In-depth study of the formation processes of single atmospheric particles in the southeastern margin of Tibetan Plateau

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Abstract

The unique geographical location of the Tibetan Plateau (TP) plays an important role in regulating global climate change, but the impacts of the chemical components and atmospheric processing on the size distribution and mixing state of individual particles are rarely explored in the southeastern margin of the TP, which is a transport channel for pollutants from Southeast Asia to the TP during the pre-monsoon season. Thus a single-particle aerosol mass spectrometer (SPAMS) was deployed to investigate how the local emissions of chemical composition interact with the transporting particles and assess the mixing state of different particle types and secondary formation in this study. The TP particles were classified into six main types: the rich-potassium (rich-K) type was the largest fraction of the total particles (30.9%), followed by the biomass burning (BB) type (18.7%). Most particle types were mainly transported from the surroundings of the sampling site and Sino-Myanmar border; but the air masses from northeastern India and Myanmar show a greater impact on the number fraction of BB (31.7%) and Dust (18.2%) types, respectively. Then, the two episodes with high particle concentrations showed that the differences in the meteorological conditions in the same trajectory clusters could cause significant changes in chemical components, especially the Dust and EC-aged types, which changed by a sum of 93.6% and 72.0%, respectively. Ammonium and Dust particles distribute at a relatively larger size (~ 600 nm), but the size peak of other types is present at ~ 440 nm. Compared with the abundant sulfate ($^{97}HSO_4^-$), the low nitrate ($^{62}NO_3^-$) internally mixed in TP particles is mainly due to the fact that nitrate is more volatilized during the transport process. The formation mechanism of secondary speciation demonstrate that the formation capacity of atmospheric oxidation is presumably affected by the convective transmission and the regional transport in TP. However, the relative humidity (RH) could significantly promote the formation of secondary species, especially $^{97}HSO_4^-$ and $^{18}NH_4^+$. This study provides insights that can improve the knowledge of particle composition and size, mixing state, and aging mechanism at high time resolution over the TP region.

Keywords

Southeastern Tibetan Plateau, Individual particles, Chemical characteristics, mixing state, Secondary formation

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1 Introduction

Atmospheric aerosols have complex components and sources and can be coated with inorganic or organic materials during transport and atmospheric processing (Crippa et al., 2013). After further coating through coagulation, condensation and photochemical oxidation, its sizes, chemical compositions, mixing states and optical properties would change greatly, leading to its influence in the atmosphere more uncertain (Jacobson, 2002; Zaveri et al., 2010; Matsui, 2016; Budisulistiorini et al., 2017; Ma et al., 2012). Currently, the influences of the complex chemical components on aerosol size and mixing state show large regional differences due to the variations in the pollution sources, atmospheric formation mechanism and meteorological conditions, which have been widely studied in an urban area at a low altitude (Pratt et al., 2011; Liu et al., 2020a; Xu et al., 2017; Wang et al., 2022). However, previous study found that the migration or formation of low-volatile component (such as nitrate and organic matter) could effectively be reduced due to evaporation during the upward transportation process (Liu et al., 2020b), which further alter the chemical compositions and the particle sizes. The transportation of the aerosols to a relatively cleaner environment prevails the formation of secondary chemicals at a high altitude (Liu et al., 2020b). Therefore, a comprehensive investigation of the detailed characteristic of aerosol formation and mixing states is required to understand their environmental effects in low-, and high-altitude.

As a typical high-altitude region, the Tibetan Plateau (TP) has the highest and largest mountain area in the world, which is the most sensitive and obvious indicator of climate change in the entire Asian continent (Liu et al., 2017; Chen and Bordoni, 2014; Immerzeel et al., 2010). In recent decades, many studies have shown that the melting and retreat of glaciers in the TP regions is accelerating, and the main reason is attributable to anthropogenic emissions, such as greenhouse gases and aerosols (Luo et al., 2020; Hua et al., 2019). Atmospheric aerosols also can act as cloud condensation nuclei to impact the local hydrological cycles and monsoon patterns by changing the microphysical properties and life span of clouds (Qian et al., 2011; Gettelman et al., 2013; Kumar et al., 2017). The southern part of the TP is always affected by the transport of more polluted air from South Asia along the mountain valleys, especially during the pre-monsoon (i.e., March-May) with the

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删除[Li]: The ability of aerosol particles to affect atmospheric conditions is dependent on their sizes, chemical compositions, and mixing states (Mc Figgans et al., 2006; Dusek et al., 2 ⋯

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southwest prevailing wind (Chan et al., 2017; Zhao et al., 2017; Han et al., 2020). Most studies have focused on the optical characteristics within the TP; however, only a few research has been conducted on aerosol components.

Present aerosol components studies conducted in TP mostly focus on exploring the influence of light-absorbing carbon aerosols and dust particles on climate change by optical or offline sampling methods (e.g., Wang et al., 2019a; Liu et al., 2021). There is a lack of studies on the chemical composition, mixing states, and formation mechanism of aerosols in the southeast margin and even the entire TP, especially using high-time resolved measurements. Although time-integrated sampling with filter collection followed by laboratory analyses has been widely adopted for the chemical characterization of aerosols (Li et al., 2022a; Shen et al., 2015; Zhang et al., 2013). Drawbacks of the traditional approach include low time resolution, high detection limit, and time- and labor-intensive procedures. More advanced aerosol online measurement equipment with high-time resolution, such as the aerosol chemical speciation monitor (ACSM) and aerosol mass spectrometer (AMS) (Ng et al., 2011; Canagaratna et al., 2007) are mainly used to achieve online observation datasets of non-refractory submicron aerosol (including the mass concentration of sulfate, nitrate, ammonium, chloride, and organic; and their corresponding mass spectral). This is beneficial to recognize the dynamic processes of source emission in the atmosphere (Du et al., 2015; Zhang et al., 2019a). At the same time, aerosol time-of-flight mass spectrometry (ATOFMS) (Dall'Osto et al., 2014), and single particle aerosol mass spectrometer (SPAMS) (Zhang et al., 2020), are popular for characterizing atmospheric individual particles. These devices can determine the chemical composition and size distribution of the particles in detail, such as the dynamic processes of chemical aging, mixing state, and transport of the aerosols (Liang et al., 2022; Li et al., 2022b; Zhang et al., 2019b). To the best knowledge, the advanced measurement device has not yet been applied for the studies conducted in TP, leading to a lack of in-depth research on the PM$_{2.5}$ pollution in TP, especially in the southeastern margin, which hinders our understanding of the distribution characteristics and formation mechanism of aerosol components in high-altitude regions.

The southeastern margin of the TP is an important transitional zone between the high-altitude TP and the low-altitude Yungui Plateau (Wang et al., 2019a; Zhao et al., 2017),

an ideal place for investigating the impacts of pollutants transport and formation in the high-altitude zone. In this study, continuous field observation of individual particles (SPAMS) was deployed on the southeastern margin of the TP during the pre-monsoon period, to (i) investigate the changes of chemical characteristics between transport and local fine particles during pre-monsoon, (ii) determine the particle size distributions, and the mixing states of different particle types, and (iii) assess the contributions of photooxidation and aqueous reaction to the formation of the secondary species. These results would expand our understanding of the chemical components, size distribution, mixing state, and aging pathways of aerosols in the high-altitude areas in the TP and surrounding areas.

2 Methodology

2.1 Sampling site

Intensive 1-month field observation was deployed at the rooftop (∼ 10 m above ground level) of the Lijiang Astronomical Station, Chinese Academy of Sciences (3260 m above sea level; 26°41′24″N, 100°10′48″E), Gaomeigu County, Yunnan Province, China, during the pre-monsoon period (from April 14th to May 13th, 2018). The nearest residential area is the Gaomeigu village (3–5 km away) with a small population size of 113 residents in 27 households. Villagers earn a living by farming (e.g., potato and autumn rape), and biomass is the major domestic fuel (Li et al., 2016). The site is surrounded by rural and mountainous areas and has no obvious industry or traffic emissions. During the observation period, the average temperature (T) and relative humidity (RH) are 8.4 ± 3.1℃ and 69% ± 21%, respectively. The wind speed (WS) is 2.2 ± 1.2 m·s^{-1} with the prevailing wind in the north and northeastern (Fig. S1).

