Mixing state of refractory black carbon in fog and haze at rural sites in winter on the North China Plain

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Abstract. The variability of the mixing state of refractory black carbon aerosol (rBC) and the corresponding complicated light absorption capacity imposes great uncertainty for its climate forcing assessment. In this study, field observations using a single particle soot photometer (SP2) were conducted to investigate the mixing state of rBC under different meteorological conditions at a rural site on the North China Plain. The results showed that the hourly mass concentration of rBC during the observation periods was 2.6 ± 1.5 µg m^{-3} on average with a moderate increase (3.1 ± 0.9) during fog episodes. The mass-equivalent size distribution of rBC exhibited an approximately lognormal distribution with a mass median diameter (MMD) of 213 nm. We found that the count median diameter (CMD) of rBC particles during snowfall episodes was larger than that before-snowfall, and the number of rBC with $Dc <$ 121 nm decreased 28.4% after snow. Indicating that rBC-containing particles with small core sizes (Dc) were much more effectively removed by snow with light snow intensity (0.23 mm/h). Based on the Mie-scattering theory simulation, the relative and absolute coating thicknesses of rBC-containing particles were estimated to be 1.6 and ~52 nm for the rBC core with a mass-equivalent diameter (Dc) of 170~190 nm, respectively, indicating that the most of rBC-containing particles were thinly coated. Furthermore, moderate light absorption enhancement (E_{abs} = 1.3) and relatively low absorption cross-section (MAC = 5.5 m^2/g) at 880 nm were observed at the GC site in winter compared with other typical rural sites.

The relationship between the microphysical properties of rBC and meteorological conditions was also studied. Relative warm and high RH environments (RH> 50%, -4 ℃< T < 4 ℃) were more favorable to rBC ageing than dry and cold environments (RH< 60%, T < -8 ℃). And the increase in ambient RH at the same temperature favors rBC aging. Increasing mass fraction of secondary inorganic aerosols (SIA, especially sulfate and nitrate) and decreasing mass fraction of organic aerosols in the environment support the formation of thick coatings by rBC. The RH dependence of E_{abs} was likely caused by the RCT as supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH in the ambient. The mass fractions of aqueous-phase formation of secondary components had a limited effect on E_{abs} under a high RH environment. The

measured *r*BC concentrations and the mixing state of rBC in different meteorological environments will be useful for evaluating the radiative forcing of *r*BC in regional climate models.

45 **Keywords:** Refractory black carbon; Mixing state; Aging process; Snowfall

1 Introduction

As an absorbing aerosol produced by incomplete combustion, black carbon plays a leading role in the absorption of shortwave solar radiation and changes atmospheric thermodynamics through the heating effect. Black carbon is

50 an important factor leading to highly uncertain climate warming (Bond et al., 2013; Moffet and Prather, 2009). Black carbon aerosols enhance haze pollution by absorbing solar radiation to change the meteorological characteristics of the planetary boundary layer (PBL) (Ding et al., 2016). Freshly discharged black carbon is hydrophobic and becomes hydrophilic when mixed with hydrophilic material during aging, acting as cloud condensation nuclei (Sarangi et al., 2019; Twomey, 1974) and increasing the cloud drop number concentration

55 (Cherian et al., 2017). Wet removal is a key factor in determining the lifetime of black carbon aerosols (BCS) in the atmosphere, which determines their vertical and horizontal distribution, local and regional diffusion, and direct, semidirect, and indirect radiative forcing effects (Liu et al., 2013).

The burning of fossil fuels such as coal and oil and the burning of biomass in agricultural production caused by human activities are the main factors leading to the increasing black carbon emissions. Using a transmission

60 electron microscope fitted with an environmental cell (ETEM), we observed that fresh black carbon presents a loose and porous chain structure, and then in the atmosphere through condensation, coagulation, heterogeneous oxidation condensation, or heterogeneous chemical reaction, *r*BC is coated by soluble material, and the coating gradually becomes thicker with increasing RH (He et al., 2015; Freney et al., 2010). The mixing state determines the morphology and optical properties of black carbon, affects its wet removal efficiency, and thus affects the

65 atmospheric lifetime (Yu et al., 2020). The extent of *r*BC coating has been considered to be an important factor in evaluating aerosol radiative capacity (Oshima et al., 2009). It has been shown that the mixing of BC and other non-BC materials can enhance radiation absorption, which is interpreted as exhibiting a "lensing effect," with the

magnitude strongly dependent on the rBC coating amount (Cappa et al., 2012; Liu et al., 2015; Moffet and Prather, 2009). Generally, rBC-containing particles with thicker coatings have higher hygroscopic properties and can act as

70 CCN under severe pollution conditions (Ding et al., 2019; Schwarz et al., 2008). Under high pollution and RH conditions, the absorption efficiency of black carbon was significantly increased. Reducing the emission of BC and related secondary coating precursors is an effective way to alleviate the direct radiation effect of BC in urban environments. However, the change in the mixing state of black carbon aerosols and their driving factors are not clear under high pollution and RH conditions.

75 The North China Plain is one of the BC hotspots with intensive anthropogenic emissions (Cao et al., 2006; Yang et al., 2016). Recently, the spatiotemporal distribution and optical properties of rBC and its influence on climate and the environment have been reported (Zhao et al., 2020a; Ding et al., 2019; Liu et al., 2020b). However, observations of the mixing state of black carbon under different meteorological conditions are still insufficient. SP2 with high resolution and high precision have significant advantages in estimating the mixing state of black carbon. During

80 recent years, an increasing number of observations have used SP2 to observe the mixing state of black carbon in China (Wang et al., 2014; Gong et al., 2016; Liu et al., 2020b; Pan et al., 2017). Most studies have focused on the mixing state of BC on severe haze days because of the presence of large amounts of particulate matter and gaseous precursors in the atmosphere. Studies have focused on the chemical mechanisms such as condensation, coagulation, and heterogeneous oxidation for coating formation (He et al., 2015; Bond et al., 2013), while the connection

85 between meteorological factors and coatings is relatively rare. Previous studies have shown that wind speed and variation in the planetary boundary layer (PBL) can influence the concentration of BC (Talukdar et al., 2019) and that relative humidity (RH) affects the morphology and hygroscopic growth of rBC-containing particles (Laborde et al., 2013; Freney et al., 2010). Meteorological fields may have an impact on the mixing state of rBC.

In this study, SP2 was deployed at a moderately polluted rural site on the North China Plain in winter to characterize

90 the size distribution and mixing state of rBC. This work is unique due to the largely different meteorological conditions during the campaign. The differences in rBC particle size under different meteorological conditions are illustrated, and the main factors influencing the rBC mixing state are discussed. The results of this paper were

exhibited in the following sequence: (1) the size distribution of rBC in different campaigns; (2) the relative and absolute coating thickness and optical properties of rBC-containing particles and their influencing factors; and (3) the relationship of the aging process of rBC with meteorological conditions such as RH, temperature and pollution conditions.

