1	An overview on the airborne measurement in Nepal,-part 1: vertical profile of aerosol size-number,			
2	spectral absorption, and meteorology			
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13 Abstract

14 The paper provides an overview of an airborne measurement campaign with a microlight aircraft, 15 over the Pokhara Valley region, Nepal, a metropolitan region in the central Himalayan foothills. This is 16 the first aerial measurements in the central Himalayan foothill region, one of the polluted but relatively 17 poorly sampled regions of the world. Conducted in two phases (in May 2016 and December 2016-18 January 2017), the goal of the overall campaign was to quantify the vertical distribution of aerosols over 19 a polluted mountain valley in the Himalayan foothills, as well as to investigate the extent of regional 20 transport of emissions into the Himalayas. This paper summarizes results from first phase where test flights were conducted in May 2016 (pre-monsoon), with the objective of demonstrating the potential of 21 22 airborne measurements in the region using a portable instrument package (size with housing case: 0.45 23 m x 0.25 m x 0.25 m, 15 kgs) onboard an ultralight aircraft (IKARUS-C42). A total of five sampling test 24 flights were conducted (each lasting for 1-1.5 h) in the Pokhara Valley to characterize vertical profiles of 25 aerosol properties such as aerosol number and size distribution (0.3-2 μ m), total particle concentration 26 (>14 nm), aerosol absorption (370-950 nm), black carbon (BC), and meteorological variables. Although 27 some interesting observations were made during the test flight, the study is limited to a few days (and 28 only a few hours of flight in total) and thus, the analysis presented may not represent the entire 29 pollution-meteorology interaction found in the Pokhara Valley

30 The vertical profiles of aerosol species showed decreasing concentrations with altitude (815 to 31 4500 m a.s.l.); steep concentration gradient below 2000 m (a.s.l.) in the morning and mixed profiles (up to ca. 4000 m a.s.l.) in the afternoon. The near-surface (<1000 m a.s.l.) BC concentrations observed in 32 33 the Pokhara Valley were much lower than pre-monsoon BC concentrations in the Kathmandu Valley, 34 and similar in range to Indo-Gangetic Plain (IGP) sites such as Kanpur in India. The sampling test flight 35 also detected an elevated polluted aerosol layer (around 3000 m a.s.l.) over the Pokhara Valley, which 36 could be associated with the regional transport. The total aerosol and black carbon concentration in the 37 polluted layer was comparable with the near-surface values (<1000 m a.s.l.). The elevated polluted layer 38 was also characterized by high aerosol extinction coefficient (at 550 nm) and was identified as smoke 39 and a polluted dust layer. The observed shift in the westerlies (at 20-30° N) entering Nepal during the 40 test flight period could be an important factor for the presence of elevated polluted layers in the 41 Pokhara Valley.

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43 **1. Introduction**

44 The Himalayas and surrounding regions are one of the unique ecosystems in the world, with a great 45 variety in the geography and socio-economics, and a notable significance in the context of regional and 46 global environmental change. Areas in the foothills of the Himalayas still constitute large regions of 47 rural populations along with pockets of rapidly growing cities. Consequently, there is a complex 48 interaction among changing emission sources and their interaction with regional and global climate 49 change. Among emitted air pollutants, the chemical and physical properties of aerosols have been linked 50 to significant burdens of disease, to melting of glaciers, to crop loses, to hydrological changes and to 51 cloud properties (Bollasina et al., 2011; Vinoj et al., 2014; Lau, 2014; Burney and Ramanathan, 52 2014;Brauer et al., 2012;Cong et al., 2015;Li et al., 2016).

53 Sources of aerosols in the Himalayas and the nearby Indo-Gangetic Plain (IGP) typically vary 54 between urban, peri-urban and rural locations; fossil fuel and industrial emissions such as vehicles, brick 55 kilns, waste burning, cement factories etc., are typically urban and peri-urban; biomass cookstove, 56 agriculture and waste burning and forest fires are often linked to emissions from rural areas (Guttikunda 57 et al., 2014;Venkataraman et al., 2006;Stone et al., 2010). Secondary chemical pathways also contribute 58 to the aerosols in the ultrafine and accumulation-mode range via particle formation events (Venzac et 59 al., 2008;Neitola et al., 2011).

60 Aerosol properties in the Himalayas have large spatial and temporal variations, especially in the pre-61 monsoon and monsoon season. These observed variations are influenced by emission sources, regional 62 meteorology, and geography (Dey and Di Girolamo, 2010). The influence of aerosol particles on local and regional weather during these adjacent seasons has significant implications for timing, intensity and 63 64 spatial distribution of the summer monsoon in the region (Bollasina et al., 2011;Ramanathan et al., 65 2001). Studies describing the aerosol-meteorology interaction are often missing in the Himalayan region partly due to lack of surface and airborne measurements of aerosol properties along with meteorology. 66 67 Most past campaign-mode measurements in the Himalayan regions, to our knowledge, have been 68 ground measurements, which have aided in evaluating aerosol properties, and their transformation and 69 transport mechanisms (Shrestha et al., 2013;Shrestha et al., 2010;Ramana et al., 2004;Marcq et al., 70 2010; Panday and Prinn, 2009; Cho et al., 2017). Long-term continuous measurements of aerosols and 71 meteorology are limited to a few stations in the High Himalayas, such as the recently discontinued Nepal 72 Climate Observatory at Pyramid (NCO-P, 27.95° N, 86.81° E, 5050 m a.s.l.), a high altitude observatory 73 located near basecamp of Mt. Everest. Columnar and satellite measurements such as AERONET and 74 CALIPSO have provided a regional overview of aerosol type and vertical distribution, as well as

estimation of the aerosol heating rate in the atmospheric column (Kuhlmann and Quaas, 2010;Gautam
et al., 2011;Pandey et al., 2017). However, these measurement techniques often suffer from large
uncertainty and biases while retrieving the complex nature of the aerosols observed in the region (Jai
Devi et al., 2011).