2.2 On-line instrument

A detailed operational principle and the calibrations of the single-particle aerosol mass spectrometer (SPAMS, Hexin Analytical Instrument Co., Ltd., Guangzhou, China) has been described elsewhere (Li et al., 2011). Briefly, individual particles are drawn into SPAMS through a critical orifice. The particles are focused and accelerated, then aerodynamically sized by two continuous diode Nd: YAG laser beams (532 nm), subsequently desorbed and

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ionized by a pulsed laser (266 nm) triggered exactly based on the velocity of the specific particle. The generated of positive and negative molecular fragments are recorded with the corresponding size of individual particles. In summary, a velocity, a detection time, and an ion mass spectrum are recorded for each ionized particle, while there is no mass spectrum for not ionized particles. The velocity could be converted to d_{va} based on a calibration using polystyrene latex spheres (PSL, Thermo Scientific Corp., Palo Alto, USA) with predefined sizes. The average ambient pressure is 690 hPa (in a range of 685–694 hPa) during the measurements and calibration. Particles measured by SPAMS mostly are within the size range of vacuum aerodynamic diameter (d_{va}) 0.2–2.0 μm. A hollow, silicone dryer was installed in front of the inlet. This reduces the uncertainty of particle collection efficiency due to the changes of humidity in sampled airs.

Meteorological parameters, including the planetary boundary layer (PBL), temperature (℃), RH (%), WS (m·s^{-1}), and wind direction (WD) were continuously achieved using an automatic weather station (Model MAWS201, Vaisala HydroMet, Helsinki, Finland) at a time resolution of 5 min. Gaseous concentrations (ppbv) were obtained using a multiple gas analyzer (Thermo Scientific Corp.), including ozone (O$_3$, model 49i) and nitrogen oxides (NO$_x$, model 42i) in a 5-min resolution. The SPAMS and gas analyzers are co-located in the same position, while the weather station was uncovered outside ~5 m from the sampling house. Time series of SPAMS particles, gaseous concentrations (NO, NO$_x$, O$_3$, and CO) and meteorological parameters (PBL, temperature, RH, WD, and WS) were shown in Fig. S2.

2.3 Individual particle classification

During the observation period, a total of 461,876 ambient particles with the size (d_{va}) of 0.2–2.0 μm were collected, including 55,583 in Episode I (E1; from April 18th 08:00 local time (LT) to April 19th 08:00 LT) and 62,110 in Episode II (E2; from April 26th 17:00 LT to April 28th 02:00 LT). The analyzed particles are classified into 1,557 groups using an adaptive resonance theory neural network (ART-2a) with a vigilance factor of 0.8, a learning rate of 0.05, and 20 iterations (Song et al., 1999). Finally, eight major particle clusters [i.e., potassium-rich (rich-K), biomass burning (BB), organic carbon (OC), Ammonium, aged element carbon (EC-aged), Dust, sodium (Na)-potassium (K)-containing (NaK-SN), and iron

(Fe)-lead (Pb)-containing (Metal)] with distinct chemical patterns were manually combined, which represent ∼99.7 % of the population of the detected particles. The remaining particles are grouped as "Other". The characteristics of the positive and negative mass spectra (MS) of each particle type are shown in Fig. S3. A detailed description of classification criteria for individual particles and the characteristic ion fragments for each particle type can be found in Text S1. The criteria used for searching the some secondary species in the SPAMS datasets are summarized in Table S2.

2.4 Trajectory-related analysis

To determine the influence of regional transport on different particles at Gaomeigu, the trajectory clusters analysis was carried out using the 72-h backward air mass trajectories at arrival heights of 500 m above ground level. The trajectories were calculated with the Hybrid Single-Particle Lagrangian Integrated Trajectory model (Draxler and Hess, 1998), and the meteorological data were obtained from the Global Data Assimilation System (GDAS; ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1, last access: 6 April, 2022). The cluster analysis employs a Euclidean-oriented distance definition to differentiate and cluster the major spatial features of the inputting trajectories. Details of the trajectory clustering method can be found in Sirois and Bottenheim (1995). To investigate the effects of transport on the chemical characteristic of the individual particles, trajectories with particle number concentrations high than the 75th percentile are considered as pollution (Liu et al., 2021).

3   Results and Discussion

3.1 Characteristics of particle composition

Table 1 summarizes the numbers of concentrations, relative percentages, and characteristic ions of each particle type. The most dominant particle type in Gaomeigu during pre-monsoon is rich-K, accounting for an average of 30.9% of the total resolved particles, followed by BB (18.7%), OC (12.8%), Ammonium (11.9%), EC-aged (10.9%), and Dust (10.7%). Similar to the results of some studies in urban areas, rich-K or carbonaceous-containing type is the dominant particle type (15-50%) (Xu et al., 2018; Wang et al., 2019b; Li et al., 2022). The difference is that few researchers can capture the high

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proportion of Ammonium particles as shown in this study (Shen et al., 2017; Xu et al., 2018), which is ascribed to the conversion of ammonia (NH_3) precursor emitted from large-scale agricultural activities and mountain forest (Engling et al., 2011; Li et al., 2013). It is necessary to point out that 60% of Ammonium particles contain signals of amine fragment (m/z 58, $C_2H_5NH=CH_2^+$), implying their similar formation pathway (Zhang et al., 2012). Moreover, the amine-containing particle represented 12.5% of the total ambient particles, which is significantly higher than that in some urban areas at low altitudes (around 2%) (Cahi et al., 2012; Zhang et al., 2015; Li et al., 2017) but is comparable to that at observed sites with high RH, or during fog and cloud events at a high altitude (> 9%) (Roth et al., 2016; Lin et al., 2019). This suggests that the formation of amines under high RH and fog conditions might exist in the Gaomeigu area (with an altitude of 3260 m), for example, the high relative fraction of amine-containing particle corresponds to a high RH (Fig. S4), and the existence of amine sources govern the ammonium formations (Bi et al., 2016; Rehbein et al., 2011). The relatively larger fraction of Dust particles is related to the short-time occurrences of dust events in spring (Fig. S5), leading to a wide contribution ranging between 10% and 70% in the period of 19:00 LT on April 16th to 10:00 LT on April 17th.

Fig. 1 shows the diurnal variations of each particle type. The rich-K, BB, and OC particles decrease after midnight until 06:00 LT, possibly explained by the curtailment of local traffic and biomass-burning activities at nighttime even though both the planetary boundary layer (PBL) height and WS decrease (Fig. S6). Then, their intensities rapidly increase in the morning (around 07:00 LT) due to more pollutants from biomass burning and traffic emissions at the upwind region. The increases of PBL height and WS also lead to the air pollutants transported from the surrounding environment to the sampling site (Liu et al., 2021). At 11:00 LT, the particle counts sharply decrease till 16:00–17:00 LT, caused by the pollutant dispersion with the increases of the PBL height and WS. Increasing trends are observed after 17:00 LT due to the reduction of PBL height and WS. In contrast, the Ammonium, EC-aged, and Dust particles show a unimodal pattern of the daily diurnal variation (Fig. 1d–f). From 00:00 to 06:00 LT, minor fluctuation of particle concentrations of Ammonium, EC-aged, and Dust is observed for these particle types. After that, their levels continuously elevate until ~11:00 LT due to the regional transport, traffic emission, and

fugitive dust (Text S2). While the PBL height and WS increase continuously, the Ammonium, EC-aged, and Dust particles begin to decline from 12:00 to 17:00 LT. The subsequent increases of these three particles after 17:00 LT are attributed to the reduction of PBL height, as a result of the accumulation of pollutants in the near-surface atmosphere.

Based on the transport pathways, four air mass clusters are identified to investigate the effect of regional transport on the major particle types (i.e., rich-K, BB, OC, Ammonium, EC-aged, and Dust) (Fig. 2). Cluster 1, 3 and 4 originated from northeastern Myanmar, accounting for 59.8%, 33.2% and 4.6% of the total trajectories, respectively. Cluster 1 had an average percentage of 32.7%, 18.5%, 12.0%, 12.5%, 11.1%, and 8.9%, respectively, on the rich-K, BB, OC, Ammonium, EC-aged, and Dust particles (Table S1). Clusters 3 and 4 have comparable contributions of BB, OC, Ammonium, and EC-aged to those of Cluster 1, but with a high contribution of Dust, which approximately 16.6% of Clusters 3 and 4 are referred to as dust pollution. The diurnal variations of the BB and OC fractions are similar which rapidly elevate at 07:00 LT (Fig. S7) due to the increased contribution of biomass burning and vehicle emissions from Cluster 1, Ammonium and EC-aged particles (peak at 07:00 LT) caused by the effect of Cluster 1 and 3 together. The similar diurnal trend of Clusters 3 and 4 are both associated with dust contributions, which decrease at 04:00 LT and increase at noon. The increased nighttime particles could be attributed to the pollutant accumulation with the decreased PBL height. Cluster 2 originate from northeastern India and passes over Bangladesh. This cluster accounts for only 2.4 % of the total trajectories, in which ~30.8% and ~35.9% are mainly associated with the rich-K and BB particles, respectively. Even though Clusters 2 and 4 are composed of a small fraction of total trajectories (2.4% and 4.6 %, respectively), BB and dust particles are identified as the major pollutants, suggesting significant influences from India and northeastern Myanmar during the campaign.