2 Observations

2.1. Site description and measurements

The observation of refractory BC (rBC) particles was performed from December 7th, 2019 to January 1st, 2020, in an air-conditioned container located in farmland of the Gucheng (GC, latitude:39°7.8′N; longitude:115°48′E) site in Dingxing County, Hebei Province, China, which is part of the China Atmosphere Watch Network (CAWNET) operated by the Chinese Meteorological Administration. As shown in Fig.S1(a), the observation station is located ~120 kilometers southwest of Beijing, adjacent to National Highway 107 (G107) and the Beijing-Hong Kong-Macau Expressway (G4). This result indicates that the sampling site was affected by traffic emissions sources, which is a feature compared to other typical rural sites mainly affected by biomass burning sources at home and abroad. A single particle soot photometer (SP2, Droplet Measurement Technologies Inc. USA) was used to measure rBC. A high–resolution time–of–flight aerosol mass spectrometer (HR-AMS, Aerodyne Research Inc., USA) was used to measure the non-refractory submicron (NR-PM1) aerosol species (organics, sulfate, nitrate, ammonium, and chloride). More details on this instrument can be found in previous studies (Jimenez, 2003; Kumar et al., 2016). Ambient relative humidity varied from 9% to 100% during the observation period, with an average relative humidity of 66%.

2.2 SP2 data analysis

2.2.1 Instrument and calibration

The measurement of the size distribution and mixing state of individual rBC-containing particles in GC was characterized using an SP2. The aerosol sampling inlet was placed on a lift tower 3.5 m above the ground, and ambient air entered the container through a 1/2 in. soft tube. The SP2 was operated at a sampling flow rate of 100 vcmm. There was a continuous intracavity Nd: YAG laser beam (1064 nm, TEM00 mode) in the SP2 measurement chamber. SP2 used particle identification by laser-induced incandescence technology to observe and identify rBC

(Stephens et al., 2003; Schwarz et al., 2006b). All particles scatter light, but only when they contain highly absorbent and refractory components, such as rBC, they absorb the laser radiation and are heated to their boiling point temperature before emitting blackbody radiation (Liu et al., 2010; Moteki and Kondo, 2007). When particles cross the laser, particles' composition and size can be determined by detecting the incandescence signal when particles evaporated and the elastic scattering signal of particles. The ratio of the signals observed at the detectors for the narrowband and broadband is used for estimating the color temperature of thermal radiation from rBC–containing particles and, thereby, constrains particle composition(Schwarz et al., 2006a). If only the scattering signal was detected, it was proven that the particles do not contain rBC and are purely scattering particles (non-rBC). If both the scattering signal and the incandescence signal were recorded, it was an rBC-containing particle. In general, rBC-containing particles need more time to evaporate the nonrefractory matter before reaching their boiling point temperature. Therefore, there was a lag time (Δt) for the peak of the incandescence signal and the scattering signal, which can be used to qualitatively study the mixing state of rBC-containing particles. For SP2, the detectable size range of the scattering signal was 200-400 nm diameter. Assuming a black carbon density of 1.8 g/cm^3 and an ideally spherical structure, the mass equivalent diameter of black carbon detected by the incandescence signal is 70-500 nm.

Prior to the observation field campaign, the scattering and incandescence signals of SP2 were calibrated in the laboratory. Aquadag aerosols (Lot #9627, Acheson Inc., USA) were used to calibrate the incandescence signal. We used an atomizer (model 3072, TSI Inc, USA) and a diffusion dryer to prepare monodispersed Aquadag aerosols, which were selected by mobility diameter using a differential mobility analyzer (DMA; Model 3081, TSI Inc., 150 USA). The calibration coefficient values for broadband high gain (BBHG) and broadband low gain (BBLG) of incandescence signals are shown in Fig. S2. We corrected the incandescence signal for ambient rBC with a scaling factor of 0.75 because the SP2 is more sensitive to Aquadag than diesel emission rBC in the same mass range (Laborde et al., 2012; Baumgardner et al., 2012). The scattering signal was calibrated using polystyrene latex spheres (PSL, Nanosphere Size Standards, Duke Scientific Corp., USA). In addition

to the above instruments, a condensation particle counter (CPC, model 3775, TSI Inc., USA) was adopted to measure the number concentration of particles to compare with SP2.

2.2.2 Shell-core calculation

A single particle rBC mass was obtained by the incandescence signal. We usually refer to the mass equivalent diameter converted by the rBC mass (ρ=1.8 g/cm^3 for ambient BC) as the rBC core diameter (Dc), which is the diameter of a void-free, spherical structure, as expressed in Eq. (1):

$$Dc = \sqrt[3]{\frac{6*M_{rBC}}{\pi*\rho_{rBC}}}, \tag{1}$$

Information about the scattering signal of rBC-containing particles detected by SP2 is distorted because when individual rBC particles passed through the laser beam, they absorb the laser energy and are heated up to ~4000 K, resulting in mass loss of a single rBC particle directly. Thus, the methodology of leading-edge-only (LEO) fitting is adopted to characterize the original scattering properties and coating thickness of rBC-containing particles. The scattering signal of purely scattering particles is Gaussian, as detected by SP2. In this method, the Gaussian fitting of the original scattering signal is reconstructed from the leading edge of the scattering signal (before the volatilization of particles) of rBC-containing particles (Gao et al., 2007). In this study, rBC-containing particles are assumed to be shell-core structures. The optical diameter of rBC-containing particles or coated diameter (Dp) is calculated by Mie table calculations. Using a core refractive index $n_c = 2.26 + 1.26i$ and a coating refractive index $n_s = 1.48 + 0i$, we input the LEO fitted scattering signal and rBC core size into Mie calculations (Taylor et al., 2015). Further, the relative coating thickness (RCT), namely, the shell/core ratio and absolute coating thickness of an rBC-containing particle are calculated as D_p/D_c and $(D_p - D_c)/2$, respectively.