79 Regional meteorology in the 850-500 mb range plays an important role in the transformation and 80 transport of aerosols from Western Asia to the IGP, the Himalayan foothills, the Himalayan and Tibetan 81 Plateau region (Decesari et al., 2010; Marinoni et al., 2013; Lüthi et al., 2015). At these altitudes, 82 synoptic- scale air masses are mostly westerly/northwesterly during the pre-monsoon and 83 southwesterly/easterly during the monsoon. These air masses are often linked to dust aerosol transport 84 during the pre-monsoon season from Western Asia into the Himalayas, including populated mountain 85 valley such as Kathmandu and Pokhara Valley in Nepal. The transported dust aerosol also mixes with the 86 primary emission (or anthropogenic aerosols) in the IGP and accumulates from northern to eastern IGP 87 along the Himalayan foothills (Gautam et al., 2009b;Gautam et al., 2011). The total aerosol loading is 88 often the highest during the pre-monsoon season in the IGP (Gautam et al., 2009a;Raatikainen et al., 89 2014), intensified further by weak surface/zonal winds and numerous open biomass burning and forest 90 fires events (Kaskaoutis et al., 2012). The polluted aerosol layer in the IGP is advected into the Himalayas 91 by synoptic-scale westerlies (~500 mb) and also by the valley wind circulation within or along the 92 planetary boundary layer (PBL) (Lüthi et al., 2015). The advection is also facilitated by the strong updraft 93 and PBL expansion (the highest in the pre-monsoon in the IGP) often mixing with the synoptic-scale 94 westerlies (Raatikainen et al., 2014). Because of strong convective activity in the IGP, the polluted air masses near the surface are often lifted up to 5-7 km or higher (Kuhlmann and Quaas, 2010). In addition 95 96 to the synoptic-scale transport, thermally-driven valley winds also enable the transport of humid and 97 polluted air mass (with enhanced absorbing fraction) from IGP into the Himalayan foothills, and further 98 up into the mountain valleys and elevated locations (Raatikainen et al., 2014;Lüthi et al., 2015;Gogoi et 99 al., 2014; Putero et al., 2014; Decesari et al., 2010; Marcq et al., 2010). Strongly coupled with the 100 expansion of the PBL in the IGP, the upslope movement of polluted air masses into the foothills and 101 further east is characterized by late afternoon peaks in aerosol optical depth (AOD) in many 102 measurement sites along the Himalayan range such as Hanle Valley (Ladakh, India), Mukteswar and 103 Manora site (Nainital, India), Hetauda (Nepal), Langtang Valley (Nepal), Dhulikhel (Nepal), Kathmandu 104 Valley (Nepal) and NCO-P (Nepal). The temporal and spatial extent of this observed "ventilation" at 105 multiple locations could be indicative of a regional-scale transport than mesoscale (Gogoi et al., 106 2014;Raatikainen et al., 2014;Gautam et al., 2011;Putero et al., 2015;Marcq et al., 2010).

107 To date, there have been no observations of vertical distributions of aerosol and gaseous species 108 carried out in the Himalayan region. Therefore, the airborne measurement campaign was designed to 109 address two major questions: (i) what is the variation in the aerosol properties, notably the vertical 110 distributions, over a polluted mountain valley, and (ii) what is the quantitative extent of regional 111 transport of aerosols in the higher Himalayas? The campaign was carried out in two phases in the 112 Pokhara Valley and surrounding areas in Nepal. In the first phase, test flights were conducted in May 2016 and in the second phase, intensive sampling flights were carried out in December 2016-January 113 114 2017. This paper provides an overview of the measurement campaign and results from the test flights in May 2016 which include snapshots of vertical profiles of aerosol size, number, and composition, along 115 116 with meteorological parameters. The airborne measurements presented in this paper are supplemented 117 with observations of local and regional meteorology, as well as satellite and ground-based column-118 integrated aerosol microphysics and radiative properties (see section 3.1.1 and 3.1.2, also Supplementary S7). A companion paper will follow with more detailed observations and results based 119 120 on the intensive measurements carried out during December 2016-January 2017.

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122 2. Ultralight measurements in Nepal

123 2.1. Details of the airborne measurement unit

124 A single-engine two-seater microlight aircraft (IKARUS C-42, COMCO IKARUS, Germany) was used as 125 the aerial platform. The technical specification of the aircraft includes approximately 4 h of flying time, a short take-off run, an additional payload of up to 50 kg, and it is suitable for small spiral movement in 126 the air. The aircraft has a cruising speed of 165 kmh⁻¹, and a 5-6 ms⁻¹ rate of climb which makes it an 127 128 appropriate aerial vehicle to perform measurements at altitudes within the PBL and as close as 50 m 129 above ground level. More detail about the aircraft is available here (http://www.comco-130 ikarus.de/Pages/c42a-technik.php?lang=en). Its size, speed, and maneuverability offered a decent climb 131 to the free troposphere to capture vertical profiles in the rough terrains of Nepal. The aircraft used for the study is operated by the Pokhara Ultralight Company for recreational flights around the Pokhara 132

133 Valley.

134 The instrument package was specifically designed and tested for aerial measurements (Junkermann,

135 2001). Table 1 describes each instrument and the integration performed to prepare the package for the

aerial deployment. The instrument package consists of a GRIMM OPS (optical particle spectrometer)

model 1.108 for particle size distributions (0.3 to 20 µm, 16 size bins) with sampling frequency of 6 s,

138 and a TSI condensation particle counter (CPC) 3760 for total particle concentration (>14 nm) at 1 s 139 resolution (See Figure S1 in the supplement). The package also included a Magee Scientific aethalometer 140 (AE42) for aerosol absorption at seven different wavelengths (370 -950 nm). The instruments were 141 reduced in weight for use on the aircraft. The CPC was operated with a constant mass flow and an 142 internal direct current (DC) pump instead of the original flow regulation by a critical orifice. 143 Meteorological parameters including temperature and dew point were sampled at a rate of 1 s using 144 METEOLABOR (TPS3). All the sensors were connected to a modular computer (PC104) for data 145 acquisition. The PC104 is also equipped with a Global Positioning System (GPS), and multiple serial and 146 analog connectors. For inflight instrument checks and quick online overview of the atmospheric 147 conditions, a small liquid crystal display (LCD) was also connected to the PC104 and placed in the cockpit 148 areas for the flight crew. This display showed real-time aerosol number concentrations and

149 meteorological parameters.