A more in-depth investigation of the characteristics of the main particle types in the southeastern Tibet Plateau was conducted during two episode periods when the number concentration of particles was high (i.e., E1: from 08:00 LT April 18th to 08:00 LT April 19th, 2018; E2: 17:00 LT April 26th to 02:00 LT April 28th, 2018) (Fig. S5). Even though the two episodes are contributed by Cluster 1, the chemical components show significant differences

(Table 1). During E1, the average fractions of the rich-K, BB, OC, Ammonium, EC-aged, and Dust particle are 29.0%, 11.5%, 8.1%, 17.5%, 10.0% and 20.3%, respectively, different from 39.3%, 14.2%, 10.0%, 13.5%, 17.2%, and 1.3% respectively, during E2. It can be seen that the Dust particle is the major changed factor, which is 93.6% lower during E2 than E1, whereas the EC-aged particle shows a reversible of 72.0% higher during E2. The rich-K, BB, and, OC particles show 22.9%-35.5% differences between the two episode periods. For the air mass clusters (Fig. S8), E1 and E2 exhibit minor differences, mostly originating from northern Myanmar and the Sino-Burmese border, but not identical regions. The Dust particles that are much lower during E2 than E1 could be explained by higher WS (on average of 2.7 ± 1.0 m/s versus 0.4 ± 0.5 m/s) (Fig. S8) and PBL height (771 ± 717 m versus 560 ± 549 m) (Fig. S9). The Dust particles are mainly formed by re-suspension in the local areas. In addition, the quick thrown-up dust belongs to more coarse size particles, which are out of the detection range of the SPAMS. However, due to the larger dust particles deposited more easily under the low WS and the stagnant air conditions during E1, more suspended dust particles of small size fall in the detection range of SPAMS. Moreover, the increased PBL height and WS could speed up the transportation of pollutants from multiple sources (e.g., traffic and biomass burning emissions) to the observation site, leading to elevate the fraction of EC-aged, rich-K, BB, and OC particles. The decreased Ammonium particle during E2 is potentially explained by the reductions in the secondary pollutant formation with declines of RH (from 73.9% ± 23.9% to 53.1% ± 14.9%), the oxidation capacities [O_3: from 82.3 ± 5.5 ppbv to 76.8 ± 8.4 ppbv; and NO_x: from, 3.9 ± 0.8 to 2.7 ± 0.8 ppb), in comparison to those during E1.

3.2 Characteristics of size distribution and mixing state

The aerodynamic size distributions of all particle types are shown in Fig. 3. According to the characteristics of the average MS (Text S1 and Fig. S3), rich-K, BB, and OC, and EC-aged particles originated from the similar sources of vehicle emission or solid-fuel combustion. Their size distribution thus presents within a small-scale (~440 nm) (Fig. 3a). However, the relative percentage of each particle type is distinct with different size ranges, possibly due to the unique atmospheric processing. For example, as shown in Fig. 3b, the proportions of

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rich-K and BB types increases along with the increase in particle size from 200 to 420 nm, but then decrease. OC and EC-aged types are mainly distributed in relatively small sizes, and their proportions gradually decrease when the size ranges become lager. Ammonium and Dust types are mainly distributed in large sizes of ~600 nm (Fig. 3a). The proportion of Ammonium particles gradually increases with the increase of particle size and peaks at 740 nm, the relatively large size distribution is ascribed to the intense atmospheric aging during regional transport (Text S1). The proportion of Dust particles gradually increases with a size > 560 nm and peaks at 1.48 μm. This is consistent with the fact that dust is a coarse particle, generally formed at the roadside and fly ash.

Compared with the total particle size distribution, the peak values of the six main particle types show minor differences (< 80 nm) during the two different episode periods (Fig. S10a). In Fig. S10b, a relatively high proportion of the rich-K and BB particles exhibit bimodal distributions, while peaks at < 300 nm are affected by the primary emissions and > 300 nm are associated with the aging process (Li et al., 2022b; Bi et al., 2011). Hence, the percentage of the six particle types distribute in wider size ranges during E2 than during E1, due to the more intensive atmospheric aging. Relatively greater fluctuation for the large-size fractions (> 1.1 μm) could be explained by the low particle concentration (a number less than 20). It should be pointed out that further application of this method would require a co-located particle-sizing instrument to scale the size-resolved particle detection efficiency. Both particle composition and size-dependent are the predominant impacting factors on the particle detection efficiency of the SPAMS (Wenzel et al., 2003; Yang et al., 2017; Healy et al., 2013).

To investigate the mixing state of the secondary species in the six main particle types, the number fractions of six secondary markers ($^{97}HSO_4^-$, $^{195}H(HSO_4)_2^-$, $^{62}NO_3^-$, $^{18}NH_4^+$, $^{58}C_2H_5NHCH_2^+$ and $^{89}HC_2O_4^-$) are selected (Fig. 4). The presences of amine (m/z $^{58}C_2H_5NHCH_2^+$) and sulfuric acid (m/z $^{195}H(HSO_4)_2^-$) signals are possibly indicative of the water uptake (Chen et al., 2019) and acidic property of the particles (Rehbein et al., 2011), respectively. The mixing states are calculated by the ratio of the number concentration of the selected ions to each particle type.

The most abundant of $^{97}HSO_4^-$ and $^{18}NH_4^+$ fraction are seen in Ammonium (99% and

306 94%, *respectively*) and EC-aged (92% and 31%, *respectively*) particles, whereas much low

307 fraction of $^{62}NO_3^-$ is found (2% and 7%, *respectively*). These suggest that ammonium sulfate

308 is not a predominant form instead of ammonium nitrate (Zhang et al., 2013). The high

309 contribution of sulfate in EC-containing particles also suggests a significant influence of

310 anthropogenically emitted sulfate precursors (e.g., SO_2) on the aging of EC-containing

311 particles at the high altitude (Peng et al., 2016; Zhang et al., 2017a). Meanwhile, relatively

312 high number fractions of $^{195}H(HSO_4)_2^-$ and $^{58}C_2H_5NHCH_2^\pm$ are also observed in Ammonium

313 (63% and 60%) and EC-aged (4% and 19%) particles. These abundant mixtures potentially

314 represent the high hygroscopicity of Ammonium and EC-aged particles, and their ability to

315 neutralize the acidic particles of Ammonium particle (Sorooshian et al., 2007). Then, a

316 moderate fraction of $^{97}HSO_4^-$ and $^{18}NH_4^+$ are seen on the rich-K (65%, 7%) and OC (56%,

317 4%) particles. In contrast, more $^{62}NO_3^-$ contribute to the rich-K (38%) and OC (68%)

318 particles, mainly affected by vehicle emissions (Text S1). Following BB (18%) and Dust (6%)

319 particles are found in a relatively low number fraction of $^{97}HSO_4^-$, while the moderate $^{62}NO_3^-$

320 accounts for 45% of the BB particle but only 3% of the Dust particle. Besides, the number

321 fraction of $^{18}NH_4^+$ is minor (<1%), which suggests the aging degree of BB and Dust particles

322 is relatively low. In addition, oxalate ($^{89}HC_2O_4^-$), a representative component of secondary

323 organic formation, is mainly mixed with BB (13%) and rich-K (12%) particles. This is

324 because the substantial precursors of oxalic acid, including acetate ($^{59}C_2H_3O_2^-$),

325 methylglyoxal ($^{71}C_3H_3O_2^-$), glyoxylate ($^{73}C_2HO_3^-$), are emitted from biomass burning, while

326 oxalate heterogeneously formed in BB related particles (Zhang et al., 2019b; Zauscher et al.,

327 2013). A relatively low fraction (<5%) of the oxalate-containing particles in OC, Ammonium,

328 EC-aged and Dust particles potentially limited by the contributions of precursor oxalic acid.

329 Compared to the individual particle mixing state in urban or suburban areas that are

330 located close to emission sources (Chen et al., 2016; Dall'Osto and Harrison, 2012; Zhang et

331 al., 2017a; Li et al., 2022b), the high fractions of sulfate and ammonium at the high altitude

332 area demonstrate a high degree of aging of the individual particles, whereas the low fraction

333 of nitrate with high volatility indicates its loss during transportation processing.