2.2.3 Calculation of rBC optical properties

The ensemble effect of light absorption by rBC-containing particles is calculated at 880 nm because the optical properties of rBC-containing particles at this wavelength are hardly affected by brown carbon (BrC). Based on the size information of rBC-containing particles (i.e., Dc and Dp) obtained by SP2 observation, optical parameters such as the mass absorption cross-section (MAC), absorption enhancement (E_{abs}), and absorption coefficient (σ_{abs}) of rBC-containing particles can be obtained based on Mie scattering theory. The refractive index of the rBC core was

2.26 + 1.26i, and the refractive index of 1.48 + 0i, was used for coatings in the Mie calculation assuming rBC-containing particles are a core-shell model. In addition to the absorption of light by the rBC core, coating materials on the surface of rBC can attract more light to the rBC core, which is called the lensing effect, enhancing the light absorption of rBC. The E_{abs} can be used to quantify the theoretical light absorption capability of BC-containing particles. E_{abs} is determined by the ratio of the absorption cross-section of the whole rBC-containing particle ($C_{abs,s}$) to the absorption cross-section of the bare rBC core ($C_{abs,c}$), as shown in Eq. (2):

$$E_{abs} = \frac{C_{abs,s}}{C_{abs,c}} \tag{2}$$

MAC reflects the optical absorption characteristics of rBC-containing particles in unit mass concentration. The larger the value is, the stronger the radiation absorption capacity of rBC-containing particles is. The mass absorption cross-section (MAC$_i$) of a single rBC-containing particle is defined as the absorption cross-section of the whole rBC-containing particle ($C_{abs,s}$) per unit rBC mass ($m_{rBC,i}$), as expressed in Eq.:

$$\text{MAC}_i = \frac{C_{abs,s}}{m_{rBC,i}} \tag{3}$$

The MAC in bulk for a given period is calculated as:

$$\text{MAC} = \frac{\sum_i \text{MAC}_i \times m_{rBC,i}}{\sum_i m_{rBC,i}}. \tag{4}$$

The σ_{abs} of rBC-containing particles is calculated based on the MAC and the mass concentration (C_{rBC}) measured by SP2, as shown in Eq. (4):

$$\sigma_{abs,calculated} = MAC \times C_{rBC} \tag{5}$$

Previous studies have shown that the light absorption of rBC–containing particles is related to their morphology (Liu et al., 2017; Liu et al., 2020b). Simple assumptions of core-shell structure and ideal spheric shape of rBC particles, used in Mie theory to calculate optical properties, cause uncertainty due to the lack of observation of optical size information and morphology.

2.3 Dispersion and trajectory analysis

The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model developed by NCEP (National Center for Environmental Prediction) and NCAR (National Center for Atmospheric Research) was widely used to simulate the air footprint area for dispersion and trajectory analysis, which was based on the Lagrangian transport model (https://ready.arl.noaa.gov/HYSPLIT_traj.php, last access: February 28th, 2020) (Cohen et al., 2015; Fan et al., 2021b; Fan et al., 2021a). The potential uncertainty of the HYSPLIT model is related to the meteorological assimilation data sets that drive the model simulation (Su et al., 2015). In this study, the HYSPLIT model was used to investigate the source of air mass on the north China plain. The dataset provided for HYSPLIT is the global reanalysis data in GDAS format (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1, last access: February 26th, 2020). It produces meteorological data four times a day, namely, at 00:00, 06:00, 12:00, and 18:00 UTC, and the horizontal resolution is 2.5°×2.5°. The vertical direction has 17 levels, ranging from the ground surface to 10 hPa. Meteorological parameters including wind, temperature, humidity, potential height, and ground precipitation are also provided. During the simulation, the trajectory ensemble option starts multiple trajectories from the first selected starting location. Each member of the trajectory ensemble is calculated by offsetting the meteorological data by a fixed grid factor (one meteorological grid point in the horizontal direction and 0.01σ units in the vertical direction). Air samples were released at 150 m above ground level from the GC site, and the simulation time of the backward trajectory was 5 days.

3 Results and Discussion
3.1 Overview of the field observation
3.1.1 Meteorology

The field campaign was performed from December 8th to 31st 2019. During the observation period, the site was affected by the winter monsoon with a prevailing northwesterly wind (Fig. S1(b)). The meteorological conditions and the concentration of aerosol particles during the sampling period are shown in Fig. 1. The average ($\pm 1\sigma$) wind speed was 1.2 (± 1) m s^{-1}, with the highest value up to 5.9 m s^{-1}. According to the wind rose diagram, the northwest wind and the southwest wind prevailed during the observation, and the wind speed reached a maximum in the

southwest direction during the whole sampling period according to the wind rose diagram (Fig.S1(c)). The relative humidity (RH) during the entire period was relatively high, with an average ($\pm 1\sigma$) of 66 ($\pm$ 25) %. There were fog events and snowfall during the observation period. The period when the RH reached 100% between December 7th and 11th was defined as the fog event, and the visibility was almost 0 km during this period. Snowfall occurred at 20:00 on December 15th and lasted until 14:00 on December 16th, with snow intensity of 1.37 mm/6h. It was of great significance to study the mixing state and removal of rBC-containing particles during fog and snow events to improve the model's simulation.

3.1.2 Mass concentration of rBC

As shown in Fig. 1, the number concentrations of rBC and purely scattering particles showed similar variation trends. The average ($\pm 1\sigma$) number concentration of rBC and purely scattering particles were 650 ($\pm$368) and 1184 ($\pm$560) # cm^{-3}, respectively. The mass concentration of rBC in the ambient environment was 2.6 ($\pm$1.5) µg m^{-3}, which was much lower than the observations in the winter of 2016 and 2018 at the urban site in Beijing. Liu et al. (2019) reported that the mass concentration of rBC in winter 2016 in Beijing was 6.4 µg m^{-3}. Due to the strict air pollution control on coal combustion and biomass burning (Ji et al., 2017), Xie et al. (2020) found a decrease in mass concentration (3.2 µg m^{-3}) in winter 2018. A similar urban-high and rural-low pattern was also found in Shenzhen, and the average mass loading of rBC in rural areas (2.6 µg m^{-3}) in autumn was much lower than that in urban areas (6 µg m^{-3}) (Huang et al., 2012). One possible explanation was that rural sites have lower traffic emissions than urban sites, open biomass burning has been strictly controlled in recent years, and the project of changing fuel from coal to natural gas has been well implemented at least in NCP (Huang et al., 2006; Ji et al., 2017).

Based on the meteorological conditions and concentrations of the total particles, six observation periods were chosen, as denoted in Fig. 1a: EP1 (December 7th 17:40 - December 11th 09:00), EP2 (December 12nd 19:00 - December 14th 00:00), EP3 (December 15th 00:00 - December 17th 00:00), EP4 (December 21st 00:00 - December

26th 00:00), EP5 (December 27th 00:00 - December 29th 20:00) and EP6 (December 30th 00:00 - December 31st 00:00). The periods with RH = 100% during EP1 were defined as the fog period. The average RH was 96% and the average rBC mass concentration was 3.6 ($\pm$1.4) µg m^{-3} during EP1. During EP3, there was snow from December 15th 20:00 to December 16th 14:00. Therefore, during EP3, the discussion is divided into before-snow, snow, and after-snow. In the other four periods, EP6 was the clean period with an average rBC mass concentration of 0.5 ($\pm$0.3) µg m^{-3}, while EP2, EP4, and EP5 represented polluted periods with average rBC mass concentration of 3.4 ($\pm$1.4), 1.2 ($\pm$1.2) and 3.4 ($\pm$1.3) µg m^{-3}, respectively. The variables related to rBC and the meteorological conditions in the selected different periods are shown in Table S2. The Hysplit model was used to analyze the origin of the air masses during the different periods. As shown in Fig. 2a-f, the air masses over GC mainly came from the northwest and northeast directions during the clean period, and the wind speed was relatively high. During the pollution period, the air masses mainly came from the southwest and south directions and local areas. During the period of EP1 and EP3 with high RH, part of the air mass originated from the eastern Bohai Sea direction.