150 Table 1. Instrument package deployed in the microlight aircraft

151 The instrument package weighs approximately 15 kg and consumes <60 W, well within the 152 power supply range of the aircraft battery. It is housed in an aluminum box (0.45 m x0.25 m x0.25 m) and 153 can be easily integrated with a mobile platform such as the IKARUS (See Figure S1). In IKARUS, the 154 instrument was placed in the rear section behind the seats which is otherwise almost empty or used for 155 cargo, and only contains the fuel tank and supporting aluminum bars. The sample inlet line (internal 156 diameter of 0.004 m or ~4.0 mm ID brass tubing) ran along the wingspan and was approximately 1.8 m 157 (in length, sampling tube) from the cockpit. Once the sample line is inside the aircraft, it is distributed to 158 all the aerosol instruments using a simple metal flow splitter (0.006 m ID). No external pump was used 159 to pull the aerosol into the sampling line, the total flow (~ 3.0 lpm) in the sampling line (before the split) 160 was due to the internal pump of all the instruments. The sample inlet positioning at the end of the 161 wingspan also minimizes the influence of the aircraft propeller, located in the front of the cockpit.

162 2.2. Site description

Pokhara Valley is Nepal's second largest populated valley (pop. >250,000) after the Kathmandu Valley (CBS, 2011). The valley is approximately at 815 m (a.s.l.), ~150 km west of the Kathmandu Valley, and ~90 km northeast of the southern plains (~100 m a.s.l.) bordering IGP. The valley is surrounded by mountains which are approximately 1000-2000 m (a.s.l.). Further north of the Pokhara Valley, within 30 km the elevation gradient increases rapidly to over 7000 m (a.s.l.) or higher (see Figure 1). This steep

168 elevation gradient is conducive for the orographic lift of humid air masses, and thus the valley also

169 receives one of the highest rates of precipitation in Nepal and occasional strong convective updrafts

170 leading to hailstorms and thunderstorms (Aryal et al., 2015). The mixing of dry westerly air masses with

171 heated moist air masses from the Bay of Bengal produces strong convection over the Pokhara Valley,

and thus results in strong updrafts. These strong convective activities are frequent in the pre-monsoon

and monsoon season but do not occur during the winter season.

174 *2.3. Test flight patterns over the Pokhara Valley*

175 Five test flights were conducted in the morning and evening period around Pokhara Valley (83.97° 176 E, 28.19° N, 815 m a.s.l.) with each flight lasting for about 1 to 1.5 h from 5-7 May 2016. The flight 177 pattern was consistently flown over the northwest part of the valley (Figure 1). A typical flight 178 commenced from the Pokhara Regional Airport (818 m a.s.l.) and steadily flew 5-10 km northwest along 179 the Pokhara Valley leaving the direct airport vicinity toward the Himalayas. This was followed by spirals 180 up and down sampling from approximately 1000 to 4000 m, often reaching close to the lower base of 181 the clouds in the free troposphere. Further climbs into the cloud layer were avoided during the test 182 flights.

Figure 1. A typical test flight within the Pokhara Valley on 5 May 2016. The plot is generated using a
 Matlab-Google Earth toolbox (<u>https://www.mathworks.com/matlabcentral/fileexchange/12954-google-</u>
 <u>earth-toolbox</u>). Each dot is a single sample point (sampling frequency of 1Hz); the color of the dot
 indicates the total aerosol number concentration and the value of each color is shown as a color bar.

187 2.4. Data processing and quality

188 The data from all the instruments was synced with the GPS clock, and the PC104 received all the 189 data simultaneously and creates a common time-stamped data file. Prior to each test flight, a zero test 190 was conducted to identify any possible leaks in the sample line.

The collected data from the five test flights went through multiple steps of quality control and quality assurance. Occasionally during the radio communication by the pilot with the ground station or air traffic controller, the CPC and the temperature sensor would record exceedingly high values. This noise is an interference picked up by the sensors from the 5 W radio transmission. The CPC and aethalometer is also sensitive to vibration in the aircraft, especially during upward and downward spiral motion, which may result in flow imbalance in these analyzers. This resulted in random noise segments for a few seconds in the data, which were flagged and not included in the analysis.

198 **3. Results**

- 199 3.1. General meteorology and air quality, aerosol properties in the Pokhara Valley
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201 3.1.1. Local and synoptic meteorology in the Pokhara Valley

202 Climatologically, Pokhara Valley has a humid subtropical climate, characterized by a summer 203 monsoon season from late June to September, preceded by a dry pre-monsoon (March-May, see Figure 204 S2 in the supplement). Dominant winds in the valley are from the southeast and southwest with a strong 205 diurnal variability in the wind speed (Aryal et al., 2015). On a local/regional, the winds in May 2016 were 206 predominantly from the southeast with only occasional strong winds from the southwest (see Figure S3 207 in the supplement, using data available at the regional meteorological station at the Pokhara Airport). 208 During the test flight period (5-7 May 2016), the wind was similar in directionality, with an hourly mean 209 wind speed of 1.8 to 3.0 ms⁻¹, with low wind speed (<2.0 ms⁻¹) before noon, usually from the southeast, followed by stronger winds from the southwest and northwest (>2.4 ms⁻¹) which can continue until late 210 211 night. The increased wind speed in the afternoon could be katabatic in nature as a result of differential 212 heating of the mountain valley slopes and could be linked to pollution transport from surrounding 213 regions (Gautam et al., 2011).