334 The number fractions of six markers in the four clusters were used to further investigate

335 the impacts of regional transport. As shown in Fig. 5a and c, the dominant mixing ion types in

each particles (except for Dust type) are similar among the four Clusters. For Cluster 1, the number fractions of $^{97}HSO_4^-$ and $^{89}HC_2O_4^-$ have larger values in five particle types (except for Dust type) than those in other clusters. Similar to Cluster 1, 3 and 4 are impacted by regional transport from northeastern Myanmar. Moreover, the fractions of the six markers are similar in in OC, Ammonium, and EC-aged types. However, $^{97}HSO_4^-$ is decreased in rich-K, BB and Dust types, while $^{62}NO_3^-$ is increased in rich-K and decreased in Dust types. As discussed in Section 3.1, these phenomena demonstrate that the aging degree of Cluster 3 and 4 might be lower than that of Cluster 1. For Cluster 2, the fraction of $^{97}HSO_4^-$ is obviously decreased in rich-K, BB and EC-aged types but slightly increased in Dust type (Fig. 5f). Such pattern inverse the observations in rich-K, OC and Dust types for $^{62}NO_3^-$ ions. These variations in Cluster 2 are more likely due to influences of biomass-burning activities from the surrounding area of the sampling site, rather than regional transport. Furthermore, Cluster 2 is associated with regional transport from northeastern India along the afternoon to nighttime (from 15:00 LT on 11 May to 07:00 LT on 12 May), which is favorable to the nitrate formation N_2O_5 by heterogeneous hydrolysis (Wang et al., 2017; Ding et al., 2021). However, these cases ae infrequent, as only 2% of trajectories are associated with Cluster 2.

During E1, more than 50% of $^{97}HSO_4^-$ fractions are mixed in the rich-K (81%), OC (62%), Ammonium (100%), EC-aged (98%) particles (Fig. S11), lower than in BB (37%) and Dust (4%) particles. Dissimilar with E1, the number fraction of $^{97}HSO_4^-$ increases to 34% during E2, potentially associated with the enhancement by secondary formation. However, the mixing state of $^{195}H(HSO_4)_2^-)$, $^{62}NO_3^-$, NH_4^+ and oxalate fractions are similar between the two episodes events. The fractions of $^{58}C_2H_5NHCH_2^+$ are significantly higher in E2 than E1 for Ammonium (67% versus 31%) and EC-aged particles (48% versus 17%), due to the relatively higher hygroscopic behavior (i.e., RHs) (Sorooshian et al., 2007).

3.3 Formation process of the high number concentration particle episodes

Photochemical oxidation and aqueous-phase reaction are the key formation pathways of secondary species (Link et al., 2017; Xue et al., 2014; Jiang et al., 2019). Generally, the oxidant O_x (O_3 + NO_2) concentration and RH serve as indicators of the degree of photochemical oxidation (Wood et al., 2010) and aqueous-phase reaction (Ervens et al., 2011). In this study, the relative number fractions of $^{43}C_2H_3O^+$, $^{89}HC_2O_4^-$, $^{62}NO_3^-$, $^{97}HSO_4^-$, and

[18]NH_4^+-containing particles to the total detected particles were selected to indicate the secondary formation (Liang et al., 2022). The correlations of the number fraction of each secondary species with the O_x concentrations (O_x) during daytime (from 06:00 to 20:00 LT) and RH during nighttime (from 20:00 to 06:00 next day LT) are used to present the formation pathways during the two episodes (Li et al., 2022).

As illustrated in Fig. 6, for E1, [43]$C_2H_3O^+$, [89]$HC_2O_4^-$, [97]HSO_4^-, [18]NH_4^+ show significant negative linear correlations with O_x ($p < 0.01$), and the correlation strengths range from moderate to strong (r = −0.51 to −0.81). However, the [62]NO_3^- fraction shows an upward trend with an insignificant correlation (r = 0.33, $p > 0.05$) with the increase in O_x concentration. For E2, [43]$C_2H_3O^+$ shows weak correlation with O_x (r = 0.37, $p > 0.05$), but strong correlations with [89]$HC_2O_4^-$, [97]HSO_4^-, and [18]NH_4^+ (r = 0.81~0.92, $p < 0.01$). It should be noted that [62]NO_3^- has a strong negative correlation (r = −0.85, $p < 0.01$) with O_x. In general, the opposite linear relationship between secondary aerosol and O_x during E1 and E2 might be influenced by reasons of i) the relatively low secondary formations because of the small amount of precursors emitting from anthropogenic activities around the sampling site (Li et al., 2016); ii) higher dilution rate of the particles formed in the atmosphere with the rapid rise of PBL height during E1 than E2 (Fig. S12a); iii) the degrees of contributions of regional transport due to the low WS (0.5 ± 0.6 m s^{-1}) during E1 and the high WS (3.1 ± 1.0 m s^{-1}) during E2, respectively (Fig. S8). Therefore, for E1, the increases of NO_3^- fraction could be influenced by the local nitrate formation, while the declines of other secondary components should be ascribed to the reduced contribution of regional transport. For E2, the decreased of [62]NO_3^- fraction could be caused by the relatively higher volatilization loss of nitrate than other components through the regional transport. Previous study proves that the formations of organic nitrate species (such as [27]CHN^+, [30]NO^+, [43]CHO_1N^+, and CHO_xN^+) through the $NO+RO_2$ pathway dominate 80% of the total nitrate production in tropical forested regions during summertime (Alexander et al., 2009). Aruffo et al (2022) also found that low NO_x (i.e., < 6 ppbv), compared to 2.3 ± 0.8 ppbv in this study, could even promote the particle-phase partitioning of the lower volatility of organonitrates. These results suggest that photo-oxidation reactions could promote secondary formation, among which the rate of HSO_4^- formation (slop=0.017) is the highest. Increased with O_x concentration during E2, the

concentration levels of secondary organic species of $C_2H_3O^+$ (18%-28%) imperceptibly rise, while the oxalate fraction significantly increase by 7%-20% as well. The results indicate that the secondary organic species have different formation capacities through atmospheric oxidation.

Considering that the oxalate is abundant mixed in rich-K (14%), BB (15%), EC-aged (5%), and Dust (6%) particles in Cluster 1 (Fig. 5), and the increased contributions of rich-K (39.3%), BB (14.2%) and EC-aged (17.2%) types during E2 (Table 1), the apparent formation of oxalate might be due to the enhancement of regional transport. Particularity, this presents the nearby biomass burning and combustion activities produce more precursor species of oxalate (Sullivan et al., 2007; Kundu et al., 2010; Zhang et al., 2017b).

Fig. 7 illustrates that the number fractions of $^{43}C_2H_3O^+$, $^{89}HC_2O_4^-$, $^{97}HSO_4^-$, and $^{18}NH_4^+$ have moderate to strong positive correlations with RH (r = 0.70~0.81, p < 0.01 or 0.05) in the nighttime during the two episodes, except that $^{43}C_2H_3O^+$ during E2 (p = 0.48) and $^{89}HC_2O_4^-$ during E1 (p = 0.12). Furthermore, $^{62}NO_3^-$ fraction has no obvious changes with insignificant correlation with RH during E1 (p = 0.43) and presents a moderate negative correlation with RH (r = 0.69, p < 0.01) during E2. As shown in Fig. 7e, the highest aqueous formation rate of HSO_4^- is mainly due to the properties of low volatile and high hygroscopic of sulfate (Wang et al., 2016; Zhang et al., 2019c; Sun et al., 2013). Compared with that during E2 (slop=0.014), the decreased formation rate of HSO_4^- during the E1 (slop=0.009) may be because the decreases of aerosol acidity in higher RH > 80% (Huang et al., 2019; Meng et al., 2014; Tian et al., 2021). And the increased contributions of regional transport due to the high WS (2.4 ± 0.8 m s^{-1}) during E2 are comparable to the low WS (0.08 ± 0.08 m s^{-1}) during E1 (Fig. S8). The fair production rate of NH_4^+ during the E1 (slop= 0.005) and E2 (slop=0.006) demonstrate that an aqueous-phase reaction could effectively promote ammonium formation. Meanwhile, a slightly larger slop during E2 could be also affected by the increased contributions of regional transport. Compared with those during E1, the inverse generation rates of two secondary organic species (i.e., $C_2H_3O^+$ and $HC_2O_4^-$) during E2 are possibly caused by the different formation pathways with a variety of RH levels or distinct regional transports. For example, $C_2H_3O^+$ shows a strong correlation with RH (r = 0.70, p < 0.05) during E1 (slop=0.003) but has insignificant correlation during E2. This could be explained

