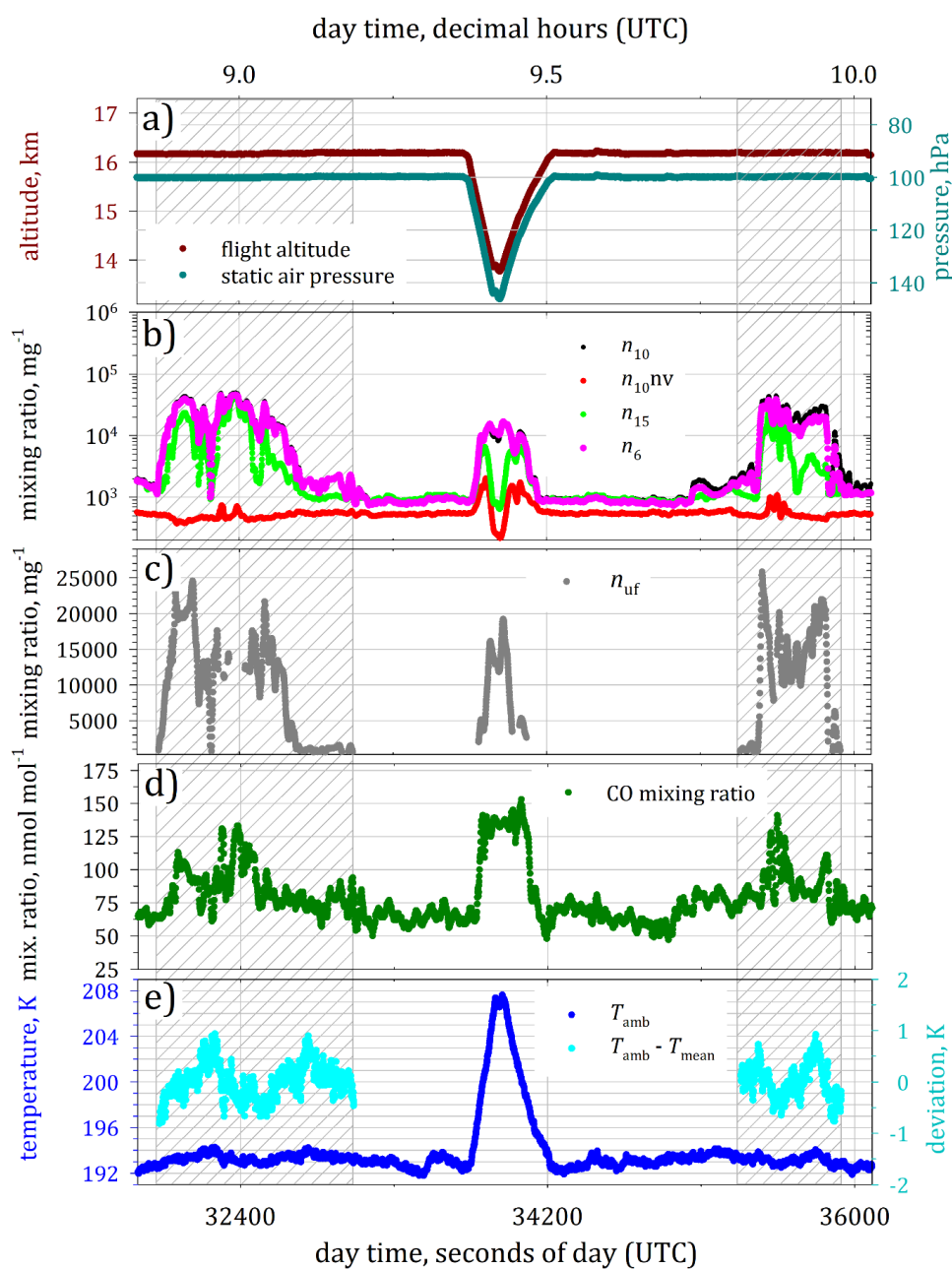


Figure 12



2235 **Tables**

pressure, hPa	particle diameter, nm										$\bar{\Lambda}_{6-15}$ %	κ_L (dimensionless)
	6	7	8	9	10	11	12	13	14	15		
	particle size dependent transmission efficiency, %											
80	60	65	70	74	77	79	81	82.5	84	85	24.25	1.32
150	70	75	77.5	81	83	84.5	86.5	87.5	88.5	89	17.75	1.22
300	77.5	81.5	84	86.5	88	89.5	90.5	91.5	92	92.5	12.65	1.14
400	80	83	85	87.5	89	90.5	91.5	92	92.8	93.5	11.52	1.13

2236 **Table 1**

2237 Re-calculated pressure-dependent corrections κ_L for number concentrations of ultrafine
 2238 particles due to particle losses ($\bar{\Lambda}_{6-15}$) in the aerosol line configuration (both COPAS instruments
 2239 attached to a single aerosol inlet) as deployed during StratoClim 2017, by using the Particle Loss
 2240 Calculator (von der Weiden et al., 2009) modified for low pressure applications. $\kappa_L = 100/(100-$
 2241 $\bar{\Lambda}_{6-15})$, correspondingly to Weigel et al. (2009).