214 Three dominant synoptic meteorology regimes characterize the seasonality of South Asia 215 (Lawrence and Lelieveld, 2010). They are summer (June-September), the winter monsoon (mid-216 November to February) and the monsoon-transition periods, which include the pre-monsoon season 217 (March-May) and post-monsoon season (mid-September to mid-November). These synoptic regimes are 218 also active in the Himalayas, including the Pokhara Valley. The monsoon transition period, during which 219 the test flights were conducted, is characterized by westerlies over 20-30° N at 850 mb and above (see 220 Figure 2). Figure 2 shows the daily wind vector over South Asia for 3, 5, 6 and 7 May 2016 generated 221 using the NCEP NCAR Reanalysis data at 2.5° x 2.5° horizontal resolution. While the reanalysis data can 222 be expected to represent the synoptic-scale phenomena in this region reasonably well, the rough terrain 223 in the Himalayas presents a significant challenge for modeling and the data is thus likely to suffer from 224 biases and other deviations from the observed meteorology (Xie et al., 2007). The wind vector at 850 225 mb in the 20-30° N latitude band was westerly with variable wind speeds in the IGP region near the 226 Himalayan foothills. The wind direction varies diurnally at the 850 mb level, with the wind direction 227 shifting to southwesterly near the Himalayan foothills. Westerlies were also generally prevalent at the 228 500 mb; however, in the mid-latitudes between 40-50° N (Central Asia), a trough and crest-like feature 229 of the westerlies moving from west to east Asia is visible (also observed by Lüthi et al., 2015), which was

230 also present prior to the study period. This wind feature was colder and more humid (see Figure S4 in 231 the supplementary material) than the westerlies observed between 20-30° N. The meandering features 232 (i.e., trough and crest) observed between 40-50° N affects the direction and magnitude of air masses (at 20-30° N) entering Nepal. For instance, the crest feature of the westerly was prevalent over the IGP and 233 Nepal prior to 3 May, transitions into the trough feature after the 3rd and continues during the study 234 235 period. The prevalence of the trough was characterized by the intrusion of wind into lower latitudes as 236 well as into the IGP, also indicated by the change in the temperature and humidity (Figure S4). The 237 intrusions of mid-latitude air masses also influence the westerlies entering Nepal in the 20-30° N sector 238 (Lüthi et al., 2015). As discussed later, variations in the vertical profiles of aerosols above 3000 m (a.s.l.) 239 could be associated with variations observed in these upper layer winds.

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Figure 2. Daily wind vector data at 850 and 500 mb, plotted using the NCEP NCAR reanalysis (2.5° x 2.5°)
 data over South Asia from 1-7 May 2016. The colors indicate the wind speed in ms⁻¹. The plots were

243 generated using the default setup at www.esrl.noaa.gov/psd/data/composites/day/.

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245 3.1.2. Overview of the aerosol properties in the Pokhara Valley during the test flight period

246 The variation in aerosol loading (as reflected by AOD) reveals a strong seasonality in the Pokhara 247 Valley (see supplementary figure S7 for a detailed description of aerosol properties in the Pokhara Valley 248 during 2010-2016). The pre-monsoon season (also the time of the test flight) has the highest AOD values 249 (AOD_{500nm}>0.6: Figure S7, S7a, S7a, and S5) followed by the monsoon low (AOD_{500nm} ~0.2-0.3), most likely 250 due to the wet removal of aerosols. AOD gradually increased (to ~0.4-0.5) during the post-monsoon 251 through winter to the pre-monsoon season. Generally, the increase of total AOD (sum of fine and 252 coarse) over the Pokhara Valley is dominated by fine-mode aerosol particles, except during the pre-253 monsoon and monsoon season, when a substantial fraction of coarse-mode particles were also present. 254 The dominant aerosol in the Pokhara Valley is mostly BC-like (Giles et al., 2012), based on the values of 255 absorption and extinction Angstrom exponent (AAE and EAE at 440-870 nm); however, a substantial 256 seasonal variation was observed from more mixed or dust-like in the pre-monsoon months, to more BC-257 like in the post-monsoon and winter months.

Figure 3. AOD and other data products from the Level1.5 AERONET direct product in the Pokhara Valley
from 1-10 May 2016. The top panel includes AOD at 500nm and the AOD for coarse and fine modes (as
shown in figure above as AOD-C, AOD-F). The bottom panel includes the Ångström Exponent (AE) for

440-870nm, the fine mode fraction and the visibility (km). The visibility data was available from the
synoptic meteorology data available at http://www7.ncdc.noaa.gov/CDO/cdo

263 The aerosol optical properties (columnar) and synoptic meteorology are shown in Figure 3 and 264 Figure S6 (in the supplementary). Prior to the flights days (1-4 May), higher AOD values were recorded in 265 the Pokhara Valley (AOD>1) and dominated by a fine-mode fraction (~0.95). Hazy condition and low 266 visibility (≤ 5 km) was recorded during the period in the valley (see Figure S6). Moving into the flight 267 days, the AOD values decreased below 1, markedly by the drop in the fine-mode fraction and the 268 improvement in haze condition and visibility. The flight day periods were also characterized by the 269 presence of scattered clouds and thunderstorms (with no precipitation) in the afternoon, which also 270 imply conditions for the strong vertical mixing of pollutants. It is indicative from Figure 3 and Figure S7 271 that the presence of high levels of pollution over the region from 1-4 May is followed by a short period 272 of (relatively) cleaner conditions, which also coincides with the changes in the synoptic situation observed in the winds at 500 mb (described in section 3.1.1.) 273

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3.2. Vertical profiles of absorbing aerosols, particle number and size distribution, temperature, anddew point

The five test flights are labeled as F1-5 in Figure 4, except F3 which is shown in the supplement (Fig. S10). F1 and F2 were conducted on 5 May, F3 and F4 on 6 May and F5 on 7 May 2016. Due to limitations of the flight permit, the test flights were conducted remaining within the Pokhara Valley as indicated by Figure 1. Among the five sampling flights, F1, F3, and F5 were morning flights, and F2 and F4 were afternoon flights (for details on sampling flights, see Table T1 in the supplement).