by high RHs that could effectively promote secondary organic formation during E1. In addition, the $HC_2O_4^-$ fraction increases slightly (9.7-13.1%) during E1 is potentially ascribed to more abundant Dust-type particles (20.3%) which compose of high calcium (Ca) (Fig. S13) that favor the formation of metal oxalate complexes (i.e., Ca oxalate). At high RHs (93.4 ± 7.6%), if oxalate ions are dissolved in the aqueous phase with the presence of Ca ions, the Ca oxalate complexes can precipitate because of their low hygroscopic and insoluble natures (Furukawa and Takahashi, 2011). This could offset the oxalate formation in the aqueous-phase reaction. However, significant linear increases (slop=0.003) with RH (r = 0.81, p < 0.01) during E2 demonstrate that the aqueous-phase reaction effectively promotes the oxalate formation (Cheng et al., 2017; Meng et al., 2020). No obvious change and insignificant correlation between $^{62}NO_3^-$ and RH are found during E1, potentially attributed to the decreases of NO_2 concentration (3.7 ± 0.4 ppbv) in the local atmosphere. Meanwhile, high RHs could promote organonitrates formation (Fang et al., 2021; Fry et al., 2014). The linearity between $^{62}NO_3^-$ and RH (r = 0.69, p < 0.01) significantly decreases during E2, mostly due to the losses of the volatile compound through the regional transport (Fig. S14).

4 Conclusions

This study presents the chemical composition, size distribution, mixing state, and secondary formation of individual particles in the southeastern margin of TP, China during the pre-monsoon season using a high-resolution SPAMS. The finding shows that the rich-K (30.9%) and BB types (18.7%) are the two dominant aerosol particles in the remote area; followed by the OC (12.8%), Ammonium (11.9%), EC-aged (10.9%), and Dust (10.7%) types; the NaK-SN, Metal and Others types contributed 0.3–2.8% to the total ambient particles. By interpreting the mass spectra and diurnal trends, the major particle types are mainly from traffic emission, biomass burning, secondary formation, and fly ash, while the dynamics of the PBL height could also affect the contributions of these particles. The observed change in the number fraction of the particle types was mainly influenced by air mass (97.61% of the total trajectories) from northeastern Myanmar, and significantly contributed to rich-K and BB types. The particle types show distinct size distributions. The two most critical particle types of rich-K and BB appear in a unimodal pattern, the fractions of OC and EC-aged gradually decrease with the increase of the particle sizes, but Ammonium and Dust types show the

opposite. Sulfate is the major secondary species and is highly mixed with rich-K, Ammonium, and EC-aged types. Nitrate has a relatively low mixing ratio due to its higher volatility than sulfate during regional transportation, except for BB and OC types. During the entire study campaign, two air episodes with the high number concentration particle occurred but with significant differences in each particle fraction due to the different meteorological conditions (RH, WS, etc.). The indicators of secondary formation, $C_2H_3O^+$, $HC_2O_4^-$, NH_4^+, NO_3^- and HSO_4^- are shown inverse linear correlation with O_x (O_3+NO_2) during episode I and II periods due to the different meteorological conditions; however, they are present a positive linear correlation with relative humidity (RH), except for NO_3^- shown the negative linear correlation with RH due to the low precursors concentration and potential organonitrates formation. These results demonstrated that the capacity of atmospheric aging of photo-oxidation and aqueous reaction have complex influencing factors. The results of this study provide useful information concerning the detailed characteristic of aerosol components, size distribution, and mixing states in the southeast TP, and highlight the importance of the cross-border transport and formation mechanism of aerosols in high-altitude regions.

删除[Li]: The results of the formation mechanism of secondary species demonstrate that the formation capacity of atmospheric oxidation is affected by the PBL height, but the relative humidity (RH) could promote the formation of secondary species, especially $^{97}HSO_4^-$ and $^{18}NH_4^+$.

471 *Data availability*. The data presented in this study are available at the Zenodo data archive

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473

474 *Competing interests*. The authors declare that they have no conflict of interest.

475

476 *Author contributions*. QW and JC designed the campaign. WR conducted field measurements.

477 LL, QW, JT, and YZ made data analysis and interpretation. LL and QW wrote the paper. All

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479

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828 https://doi.org/10.1016/j.scitotenv.2017.04.101, 2017.

829

830 Table 1. The number concentrations, average percentages and characteristic ions of nine types
831 of particles during the entire study period, and the average percentages of the major six
832 particle types during two episodes.

Type	Number count	Fraction in total (%)	Episode 1 (%)	Episode 2 (%)	Tracer ions
rich-K	151040	30.9	29.0	39.3	$^{39}K^+$, $^{26}CN^-$, $^{42}CNO^-$, $^{46}NO_2^-$, $^{62}NO_3^-$, $^{97}HSO_4^-$
BB	91322	18.7	11.5	14.2	$^{39}K^+$, levoglucosan ($^{45}CHO_2^-$, $^{59}C_2H_3O_2^-$, $^{71}C_3H_3O_2^-$, $^{73}C_3HO_3^-$), $^{26}CN^-$, $^{35,37}Cl^-$, $^{42}CNO^-$, $^{46}NO_2^-$, $^{62}NO_3^-$, $^{97}HSO_4^-$
OC	62446	12.8	8.1	10.0	$^{27}C_2H_3^+$, $^{37}C_3H^+$, $^{38}C_3H_2^+$, $^{39}K^+/C_3H_3^+$, $^{43}C_2H_3O^+$, $^{51}C_4H_3^+$, $^{26}CN^-$, $^{42}CNO^-$, $^{46}NO_2^-$, $^{62}NO_3^-$, $^{97}HSO_4^-$
Ammonium	58317	11.9	17.5	13.5	$^{12}C^+$, $^{18}NH_4^+$, $^{39}K^+$, $^{58}C_2H_5NHCH_2^+$, $^{97}HSO_4^-$, $^{195}H(HSO_4)_2^-$
EC-aged	53337	10.9	10.0	17.2	$C_n^{\pm}$ (n =1 ~ 5), $^{39}K^+$, $^{97}HSO_4^-$
Dust	52533	10.7	20.3	1.3	$^{40}Ca^+$, $^{56}CaO^+$, $^{16}O^-$, $^{17}OH^-$, $^{76}SiO_3^-$, $^{79}PO_3^-$
NaK-SN	13726	2.8	na	na	$^{23}Na^+$, $^{39}K^+$, $^{62}NO_3^-$, $^{97}HSO_4^-$
Metal	4672	1.0	na	na	$^{51}V^+$, $^{56}Fe^+$, $^{64,66,68}Zn^+$, $^{206,207,208}Pb^+$
Others	1580	0.3	na	na	No obvious characteristic peaks

833

Figure captions:

Figure 1. Box and whisker diurnal plots of the number concentration of the main particle types (a) rich-Potassium (K), (b) Biomass burning (BB), (c) Organic carbon (OC), (d) Ammonium, (e) Element carbon (EC)-aged, (f) Dust in hourly resolution. The lower, middle, and upper lines of the boxes denote the 25th, 50th, and 75th percentiles. The lower and upper whiskers show the 10th and 90th percentiles, respectively. Average values are shown in white dots.

Figure 2. Maps of the mean HYSPLIT back trajectory clusters (72 h) at the height of 500 m during the whole field observation. Embedded pie chart represents the relative fraction of each particle type in the four clusters.

Figure 3. Size distributions of (a) the total number particle counts, (b) the relative percentages (%) of the total particles for nine groups during the sampling campaign.

Figure 4. Number fractions of secondary markers associated with the six particle types (rich-K, BB, OC, Ammonium, EC-aged, Dust). Secondary species include sulfate ($^{97}HSO_4^-$), sulfuric acid ($^{195}H(HSO_4)_2^-$), nitrate ($^{62}NO_3^-$), ammonium ($^{18}NH_4^+$), amine ($^{58}C_2H_5NHCH_2^+$), and oxalate ($^{89}HC_2O_4^-$) ions.