3.2 Size distribution of rBC

The time series of the mass size distribution and number size distribution of rBC during the whole experimental period was stable with a modal size peaking at approximately 200nm and 120 nm, respectively (Fig. 3). The rBC measured by SP2 at the GC site mainly were distributed in the range of 70-500 nm and the rBC mass and number were concentrated at 100 – 400 nm and < 300nm, respectively. dM / dlogDc and dN / dlogDc showed an apparent diurnal trend. The peak period of diurnal variation was between 20:00 and 09:00 LST, while low diurnal variation occurred in the afternoon. This pattern was determined by the dual effects of diurnal variation in the boundary layer and local rBC emissions. To better study dM / dlogDc and dN / dlogDc, the mass median diameter (MMD) and count median diameter (CMD) were used to represent the characteristics of dM / dlogDc and dN / dlogDc for detailed discussion.

260 Field measurements of the size of rBC around the world are summarized in Table 1. The size distribution as a function of rBC core mass equivalent diameter (Dc) measured by SP2 is shown in Fig. 4. During the whole experiment, the mass size distribution followed the lognormal distribution and peaked at 213 nm (MMD) (Fig. 4a). As shown in Table 1, the MMD of rBC at the GC site in winter was slightly smaller than the value (222 nm) observed in rural areas in Shenzhen in November (Huang et al., 2012) and consistent with the biomass burning

265 emission observed in Texas (MMD = 210 nm) (Schwarz et al., 2008). However, the rBC emitted by biomass burning in the laboratory was smaller (189 nm) (Pan et al., 2017). In addition, this mode was identical to the rBC mode observed in the aged continental air mass during a Paris study (Laborde et al., 2013). In comparison with most studies in urban areas, we found that the MMD of rBC in urban areas was smaller than that in our study (Huang et al., 2012; Gong et al., 2016; Liu et al., 2019a; Lan et al., 2013; Liu et al., 2020b; Schwarz et al., 2008). Laborde et

270 al. (2013) found that MMD was significantly related to the source of rBC-containing particle emissions. Pan et al. (2017) showed that the MMD of rBC was also determined by combustion conditions and burning materials in a laboratory study. Many studies have indicated that the MMD of rBC from traffic emissions was smaller than that from biomass burning, coal combustion and aged air masses (Laborde et al., 2013; Schwarz et al., 2008). According to previous studies, MMD presented a trend of small size in summer large size in winter, small size in urban areas,

275 and large size in rural areas (Liu et al., 2019a; Liu et al., 2020b; Huang et al., 2012; Kondo et al., 2016; Wu et al., 2021). The dense traffic arteries in urban areas and a large amount of traffic emissions may be the reason that MMD in urban areas is smaller than that in rural areas (Huang et al., 2006; Laborde et al., 2013; Schwarz et al., 2008). The different trends of MMD in seasons and spaces may be because rBC-containing particles came from different emission sources. In addition, the MMD was higher in pollution events (i.e., EP1, EP2, EP3, and EP5). This was

280 similar to the results of previous studies in which under the condition of a higher rBC mass concentration, the rBC core size usually increased (Liu et al., 2019a). It could be that coagulation of rBC particles or regional transport causes changes in rBC-containing particle sources at higher concentrations.

The number size distribution followed the lognormal distribution and peaked at 122 nm (CMD) (Fig. 4a). Because most rBC particles (by number) emitted from urban areas were below the SP2 detection limit of 70 nm, the comparison of the number size distribution between urban and rural areas was meaningless (Schwarz et al., 2008). There was snowfall with the intensity of 0.23 mm/h during the observation period, and the mass and number size distributions of the rBC core mass equivalent diameter (Dc) during the snow event are illustrated in Fig. 4b-c. During before-snow, snow, and after-snow periods, the MMD was 213 nm, 210 nm, and 206 nm, decreasing by 3.3%, and the CMD was 120 nm, 126 nm, and 129 nm, an increase of 7.5%, respectively. We found that the number size distribution during the pre-snow and after-snow periods was consistent with a log-normal distribution. CMD of fresh BC usually ranges from 50 to 80 nm (Bond et al., 2013), so we extrapolated the fitting curves to 10 nm (Fig. 4c). The difference in areas between the two log-normal fit curves at Dc less than 121nm was 0.23, that is, the amount of rBC with Dc less than 121 nm decreased by 28.4% after snow. According to Fig. 3, the number concentration of rBC with Dc below 100 nm decreased significantly while the number concentration of rBC with Dc about 200 nm decreased slightly during the snow case. This result is similar to the observation in Beijing that the snow with a precipitation intensity of 0.37 mm reduced the volume concentration of the small particle size (< 100 nm) fraction in the accumulation mode (Su et al., 2016). Previous studies found that wet deposition is the main mechanism for removing rBC-containing particles (Taylor et al., 2014). Wet removal plays an important role in determining the size distribution of rBC during vertical transport in the free troposphere (Moteki et al., 2012), and rainfalls, as a means of wet removal, preferentially remove large rBC-containing particles, resulting in a decrease in MMD after rain events (Liu et al., 2020b; Wang et al., 2018). However, the snow with relatively weak intensity may preferentially remove the small core size of rBC. Although the CMD increased moderately, the effect of removing rBC particles with a small core size (Dc) by snow was insignificant on the MMD (Liu et al., 2020a). The mechanism of wet scavenging and its effect of the distribution of aerosol particle size is complex (Zhao et al., 2020c; Sun et al., 2019) and we will investigate in more depth how wet scavenging affects rBC particle size in a future study.