283 Figure 4. Vertical profiles of aerosol species and meteorological parameters during the 5-7 May 2016 284 test flights in the Pokhara Valley using the IKARUS microlight aircraft. The subplot in each row is 285 arranged by (i) size distribution measured by the GRIMM OPS 1.108 (0.3-20 µm), limited to 1 µm in the 286 figure, (ii) Total particle number concentration (also indicated as TPC, Dp >14 nm) measured by the CPC 287 3760, along with absorbing aerosol mass density at 370 nm and 880 nm (iii) temperature (red line, in °C) 288 and dew point (black dots, in °C) and relative humidity (or RH %), (iv) calculated absorption Ångstrom 289 exponent averaged for every 500 meters elevation band. For the size distribution plot, the x-axis 290 represents the optical diameter of the aerosol (nm), and the color bar represents the concentration (10^x 291 in #cm⁻³). Of the five test flights, only F1-2, F4-5 is shown here, F3 is in the supplementary. Number size 292 distribution data from Flight F3 is not available due to the failure of the Grimm's pump during flight

initiation. In each subplot, the y-axis is the altitude above the mean sea level (in m). The origin of the y-axis is at 815 m (a.s.l.).

295 3.2.1. Diurnal variation in the vertical profiles

296 All the vertical aerosol profiles (Figure 3) showed a strong gradient below 2000 m (a.s.l.). Because of 297 the valley geography, with surrounding mountains of about ~2000 m (a.s.l.) or higher, it is likely that the 298 gradient observed below 2000 m (a.s.l.) could be related to emissions from the Pokhara Valley. The 299 development (or dissolution) of the boundary layer during the day clearly influenced the evolution of 300 the aerosol vertical profiles in the Pokhara Valley. The shallow boundary layer in the night, which 301 continued till the morning, led to the accumulation of aerosols below 2000 m (a.s.l.) in the morning (see 302 the morning flights (F1, F3, and F5) and a strong decrease with altitude was observed. For instance, in 303 the morning profiles, the concentrations near the surface (<1000 m a.s.l.) for total particle number concentrations (also indicated as **TPC** in Figure 4) were mostly >10³ cm⁻³, but could reach ~ $3x10^4$ cm⁻³ or 304 305 higher (see F5 in Figure 4), which is attributed to the coupling of the shallow boundary layer and the 306 emissions in the contained valley topography (Mues et al., 2017). Also, all the measured aerosol 307 parameters (number size distribution for particles with diameters between 0.3 and 0.5 μ m), the total 308 particle concentration (>14 nm), and the absorption) vary similarly as a function of the altitude 309 irrespective of the timing of the profiles. The similarity in the vertical concentration gradients of the 310 absorbing aerosol mass concentrations and the aerosol number concentration above 2000 m (a.s.l.) 311 provides evidence of similar emission sources or origins.

312 Within the morning profiles, substantial variations were observed; in F1 (5 May), in addition to the 313 strong gradient below 2000 m (a.s.l.), there is a polluted layer above 3000 m (a.s.l.) which is not evident in F5. The BC concentration was close to 1 μ gm⁻³ up to 4000 m (a.s.l.) for F1 and stayed in that range 314 until F5, where it dropped to about ~0.4 μ gm⁻³. The temperature and humidity profile also showed 315 316 changes between the morning flights; the conditions during F1 are warmer (throughout the profile) and 317 dryer (near the surface), compared to F5. This observed variation in the aerosol vertical profile 318 (including the meteorology) may be indicative of cleaner atmospheric conditions (in terms of aerosol 319 number and absorption) from 5 May to 7 May and could be associated with the arrival of colder 320 airmasses in the Pokhara Valley. The near-surface BC concentrations measured in this study were much 321 lower than surface BC concentrations measured in the pre-monsoon season (2013) in the Kathmandu Valley in the Himalayan foothills (hourly average: \sim 5-40 µgm⁻³, Mues et al. (2017)), but comparable to 322 winter measurements (2004) in Kanpur in the IGP (1-3 min average: \sim 1-7 µgm⁻³, Tripathi et al. (2005)). In 323

Kanpur, Tripathi et al. (2005) observed BC concentrations close to 1 μgm⁻³ up to 2000 m (a.s.l.) and a
sharp gradient below 400 m (a.s.l.), most likely due to a shallow boundary layer in winter.

The elevated polluted air mass in F1 could be an indication of transport related to the mountain valley winds and/or synoptic transport related to the westerlies, common during this season (Gautam et al., 2011;Raatikainen et al., 2014;Marcq et al., 2010). Pre-monsoon airborne measurements over the IGP and near the Himalayan foothills during CALIPEX-2009 found a polluted aerosol layer (2-4x10³ cm⁻³ with a mean size of 0.13 µm diameter) below 4 km (a.s.l), attributed to biomass burning observed during this particular season (Padmakumari et al., 2013).

332 The afternoon profiles (F2: 5 May 2016 and F4: 6 May 2016) in contrast to the corresponding 333 morning profile (F1 and F3) showed a more relatively mixed profile up to about 2500-3000 m, decreased 334 then up to the maximum sampled altitude of just above 4000 m (a.s.l.). For instance, the concentrations 335 of measured aerosol parameters up to 3000 m (a.s.l.) were comparable to the concentrations observed at ~1000 m (a.s.l.). Slight differences exist within the afternoon profiles, which may be related to local 336 337 meteorology (boundary layer evolution) and mountain valley wind circulation in the afternoon. Cloud 338 layers were present during the afternoon flights at and above 4000 m (a.s.l.) in F4 (also indicated by the 339 sharp rise in RH from ca. 3600 m a.s.l.), which may have led to the scavenging of the aerosol by cloud 340 droplets and thus explaining the observed decrease in the measured aerosol parameters.