Figure 5. Number fractions of secondary markers associated with the six particle types (i.e., rich-K, BB, OC, Ammonium, EC-aged, and Dust) in four clusters. Secondary markers include sulfate ($^{97}HSO_4^-$), sulfuric acid ($^{195}H(HSO_4)_2^-$), nitrate ($^{62}NO_3^-$), ammonium ($^{18}NH_4^+$), amine ($^{58}C_2H_5NHCH_2^+$), and oxalate ($^{89}HC_2O_4^-$).

Figure 6. Correlations between the relative number fractions of the secondary species (a) $^{43}C_2H_3O^+$, (b) $^{89}HC_2O_4^-$, (c) $^{18}NH_4^+$, (d) $^{62}NO_3^-$, (e) $^{97}HSO_4^-$ and O_x concentration during E1 (blue square) and E2 (red dot).

Figure 7. Correlations between the relative number fractions of the secondary species (a) $^{43}C_2H_3O^+$, (b) $^{89}HC_2O_4^-$, (c) $^{18}NH_4^+$, (d) $^{62}NO_3^-$, (e) $^{97}HSO_4^-$ and relative humidity (RH) during E1 (cyan dot) and E2 (orange square).

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删除[Online_Group]: ize distributions of the (a, c) number concentrations and (b, d) fractions of the major six particle types (rich-K, BB, OC, Ammonium, EC-aged, Dust) during two episodes of (a,b) E1 and (c, d) E2

设置格式[Li]: 字体颜色: 自动设置

删除[Online_Group]: **Figure 6.** Number fractions of secondary markers associated with the six particle types (rich-K, BB, OC, Ammonium, EC-aged, Dust) in two episodes events of E1 and E2: sulfate ($^{97}HSO_4^-$), sulfuric acid ($^{195}H(HSO_4)_2^-$), nitrate ($^{62}NO_3^-$), ammonium ($^{18}NH_4^+$), amine (DEA (diethylamine, $^{58}C_2H_5NHCH_2^+$), and oxalate ($^{89}HC_2O_4^-$).

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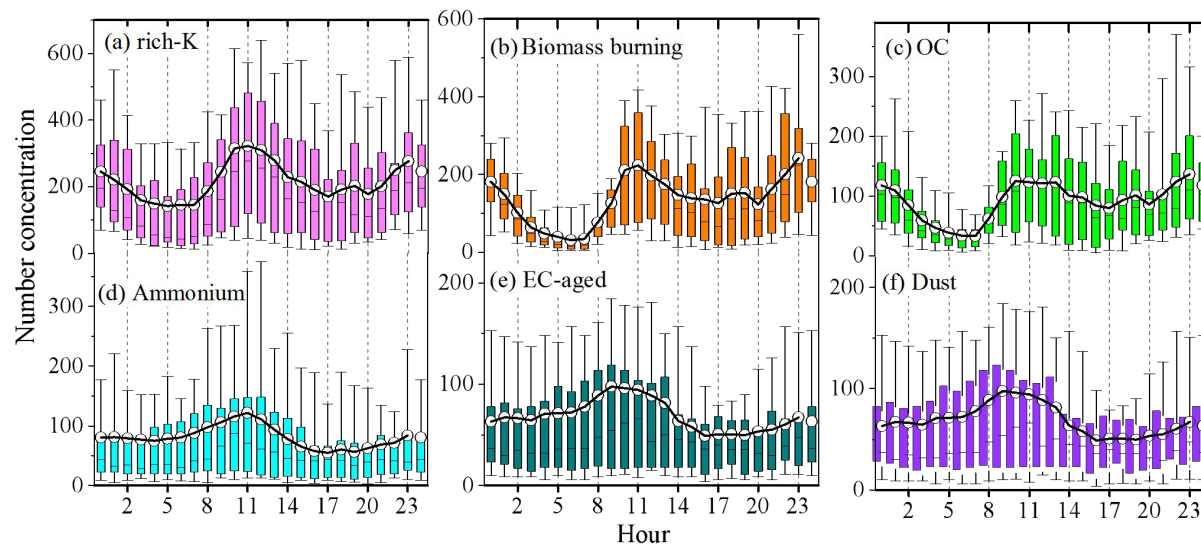
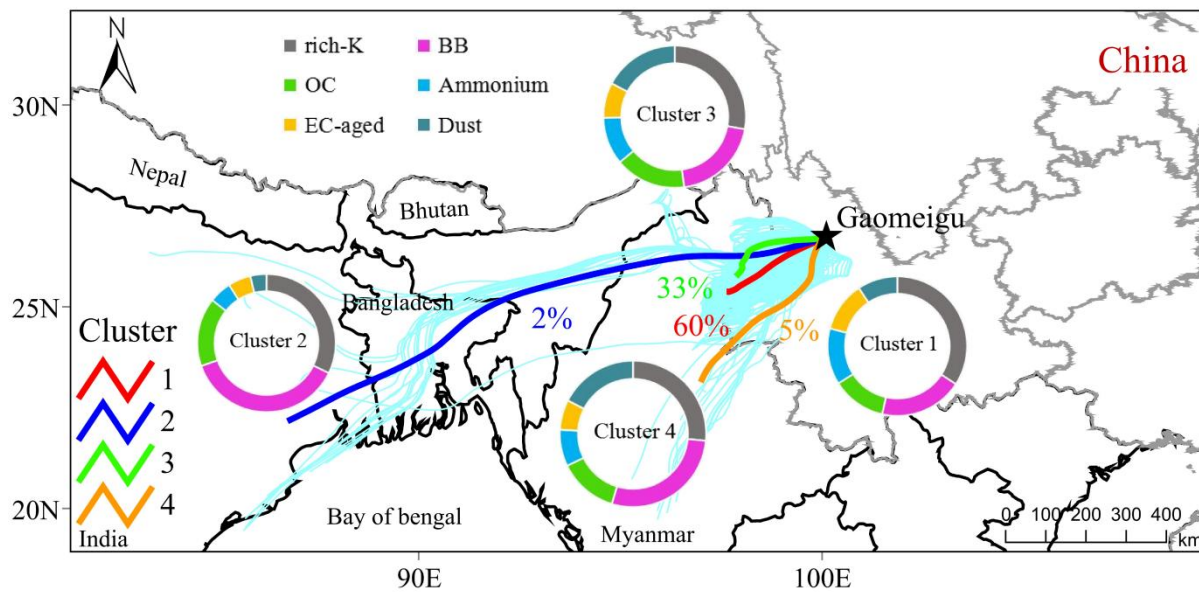

860

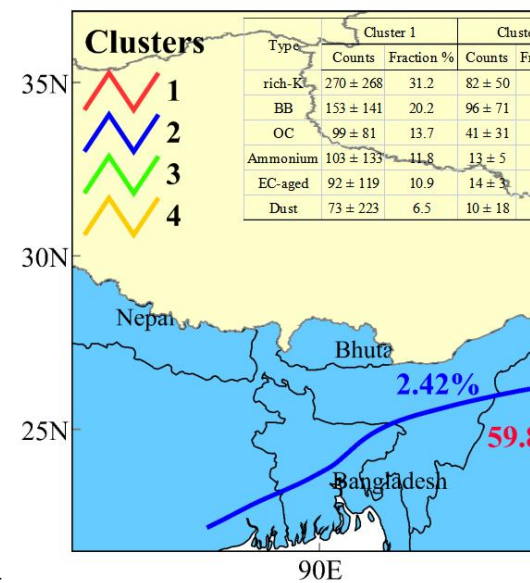
861   Figure 1. Box and whisker diurnal plots of the number concentration of the main particle types (a)

862   rich-Potassium (K), (b) Biomass burning (BB), (c) Organic carbon (OC), (d) Ammonium, (e) Element

863   carbon (EC)-aged, (f) Dust in hourly resolution. The lower, middle, and upper lines of the boxes denote the

864   25th, 50th, and 75th percentiles. The lower and upper whiskers show the 10th and 90th percentiles,

865   respectively. Average values are shown in white dots.

866

867 Figure 2. Maps of the mean HYSPLIT back trajectory clusters (72 h) at the height of 500 m during the
868 whole field observation. Embedded pie chart represents the relative fraction of each particle type in the
869 four clusters.