3.3 Mixing state of *r*BC-containing particles under different conditions
3.3.1 Coating thickness of *r*BC-containing particles

Figure 5 provides the temporal variation of ACT, RCT, and CMD of *r*BC-containing particles during the entire period. The relative coating thickness (RCT) and absolute coating thickness of *r*BC-containing particles were calculated by the LEO fitting method, see Sect. 2.2.2 for details. Herein, rBC cores with Dc = 180±10 nm and Dc = 190±10 nm were selected because the low scattering signal of small *r*BC is easily influenced by signal noise(Schwarz et al., 2008; Zhao et al., 2020b; Liu et al., 2020b). The average RCT (±1σ) (Dc@170-190nm) was 1.6 (±0.2), RCT (±1σ) (Dc@190-210nm) was 1.5 (±0.2), ACT (±1σ) (Dc@170-190nm) was 52.4 (±21.8) nm, and ACT (±1σ) (Dc@190-210nm) was 54.2 (±20.4) nm. As shown in Table 1, the RCT (Dc@190-210nm) of the GC site was consistent with the RCT (1.2-1.4) with Dc = 200 nm of *r*BC-containing particles emitted from biomass burning through laboratory research (Pan et al., 2017), but was larger than the value (RCT (@Dc= 180 nm) = 1.2) during the summer in Beijing, which was mainly influenced by traffic emissions (Liu et al., 2020b). The ACT at the GC site (Dc@190-210nm) was smaller than the ACT (@Dc= 200 nm) affected by biomass burning (65nm) in Texas and larger than the ACT (@Dc= 200 nm) affected by urban emissions in Texas (Schwarz et al., 2008). The different sources and aging processes of *r*BC-containing particles affect the coatings. Laborde et al. (2013) reported that *r*BC-containing particles emitted by traffic were essentially uncoated with a median coating thickness of 2±10 nm at a core size of 180-220 nm, the median ACT of *r*BC-containing particles emitted by biomass burning was approximately 20-40 nm, and the median ACT of *r*BC-containing particles in continental aged air masses reached ~80 nm.

In addition, we compared the average coating thickness of *r*BC-containing particles during different periods. As shown in Table S2, the average RCT and ACT (±1σ) (Dc@170-190nm) were 2.0 (±0.3) and 92.7 (±23.8) nm, respectively, during the fog event, which was the highest values during the whole observation period. During the before-snow, snow, and after-snow periods, the average RCTs (ACTs) (±1σ) (Dc@170-190nm) were

1.4 (±0.1) (39.9 (±43.6) nm), 1.5 (±0.1) (46.9 (±4.5) nm) and 1.7 (±0.1) (61.3 (±4.7) nm), respectively. During the polluted periods (EP2, EP4, and EP5), the average RCTs (ACTs) (±1σ) (Dc @170-190nm) were 1.5 (±0.1) (42.3(±9.4) nm), 1.7 (±0.1) (58.6(±12.7) nm) and 1.6 (±0.2) (37.0 (±9.5) nm), respectively. The average RCT and ACT (±1σ) (Dc @170-190nm) were 1.4 (±0.1) and 37.0 (±9.5) nm during the clean period (EP6).

335 We used the ambient mass concentrations of NR-PM1 and meteorological conditions to discuss the differences in coating thickness during different periods. According to Fig. 6, the ambient mass concentrations of total NR-PM1 were almost the same during the fog, after-snow, and EP4 periods. However, the coating thickness was different. The RH during these 3 periods was different, where the RH during the fog event was highest (RH=100%), the RH during EP4 was lowest (RH~81%), and the RH during the after-snow period was about 96%. In addition, the concentrations and mass fractions of ambient sulfate and

340 organics during fog events were the highest and lowest, respectively, in the 3 cases. This indicates that a high RH significantly affected the formation of coatings under the same concentration of pollutants in the atmosphere. Moreover, it was also likely that the high RH provides favorable conditions for the sulfate formed by the aqueous phase reaction. This also means that the high fraction of sulfate and low fraction of organics in the environment provide favorable conditions for the formation of coatings. This is similar to the conclusion obtained from previous studies, the increase of growth factor of BC (gf$_{BC}$) was

345 positively associated with the elevation of secondary inorganic material, while was suppressed by the increase of organic coating content (Liu et al., 2013). The changes of the coatings and variations of the NR-PM1 fractions in the environment were also consistent with this pattern. The secondary transformation of inorganic aerosols was intensified due to the increasing ambient RH during the snowfall. The increase in the fraction of secondary inorganic aerosols in the atmosphere and the decrease in the fraction of organic materials and chlorides to a certain extent favor the formation of rBC coatings. Similarly,

350 the coating thickness of rBC–containing particles showed a positive variation with RH and the fractions of secondary inorganic aerosols during the selected pollution periods (EP2, EP4, and EP5). Despite the higher concentration of rBC during EP2, the lower RH and the fraction of secondary inorganic aerosols during this period result in thinner coatings of rBC–containing particles. Indicating that pollution processes based on primary emissions were not conducive to the formation of rBC coatings. During dry clean days, both primary emissions and secondary transformation were weak and the coating thickness of rBC–

355 containing particles was relatively thinner compared with periods of pollution days under high RH conditions. This part of rBC–containing particles were mainly aged rBC that has not been scavenged.

3.3.2 Absorption enhancement of rBC-containing particles

The E_{abs}, $\sigma_{abs,}$ and MAC of rBC-containing particles during the whole campaign are shown in Fig.5. The average E_{abs} and MAC at 880 nm were 1.3 and 5.5 m^2/g, respectively, as calculated by Mie theory. Comparisons of E_{abs} in

360 various field studies are shown in Table 2. The E_{abs} at the GC site we simulated were consistent with the value measured by Sun et al. with TD-PAX (Droplet Measurement Technologies) at 870 nm (1.32 ± 0.15) (Sun et al., 2021). The E_{abs} calculated by Mie theory is only related to the core size of rBC, the refractive index of rBC-containing particles, and the coating thickness. It has been suggested out that the correlation between σ_{abs} calculated by Mie theory and σ_{abs} measured by AE33 is as high as 0.98, indicating that the optical properties of rBC-containing particles calculated by Mie theory can represent

365 the actual rBC-containing particles in a specific area (Zhang et al., 2018a). Therefore, we compared the differences in E_{abs} between the GC site and other regions. A moderate E_{abs} (1.21 ± 0.09) at 870 nm was found in California during winter in 2013 which was close to the present study (Zhang et al., 2016). The E_{abs} were slightly larger than the value simulated using core-shell in Beijing during summer in 2008 at 550 nm (1.15) (Liu et al., 2020b). Slightly high E_{abs} values were observed in Xian (1.8 at 870 nm) (Wang et al., 2014) and Paris (1.53 ± 0.93 at 880 nm) (Zhang et al., 2018b) which

370 represent the more polluted urban areas. Previous studies have found that the E_{abs} are generally higher in rural areas than in urban areas, such as Shouxian (2.3 ± 0.9 at 532 nm) (Xu et al., 2018) and Yuncheng (2.25 ± 0.55 at 678 nm) (Cui et al., 2016). In addition, we studied the absorption enhancement of rBC-containing particles during 6 different periods, and specific information is detailed in Table. S1. From the before-snow to after-snow period, the coating thickness and E_{abs} of rBC-containing particles increased gradually. During the clean period, rBC with a larger core size was mostly

375 emitted from primary sources, and the E_{abs} close to 1. In the fog event, the coating thickness was the highest among the 6 periods, while the E_{abs} was lower. This may be because the core size (Dc) of rBC during fog events was larger, rBC with a large core size (Dc) had a lower MAC, and the addition of coatings to rBC with a large Dc did not significantly affect the final total MAC compared to rBC with a small Dc (Liu et al., 2019a). Compared with other rural sites, the E_{abs} and MAC of rBC-

containing particles at the GC site were low. Sun et al. (2021) found that the absorption due to the lensing effect at the GC site correlated best with chloride and less well with SIA and SOA. Chloride is mainly from coal combustion and biomass burning emissions, which means that the E_{abs} at the GC site of rBC-containing particles were dominantly affected by primary species and that the coatings formed by secondary reaction products had little effect on E_{abs}. This result was consistent with the findings observed at Fresno with similar emission sources as the GC site, where the average mixing-induced absorption enhancement of BC due to the coatings can be very small (Cappa et al., 2019).