341 3.2.2. Nature of absorbing aerosols in the Pokhara Valley

342 The absorption at multiple wavelengths was used to calculate the absorption Angstrom exponent (AAE), shown in the right-most subplot in each row of Figure 4. AAE characterizes the wavelength (λ) 343 dependence of absorption coefficient (Absorption coefficient or Abs. coeff = $K\lambda^{-AAE}$, Russell et al. 344 345 (2010); Giles et al. (2012)). On a logarithmic scale, the above power relation between (Absorption 346 coefficient and wavelength) is approximately a straight line (see supplementary S11 for an example case 347 where both power and logarithmic form are plotted). The slope of the straight line is AAE. In our case, 348 all the absorption coefficient measured between the 470 and 880 nm wavelength was used for the calculation of AAE. The mass absorption coefficients (MAC) of 14.5 and 7.77 m² g⁻¹, as prescribed by the 349 350 manufacturer of the aethalometer (Hansen et al., 1984) for wavelength 470 nm and 880 nm, 351 respectively were used to calculate the absorption coefficient (the unit for the absorption coefficient is m⁻¹). The calculated AAE was averaged for each 100 m (a.s.l.), as shown in the figure. The sampling 352 353 resolution for the aethalometer is 2 min (see Table 1), which resulted in no (in a few cases) or few data

354 (after flagging) if smaller height bins were chosen. The AAE profile differed markedly between the 355 morning (F1, F3, and F5) and afternoon (F2 and F4) profiles, with morning profiles showed large 356 variations along the height. Surface AAE (~1000 m a.s.l.) was close to 0.8 to 1.2 for all the flights which 357 indicate the presence of BC from a mix of sources (biomass burning and fossil fuel combustion. A 358 source-diagnostic analysis of C-isotopes of elemental carbon (EC) in TSP (total suspended particulates) 359 collected in Pokhara during April 2013-March 2014 showed that the biomass burning and fossil fuel 360 combustion contributes nearly 50 % each to the (annual average) EC concentration (Li et al., 2016). The 361 AAE values above surface (>1000 m a.s.l.) varied from 0.5 to 2, but mostly fell into the range of 0.9-1.2, 362 which is typically reported for mixed to BC like aerosols from urban and industrial emissions (Russell et 363 al., 2010; Yang et al., 2009; Dumka et al., 2014). AAE<1 could also be indicative of a composite aerosol, 364 where a BC aerosol (or "core") is coated with absorbing or non-absorbing aerosols (Gyawali et al., 365 2009).

Figure 5. Aerosol extinction coefficient (at 532 nm) vertical profile (left) and aerosol type
classification based on the CALIPSO level 2 retrieval (right). Only the CALIPSO overpass over the Pokhara
Valley or nearby locations (such as Kathmandu Valley region, and the region to the west of Pokhara
Valley) is included. The extinction profile is averaged for the region 27-28.5° N latitude, which also
includes the Pokhara Valley.

371 3.2.3. Comparison of the satellite-derived vertical profiles with measurements

372 The measured vertical profiles were also complemented with CALIPSO retrievals over the Pokhara 373 Valley (Figure 5). Level 2 (version 4), cloud and quality screened data were used to generate the 374 vertically resolved extinction (at 532 nm) and aerosol classification. The CALIPSO satellite had only three 375 overpasses over the Pokhara Valley between 1 and 10 May 2016 (the extinction profile lines with circle 376 markers are for the Pokhara Valley). Therefore, the satellite overpasses through nearby regions such as 377 the Kathmandu Valley region to the east and the region to the west of the Pokhara Valley (denoted by WestPV in Figure 5) were also considered. The range of extinction values for the Pokhara Valley (0.15-378 379 0.25 km⁻¹ especially around 2000-4000 m a.s.l.) were similar to pre-monsoon values (0.15-3 km⁻¹) 380 reported in Nainital (a hilly station located ~2000 m (a.s.l.) in India, and 400 km west of the Pokhara Valley) and slightly less than Kanpur, a site in the IGP, about 400 km to the southwest of Pokhara 381 (Dumka et al., 2014). A large extinction (>0.5 km⁻¹) was observed on 1 May 2016 over the Pokhara 382 383 Valley at an altitude of 3-4 km (a.s.l.) which can be attributed to smoke (biomass- related) and polluted 384 dust (a mixture of dust and biomass smoke or urban pollution) as evident by the aerosol type

classification. Aerosols over the IGP and in the proximity of the Himalayan foothills were mainly "Dust"
on 1 May 2016. Although not conclusive, the 9 May aerosol type classification is markedly different from
1 May with the absence of dust in the IGP, and absence of polluted dust or smoke over the Pokhara
Valley.

Figure 6. HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory, Draxler and Hess (1998)) 3
 day back trajectories of air masses arriving at 3 different heights (800 m, 1500 m and 2500 m) from
 above the ground level (AGL~ 815 m a.s.l.) in the Pokhara Valley (28.19° N, 83.98° E) during 5-7 May
 2016. NCEP GDAS (Global Data Assimilation System) Reanalysis data with 1°x1° horizontal resolution
 were used as the input meteorology. The trajectory data is overlayed with the active fire data
 (extracted from the MODIS collection 6 database, available at
 <u>https://firms2.modaps.eosdis.nasa.gov/active_fire</u>. Each green dot with a gray edge is an active fire, and

the strength of the active fire is indicated by the *"frp" value,* which is the fire radiative power inmegawatts.

398 3.2.4. Role of synoptic circulation in modulating aerosol properties over the Pokhara Valley

399 The measured vertical profiles and available satellite data from MODIS (See Figure S8) and CALIPSO 400 suggest that the synoptic-scale circulation were changing during the study period. The changing synoptic 401 circulation also influenced the transport of polluted air into the Pokhara Valley. The regional 402 meteorology station in the Pokhara Valley reported hazy conditions till 5 May 2016(see Figure S6) which 403 disappeared from 6 May 2016 onwards followed by clear days with scattered clouds during the daytime 404 and thunderstorms in the afternoon. The variation in the AOD, AOD-F and Fine Mode Fraction (FMF) 405 from AERONET (only level 1.5 data were available, see Figure S7) also showed that high turbidity in the 406 atmospheric column, dominated by fine-mode aerosols before 5 May 2016 (AOD_{500nm}>2.0, FMF >0.9), 407 which declined sharply after 5 May 2016. The variation in the horizontal visibility (or visual range) 408 measured at the meteorology station in the Pokhara Valley further indicates that the intensity of 409 pollution declined during the study period, especially starting on 5 May 2016.