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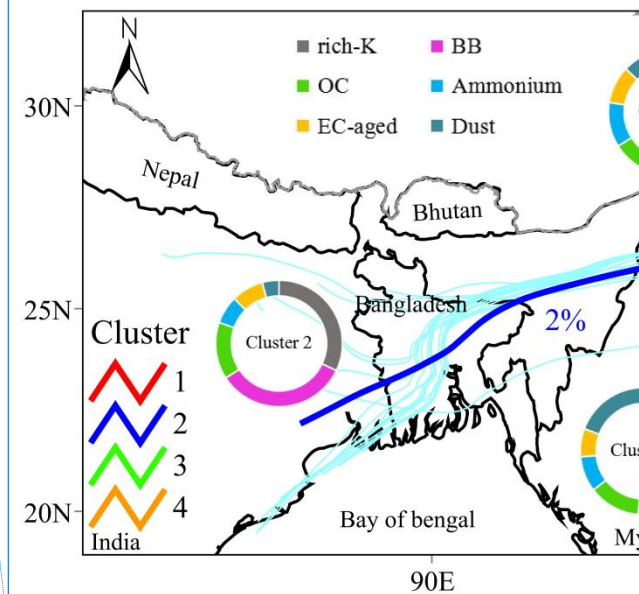
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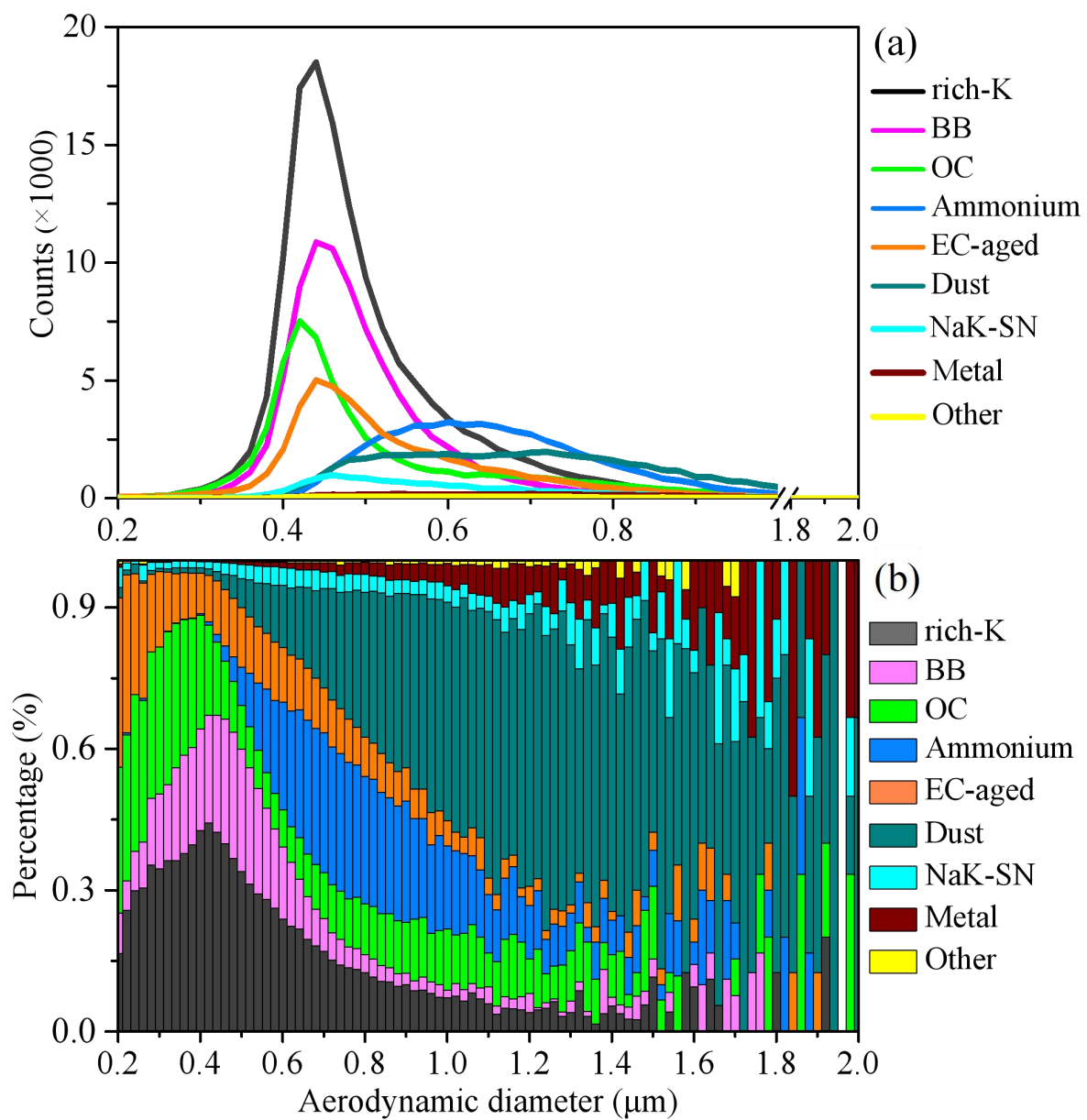
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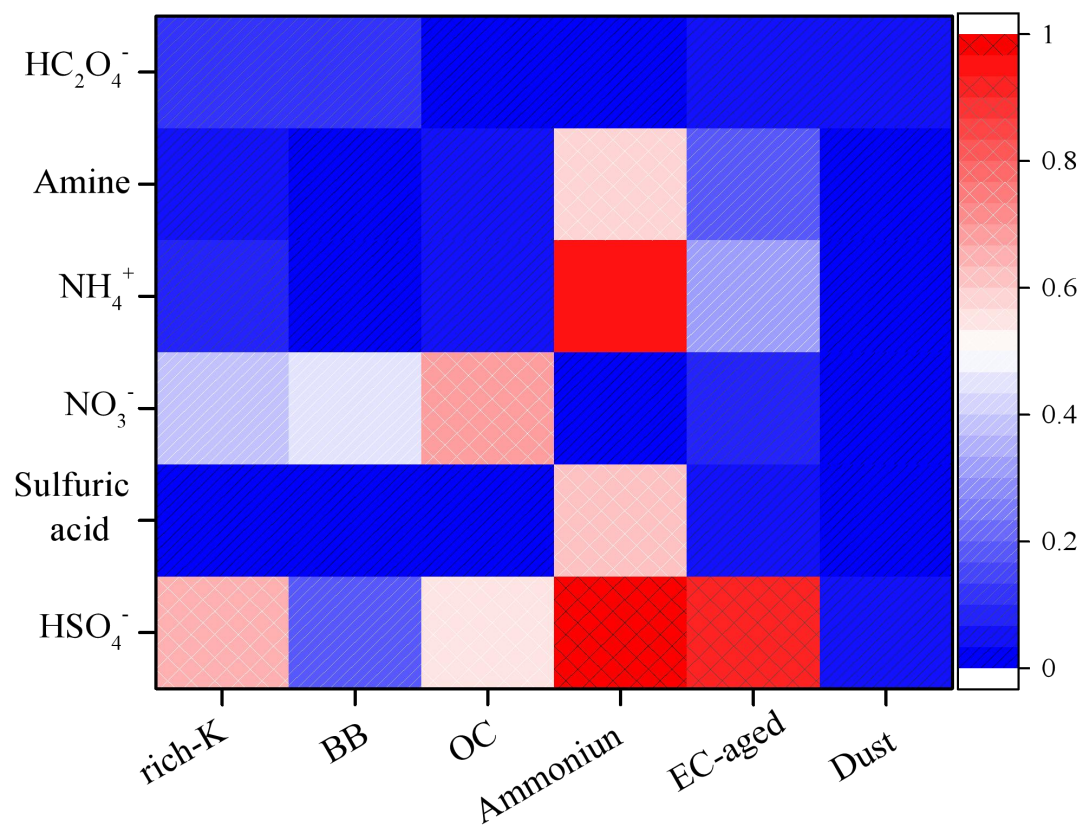
871 Figure 3. Size distributions of (a) the total number particle counts, (b) the relative percentages (%) of the

872 total particles for nine groups during the sampling campaign.