3.4 Diurnal variation of the mixing state of rBC

Figure 7 exhibits the diurnal trend of the mixing state of rBC-containing particles. The temporal variation in rBC exhibited a clear "U" shaped pattern while the diurnal variation in wind speed was the opposite (Fig. S3). The mean rBC mass loading increased during the evening, with a peak (3.5 µg m^{-3}) at 20:00 LST and a minimum (1.4 µg m^{-3}) at 16:00 LST at the GC site, which was later than that in Beijing (Liu et al., 2020). From the diurnal variation of wind speed (Fig. S3), it can be seen that the wind speed started to increase at approximately 9:00 LST and reached a maximum (2.2 m s^{-1}) at 13:00 LST. These two-time points corresponded to the start of rBC decline and the lowest value of rBC respectively. This diurnal variation in rBC and wind speed can be explained by the diurnal variation in the boundary layer (Simpson and McGee, 2012; Talukdar et al., 2019). In the early morning, calm inversion conditions predominate in the boundary layer. The temperature rose, the wind speed increased to an average of over 2 m s^{-1} at midday, and the rBC declined rapidly with decreased atmospheric stability and increased turbulence. After sunset, the temperature dropped sharply, due to inversion and increased emissions from heating activities, and the surface pollution of rBC was enhanced. The diurnal cycle of σ_{abs} was similar to that of rBC mass concentration, which may be because σ_{abs} of rBC depend on its concentration (Zhang et al., 2018b). MAC and Dc didn't show significant diurnal variations. The coating thickness of freshly emitted rBC-containing particles was thin but thicker through a complex aging process, indicating that the RCT was controlled by the competing effects of the aging process and emissions. The temporal variation in RCT did not present a clear inverted "U" shaped pattern like Beijing during summer (Liu et al., 2020b). Three peaks of the mean RCT appeared at 0:00, 8:00, and

18:00, indicating that rBC emissions increased after 0:00, 8:00, and 18:00 at the GC site and fresh rBC decreased the coating thickness. In addition, there was a significant increase in RCT from the late afternoon. The light absorption enhancement of rBC also did not present a strong diurnal variation trend like Beijing during June (Xie et al., 2019). This trend was consistent with the diurnal variation trend of MAC but different from that of RCT. As shown in Fig. S4, a good correlation was found between MAC (880 nm) and E_{abs} with an R^2 of 0.75, and a weak anticorrelation was found between RCT and E_{abs} with an R^2 of 0.14. This is consistent with the results discussed in Sect 3.3.2, where the E_{abs} at the GC site were more influenced by the MAC of the rBC than the coating thickness. The diurnal variation in the mass fraction of aerosol species is shown in Fig. 7d. The daily cycle of mass fractions in chloride, organics, and ammonium began to decrease in the late afternoon, while nitrate increased. The two higher diurnal peak values of the mass fraction of sulfate occurred at 09:00 and 16:00 LST, and the daily variation peak of the mass fraction of nitrate occurred at 16:00 LST. This may explain the increase in RCT in the late afternoon, and an increase in the mass fraction of sulfate and nitrate in the environment may favor an increase in the coating thickness of rBC-containing particles. However, the diurnal cycle of light absorption enhancement of rBC presented limited dependence on the aerosol species in the environment.

3.5 The dependence of the aging degree of rBC-containing particles on meteorological and pollution conditions

The factors that affect the aging degree of rBC-containing particles include photochemical reactions, chemical coatings, and the meteorology of the environment (Laborde et al., 2013; Saathoff et al., 2003; Schnaiter, 2005). Here, we select rBC-containing particles with a Dc size of 170-190 nm. Fig. 8 shows RCT as a function of temperature and RH. Temperature and RH ranged from -14 to 8 °C and from 9 to 100%, respectively. In general, the changes of RH were mainly controlled by diurnal variations in temperature (Fig. S3). The higher the RH, the greater the RCT at the same temperature. At temperatures lower than -8 °C, the RH was lower than 60%, the RCT was smaller than 1.5. However, at the intermediate temperature regime (-7 – 4 °C), the RH required when the RCT exceeds 1.6 decreases with the increase of temperature. Larger RCT (RCT > 2) occurred in the

temperature range of - 4 – 4 ℃ when the RH was higher than 50%. Indicating that lower RH (~60%) required for rBC to form thick coatings under the subcooled condition (0 ℃< T < 8 ℃), while higher RH (~80%) required for rBC to form thick coatings under the supercooled condition (-14 ℃< T < 0 ℃).

430 In order to exclude the influence of special cases on the general results, data from fog days and snow periods were excluded to further investigate the aging of rBC. Based on the normalized frequency of data points under different RH and temperature ranges (Fig. 9d), five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< T < 0 ℃), and subcooled condition (0 ℃< T < 8 ℃)) to investigate the dependence of the mixing state of rBC on temperature and RH. The mean RCT,

435 E_{abs}, ACT, and aerosol species as a function of RH for 2 different temperatures regimes were shown in Fig. 9 and Fig. 10. The average RCT increased from ~1.4 to 1.6 and from ~1.4 to 1.7 and the average ACT increased from ~37 nm to 51 nm and from ~39 to 65 nm at the GC site as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The coating thickness of rBC particles was thicker under subcooled conditions than under supercooled conditions. This indicates that a suitable high temperature is conducive to the aging of rBC

440 particles. This variation was also supported by the gradual increases in mass concentration and mass fractions of secondary components as a function of RH in the environment. Under subcooled conditions, the coating thickness of rBC started to increase significantly at RH >40%, whereas this phenomenon did not occur until RH >60% under supercooled conditions. This show that the RH required to form a thicker coating of rBC particles was lower under subcooled conditions. The high mass fraction of secondary components formed by liquid phase and heterogeneous

445 reactions in the environment under high RH and subcooled conditions were favorable to form coatings of rBC particles (Collier et al., 2018; Wu et al., 2016).