Three day back trajectories (72 h) were generated using HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) for air masses arriving in the Pokhara Valley at 800 m, 1,500 m and 2,500 m from above ground level (AGL) for the test flight period (see Figure 6). The NCEP GDAS reanalysis data with a 1°x1° horizontal resolution were used as the input meteorology for the trajectories. The majority of air masses (especially at 1500 and 2000 m AGL) were westerly. A high resolution (0.0625° horizontal)

simulation of air mass trajectories during the pre-monsoon period over the Himalayas and Tibetan
Plateau region by Lüthi et al. (2015) also identified synoptic-scale transport (as westerly advection
around 500 mb) and a convection-enabled polluted airmass from the IGP as a major mechanism of
transport of air pollution in the Himalayas. Transport of air pollution by both mechanisms was coupled
with the diurnal expansion of PBL height in the IGP where the trajectory height was similar to planetary
boundary layer (PBL) height thus allowing mixing up of the polluted layer, also observed by Raatikainen
et al. (2014) over Gual Pahari (IGP site) and Mukteswor (Himalayan foothill site).

422 During the study period, the direction of the trajectories varied as the air masses entered Nepal and 423 eventually into the Pokhara Valley. On 5 and 6 May 2016, the air masses (at 1500 and 2000 m AGL) were 424 mostly northwesterly traversing through northern India and western Nepal before entering the Pokhara 425 Valley. A shift in the trajectory direction from north westerly to south westerly was observed on 7 May 426 2016, where the trajectories were moving through central India and the southern foothills into the 427 Pokhara Valley. The observed shift in the trajectories at 1500 and 2500 m AGL was modulated by the 428 synoptic-scale changes in the mid-latitude (over Central Asia) air masses (40-50° N) (Lüthi et al., 2015). 429 The intrusion (in the form of a trough) of the cold and humid air masses from 40-50° N (see Figure 2) 430 into 20-30° N occurred during the study period. As the trough moves eastward, it shifts the synoptic air 431 mass at 20-30° N from northwesterly to southwesterly on 7 May 2016. The elevated polluted layer on 5 432 and 6 May 2016 (Figure 4) could be the result of this modulation of the westerly. The northwesterly 433 airmass entered Nepal via Northern India, where MODIS retrievals showed a high aerosol loading (See 434 Figure S8), which could be mainly attributed to the numerous biomass fire events (See Figure S9) 435 observed in North India. In addition, numerous forest fires were also reported in western Nepal during 436 the same period. However, the absorption signal from the flight measurement does not clearly show 437 higher absorption at shorter wavelengths compared to absorption at 880 or longer wavelengths. This 438 also implies that the observed elevated polluted layer in the Pokhara Valley is not entirely due to the 439 biomass burning plume intercepted by the westerlies or north-westerlies. As the airmass origin shifts to 440 southwesterly on 7 May 2016 (detected during flight F5), the synoptic air mass bypassed the high AOD 441 loading over north India and contained the cold and relatively clean air from Central Asia. This resulted 442 in the disappearance of the polluted layer over 2000 m (a.s.l.) during flight F5.

443 4. Conclusion

This paper provides an overview of the pre-monsoon airborne measurement carried out with a microlight aircraft platform in the Pokhara Valley in Nepal, the first-of-their-kind airborne aerosol

measurements in the Himalayan foothill region. The objective of the overall airborne campaign in the 446 447 Himalayan region was to quantify the vertical distribution of aerosols over a polluted mountain valley 448 region, as well as to measure the extent of regional transport into the Himalayas. In this paper, 449 measurements from the test flights during May 2016 are summarized. These mainly include vertical 450 profiles of aerosol number and size distribution, multi-wavelength aerosol absorption, black carbon, 451 total particle concentration, and meteorological variables. The instrument package, designed for a 452 microlight sampling was fitted to an IKARUS-C42 microlight aircraft. A total of five test flights were 453 conducted between 5 and 7 May 2016, including morning and evening flights for about 1-1.5 h each, as 454 well as vertical spirals to characterize vertical profiles of aerosols and meteorological parameters

455 The results presented in this paper should be considered as a pilot study mapping out the 456 aerosol concentrations and their interactions with meteorological processes in the Pokhara Valley due 457 to the limited flight time. In all the measured flights, the vertical profiles of aerosol parameters showed 458 strong gradients along the atmospheric column. The observed total number concentration gradient was 459 strongly influenced by the mountain valley boundary layer, which resulted in a sharp gradient below 460 about 1500-2000 m (a.s.l.). The increase of boundary layer height contributed to the differences in the 461 morning and afternoon profiles. Similar vertical profiles of BC concentrations and aerosol total particle 462 number concentrations provided evidence of common emission sources or co-located origins. The 463 observed BC concentration near the surface (~ 1000 m a.s.l.) was much lower than pre-monsoon BC 464 concentrations measured in the Kathmandu Valley but comparable to values reported during the winter 465 season in Kanpur in the IGP. The AAE estimates near the surface, based on the absorption value, fell in 466 the range of 0.9-1.2, which indicates the presence of *BC like* and mixed (dust, urban, biomass) aerosols. 467 An elevated polluted layer was observed at around 3 km (a.s.l.) over the Pokhara Valley during this 468 study. Characterized by a strong presence of dust in the IGP and polluted continental airmasses over the 469 Pokhara Valley, the polluted layer could be linked with the westerly synoptic circulations and regional 470 transport from the IGP and surrounding regions. The direction of the synoptic transport entering the 471 Himalayan foothills and into Pokhara Valley, however, was influenced by the Westerlies at mid-latitudes 472 (40-50° N). The extent of transport can be better quantified with regional airborne measurements along 473 the south-north transect through the region between the IGP and the Himalayan foothills at high 474 altitudes in the Himalayas, including the Pokhara Valley. We will explore the extent of such regional 475 transport in a subsequent publication that will be primarily based on the airborne measurements in 476 phase II (December 2016- January 2017) in the Pokhara Valley and in the surrounding region. The