874 Figure 4. Number fractions of secondary markers associated with the six particle types (rich-K, BB, OC,

875 Ammonium, EC-aged, Dust). Secondary species include sulfate ($^{97}HSO_4^-$), sulfuric acid ($^{195}H(HSO_4)_2^-$),

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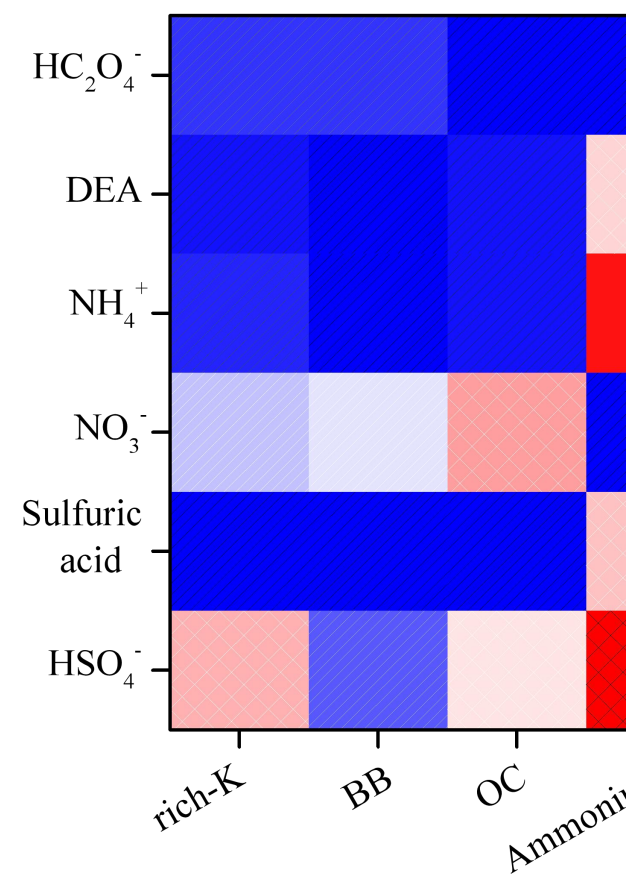
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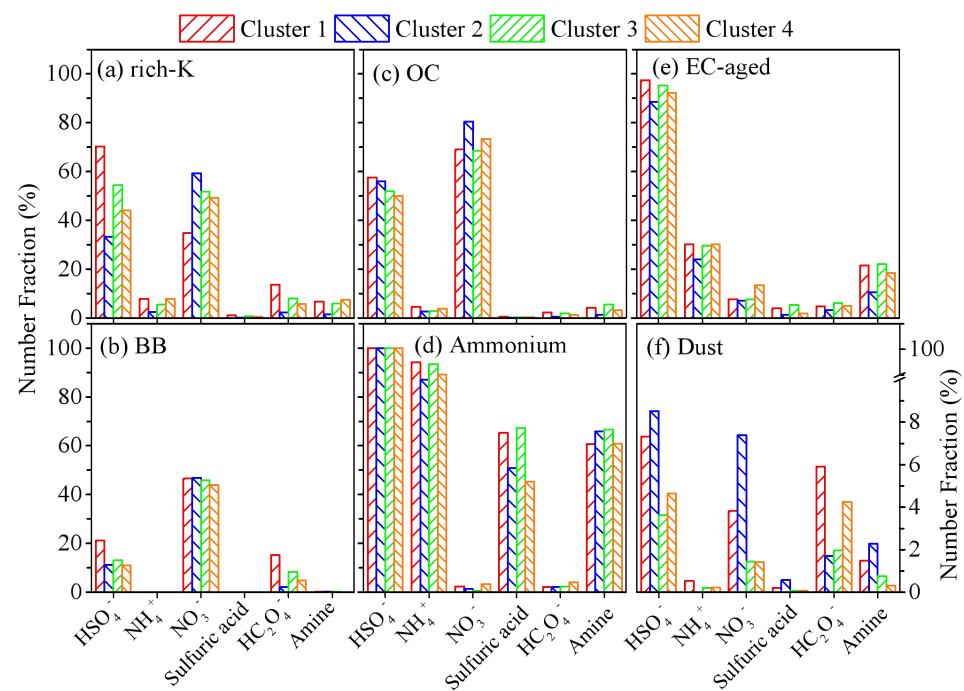
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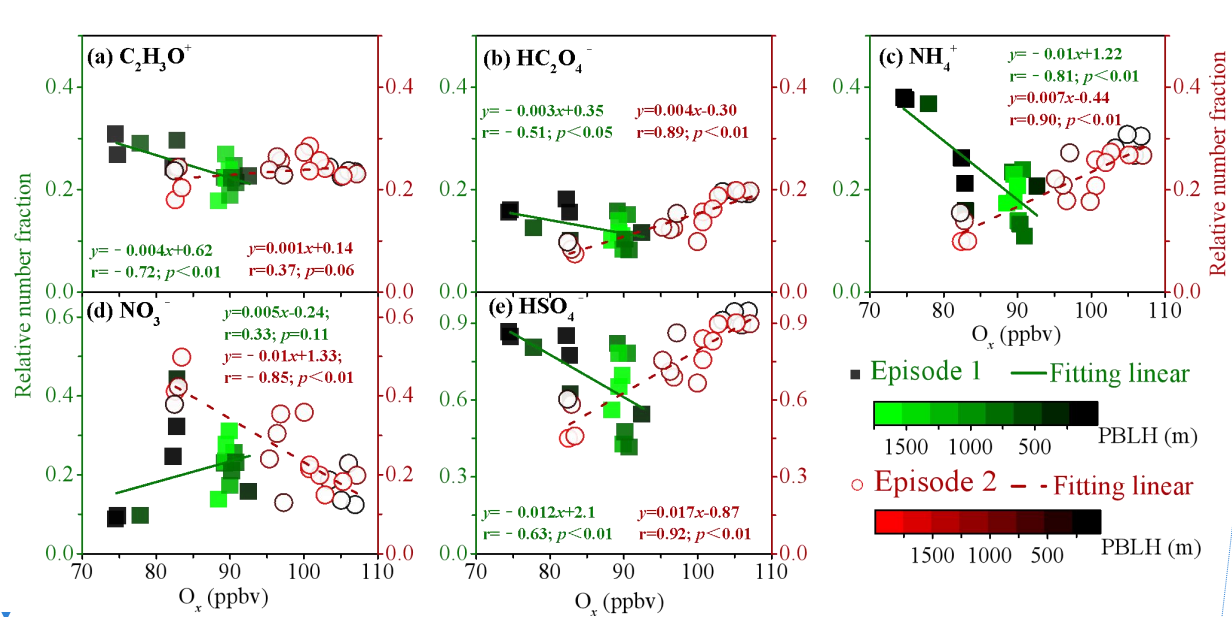

884

885   Figure 6. Correlations between the relative number fractions of the secondary species (a) $^{43}C_2H_3O^+$, (b)

886    $^{89}HC_2O_4^-$, (c) $^{18}NH_4^+$, (d) $^{62}NO_3^-$, (e) $^{97}HSO_4^-$ and O_x concentration during E1 (blue square) and E2 (red

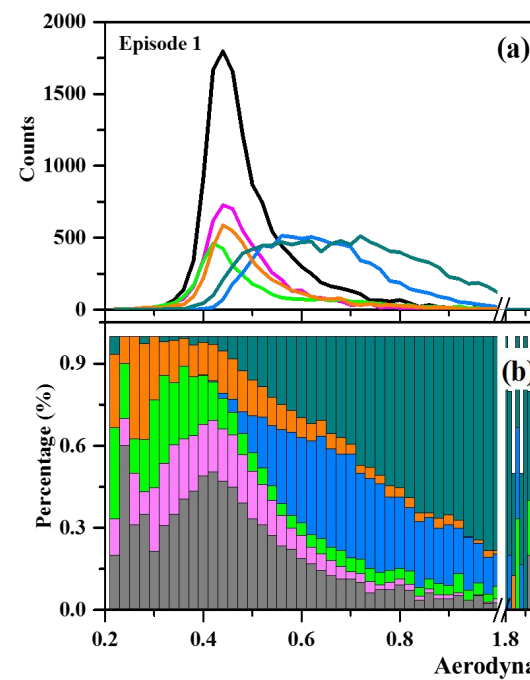
887   dot).

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Figure 5. Size distributions of the (a, c) number concentrations

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E1 and (c, d) E2.

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Figure 7. Correlations between the relative number fractions of the secondary species (a) [43]$C_2H_3O^+$, (b) [89]$HC_2O_4^-$, (c) [18]NH_4^+, (d) [62]NO_3^-, (e) [97]HSO_4^- and relative humidity (RH) during E1 (cyan dot) and E2 (orange square).

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