The RH dependence of average E_{abs} was shown in Fig. 9b. The RH dependence of E_{abs} was likely caused by the RCT as supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH in the ambient (Fig. 10a-b). The average E_{abs} increased from ~ 1.2 to 1.3 and from 1.3 to 1.4 as RH increased from 0 to 100%

450 under the supercooled condition and subcooled condition, respectively. The E_{abs} under subcooled conditions was greater than that under supercooled conditions. The average E_{abs} remained almost constant after RH exceeded 60% under subcooled

conditions. We suspected that the contribution of the aqueous-phase formation of secondary components to E_{abs} might be limited under high RH (Sun et al., 2021).

4 Conclusions

The size distribution, coating thickness, and optical properties of individual rBC-containing particles were measured by a single-particle soot photometer (SP2) during the period December 7th to December 31st, 2019 to characterize the mixing state of rBC-containing particles in rural areas on the North China Plain. There were fog, snow, and heavy pollution events during the observation period, so we analyzed the mixing state and optical properties of rBC-containing particles in different episodes and discussed the factors affecting the aging degree of rBC. The mean mass concentration of rBC at the GC site was 2.6 ($\pm$1.5) µg m^{-3}, which was much lower than the observations in the winter of 2016 and 2018 at the urban site in Beijing. The MMD and CMD of rBC at the GC site were 213 nm and 122nm, respectively. The MMD was smaller than that in other rural areas. In addition to the impact of emission sources, meteorological conditions can significantly affect the size distribution of rBC. The CMD of rBC increased after snow, suggesting that snow events with the light intensity (0.23 mm/h) preferred to remove rBC-containing particles with smaller core sizes. The average RCT (ACT) was 1.6 (52 nm) when the Dc was in the range of 170 nm~190 nm, indicating a moderate coating thickness of rBC during the winter. The coating thickness of rBC-containing particles in the fog event was thicker than that in other periods due to the high RH and high concentrations of secondary aerosols. When the concentrations of NR-PM1 in the environment were the same in the different cases, the higher the RH, the thicker the coating thickness. Higher mass fractions of secondary inorganic aerosols (SIA) and lower mass fractions of organics in the environment may favor the formation of coatings of rBC. The ensemble-average mixing-induced E_{abs} (1.3) and MAC (5.5 m^2/g) at the GC site were smaller than those at other rural areas. The RCT and E_{abs} did not show clear diurnal cycles with little variations in the range of ~1.9-2, and 1.3-1.4, respectively. A good correlation was found between MAC (880 nm) and E_{abs} with an R^2 of 0.75 and a weak anticorrelation was found between RCT and E_{abs} with an R^2 of 0.14 under diurnal cycle. This may be because the optical properties of rBC at the GC site were mainly influenced by primary emissions (such as coal

combustion, biomass burning, and traffic emission), and the absorption due to the lensing effect contributes less to it.

Our results showed that the mixing state of rBC-containing particles depends on the synergistic effects of ambient RH, temperature, and aerosol species. Higher RH made it easier for rBC to form thicker coatings when the ambient temperature was the same. At temperatures lower than -8 ℃, the RH was lower than 60%, the RCT was smaller than 1.5. Larger RCT (RCT > 2) occurred in the temperature range of - 4 – 4 ℃ when the RH was higher than 50%. The RH required to form the same coating thickness of rBC was higher under supercooled conditions than that under subcooled conditions. The RH dependence of E_{abs} was likely caused by the RCT as supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH in the ambient. The mass fractions of aqueous-phase formation of secondary components had a limited effect on E_{abs} under a high RH environment. The increasing mass fraction of secondary components in the environment favored the aging of rBC under high RH, subcooled conditions. Our results highlight that the aging degree of rBC in different meteorological environments needs to be considered in regional climate models to better evaluate the radiative forcing of rBC.

Data availability. To request the data given in this study, please contact Xiaole Pan at the Institute of Atmospheric Physics, Chinese Academy of Sciences, via email (panxiaole@mail.iap.ac.cn).

Author contributions. YZ, SL, HL, and XP designed the research. YZ, SL, HL, XP, WX, YT, WY, XL, QL, JL, CC, YS, PF, JX, JC, and ZW performed the experiments. YZ, SL, HL, and XP performed the data analysis. YZ, HL, XP, and SL wrote the paper.

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

Financial support. This research has been supported by the National Natural Science Foundation of China (grant nos. 4187072067).