- 477 subsequent paper will also characterize the extent of vertical transport from three different mountain
- 478 valleys located at different elevations along the south-north transect.
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- 645

647 List of Tables

648 Table 1. Instrument package deployed in the microlight aircraft

erosol particle umber size stribution (0.3 -) μm)	GRIMM 1.108	Light scattering	resolution 6 s
erosol particle umber size stribution (0.3 -) μm)	GRIMM 1.108	Light scattering	6 s
umber size stribution (0.3 -) μm)			
stribution (0.3 -) μm)			
) μm)			
otal particle	TSI CPC 3760	Condensation/light	1 s
umber		scattering	
oncentration (>14			
n)			
erosol spectral	Magee AE42	7 wavelengths, light	2 min
osorption		attenuation	
ew point sensor	METEOLABOR, TPS3	Chilled Mirror	1 Hz
emperature	Thermocouple	-	1 Hz
ata acquisition	PC 104+ GPS		
stem			
ower supply	Aircraft battery pack,	12 V, >15 AH	
	LiFEPO ₄ battery		
	tal particle mber ncentration (>14 n) rosol spectral sorption w point sensor mperature ta acquisition stem wer supply	tal particle TSI CPC 3760 mber ncentration (>14 n) rosol spectral Magee AE42 sorption w point sensor METEOLABOR, TPS3 mperature Thermocouple ta acquisition PC 104+ GPS stem wer supply Aircraft battery pack, LiFEPO4 battery	tal particle TSI CPC 3760 Condensation/light mber Scattering ncentration (>14 n) rosol spectral Magee AE42 7 wavelengths, light sorption TEEOLABOR, TPS3 Chilled Mirror mperature Thermocouple - Ita acquisition PC 104+ GPS stem wer supply Aircraft battery pack, 12 V, >15 AH LiFEPO4 battery



- **Figure 1.** A typical test flight within the Pokhara Valley on 5 May 2016. The plot is generated using a
- 656 Matlab-Google Earth toolbox (<u>https://www.mathworks.com/matlabcentral/fileexchange/12954-google-</u>
- 657 earth-toolbox). Each dot is a single sample point (sampling frequency of 1Hz) and the color of the dot
- 658 indicates the total aerosol number concentration (in # cm⁻³) and the value of each color is shown as a
- 659 color bar.



Figure 2. Daily wind vector data at 850 and 500 mb plotted using the NCEP NCAR reanalysis (2.5° x 2.5°) data over South Asia from 1-7 May

- 662 2016. The colors indicate the wind speed in ms⁻¹. The plots were generated using the default set-up at
- 663 www.esrl.noaa.gov/psd/data/composites/day/.



Figure 3. AOD and other data products from the Level1.5 AERONET direct product in the Pokhara Valley from 1-10 May 2016. The top panel

667 includes AOD at 500nm and AOD coarse and fine (as shown in figure above as AOD-C, AOD-F). The bottom panel includes angström Exponent

668 (AE) 440-870nm, fine mode fraction and visibility (km). The visibility data was available from the synoptic meteorology data available at the

669 http://www7.ncdc.noaa.gov/CDO/cdo/

- 671 Figure 4







the IKARUS microlight aircraft. The subplot in each row is arranged by (i) Aerosol number size distribution measured by the Grimm OPC 1.108

- 691 (0.3-20 μm), limited to 1 μm in the figure, (ii) Total particle number concentration (also indicated as *TPC*, Dp >14 nm) measured by the CPC 3760
- 692 and absorption aerosol at 370 nm and 880 nm (iii) temperature (°C) and dew point (black dot, in °C) and relative humidity (or RH %), (iv)

- 693 calculated absorption Ångstrom exponent averaged for every 500 meters elevation band. For the size distribution plot, the x-axis represents the
- optical diameter of the aerosol (nm), and the color bar represents the concentration (10^x in #cm⁻³). Of the five test flights, only F1-2, F4-5 is
- 695 shown here, F3 is in the supplementary. Number size distribution data from Flight F3 is not available due to the failure of the Grimm's pump
- 696 during flight initiation. In each subplot, the y-axis is the altitude above the mean sea level (in m). The origin of the y-axis is at 815 m (a.s.l.).
- 697
- 698
- 699



- **Figure 5**. Aerosol extinction coefficient (at 532 nm) vertical profile (left) and aerosol type classification based on the CALIPSO level 2 retrieval
- 702 (right) available during May 2016. Only the CALIPSO overpass over the Pokhara Valley or nearby locations (such as Kathmandu Valley region,
- and the region to west of Pokhara Valley) are included). The extinction profile is averaged for the region 27-28.5° N latitude which also includes
- the Pokhara Valley.
- 705



- **Figure 6**. HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) 3 day back trajectories of air masses arriving at 3 different heights
- 707 (800 m, 1500 m and 2500 m) from above the ground level (AGL~ 815 m a.s.l.) in the Pokhara Valley (28.19° N, 83.98° E) during 5-7 May 2016.
- 708 NCEP GDAS (Global Data Assimilation System) Reanalysis data with 1°x1° horizontal resolution were used as the input meteorology. The
- trajectory data is overlayed with the active fire data (extracted from the MODIS collection 6 database, available at
- 710 <u>https://firms2.modaps.eosdis.nasa.gov/active_fire/</u>. Each green dot with a gray edge is an active fire, and the strength of the active fire is
- 711 indicated by the *"frp" value,* which is the fire radiative power in megawatts.