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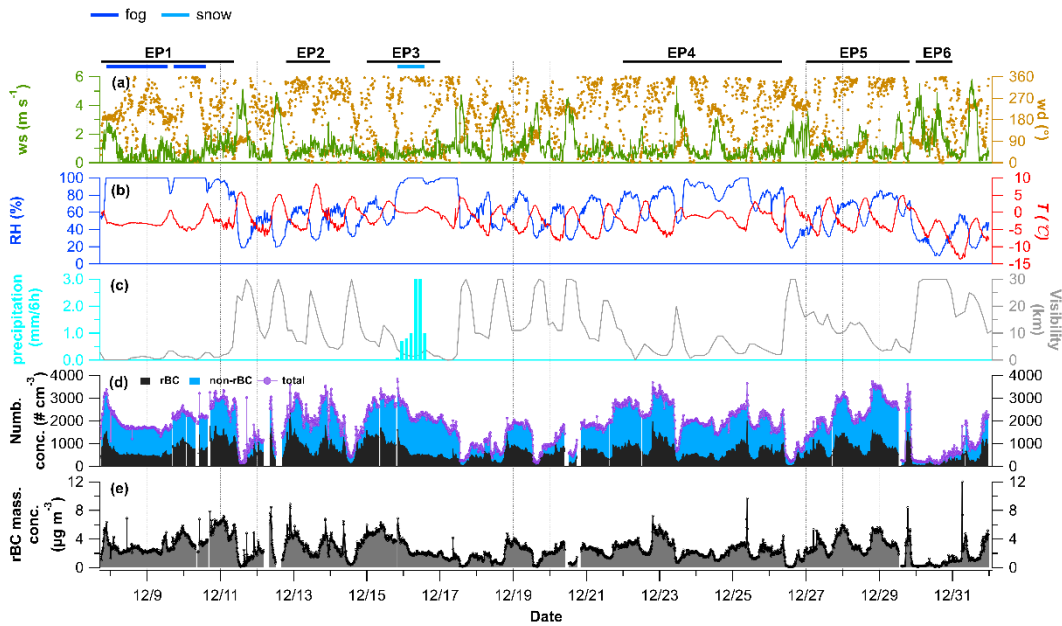
Figure 1: Time series of (a) wind speed (WS) and wind direction (WD), (b) relative humidity (RH) and ambient temperature (T), (c) precipitation and visibility, (d) number concentration of *r*BC-containing particles, purely scattering particles and total particles measured by SP2 in GC and (e) *r*BC mass loading.
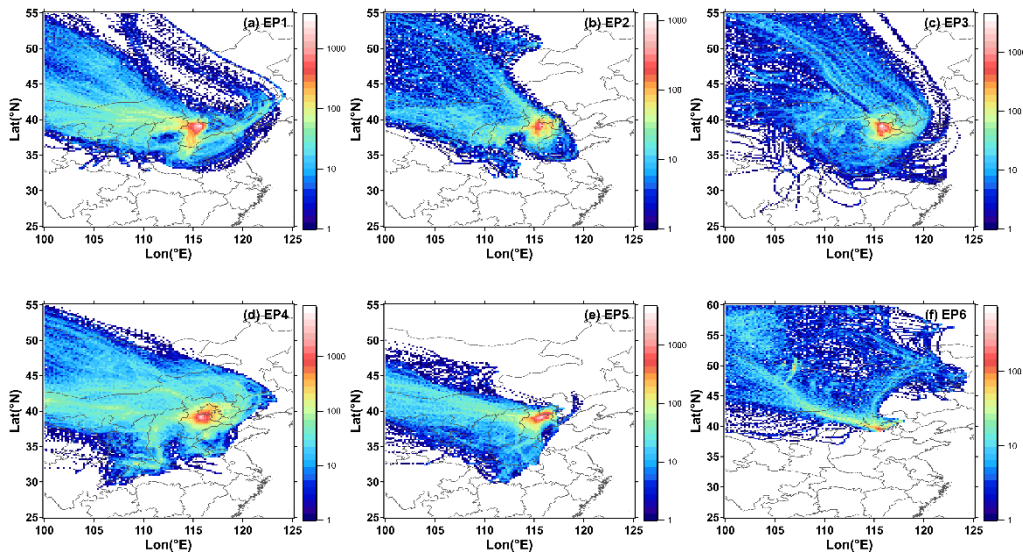
Figure 2: The backward trajectories from which the air mass over GC originated in varying periods.
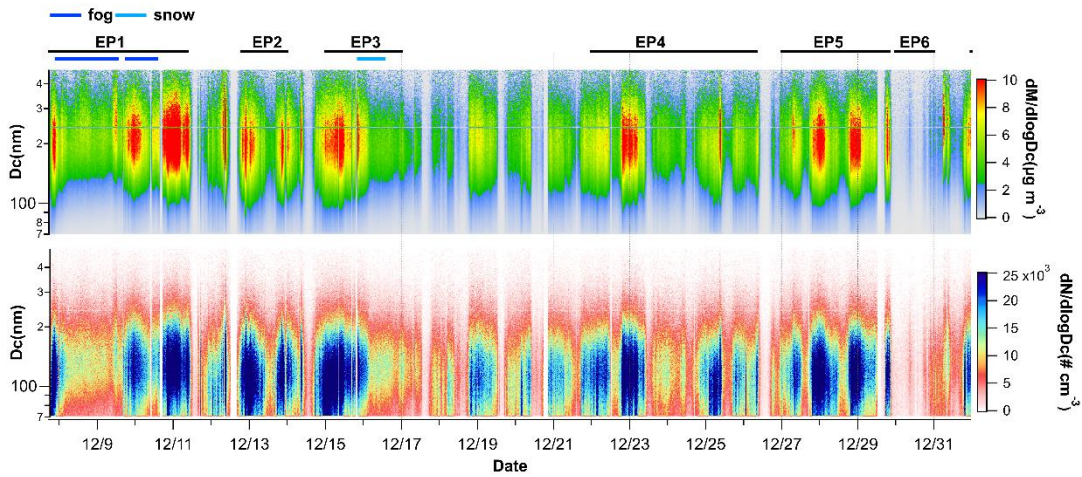
Figure 3: Time series of the mass size distribution and number size distribution of rBC, as measured by the SP2.
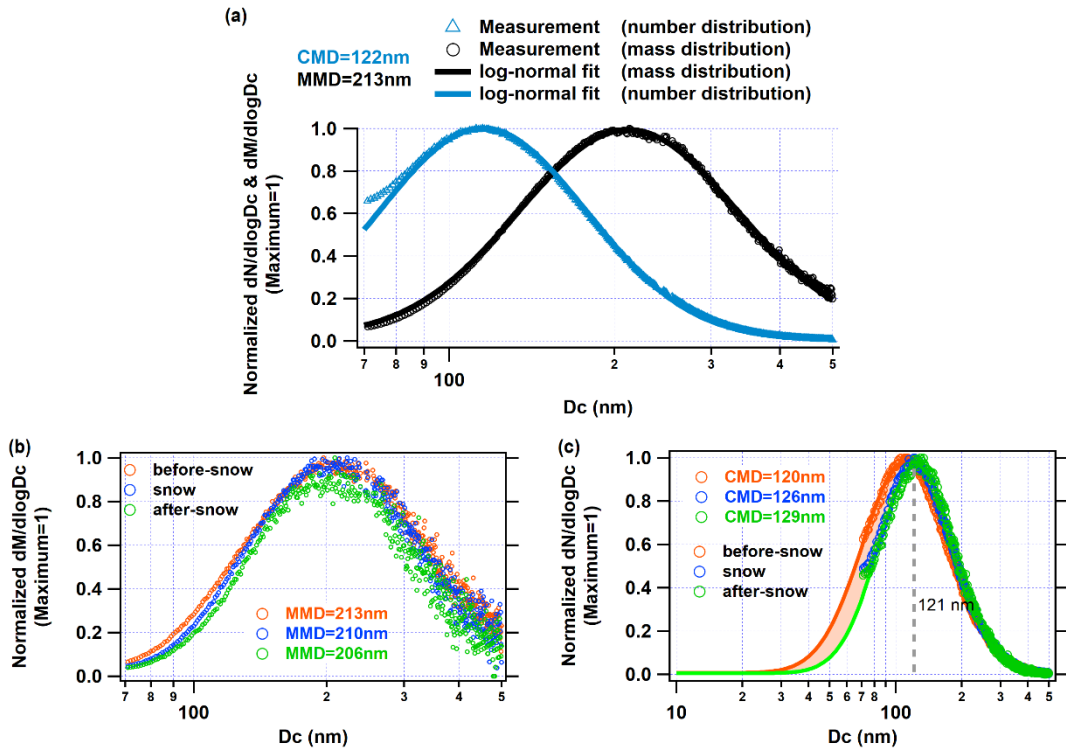
Figure 4: (a) Number and mass size distribution (dN / dlogDc and dM / dlogDc) of rBC core during the whole campaign. (b) Mass size distribution (dM / dlogDc) during snowfall and (c) number size distribution (dN/ dlogDc) during snowfall. Note that the orange and green lines in (c) are the log-normal fit of number size distribution during the before-snow and after-snow periods, respectively. And the orange shaded areas denote the difference in area between the two log-normal fit curves at Dc less than 121nm.
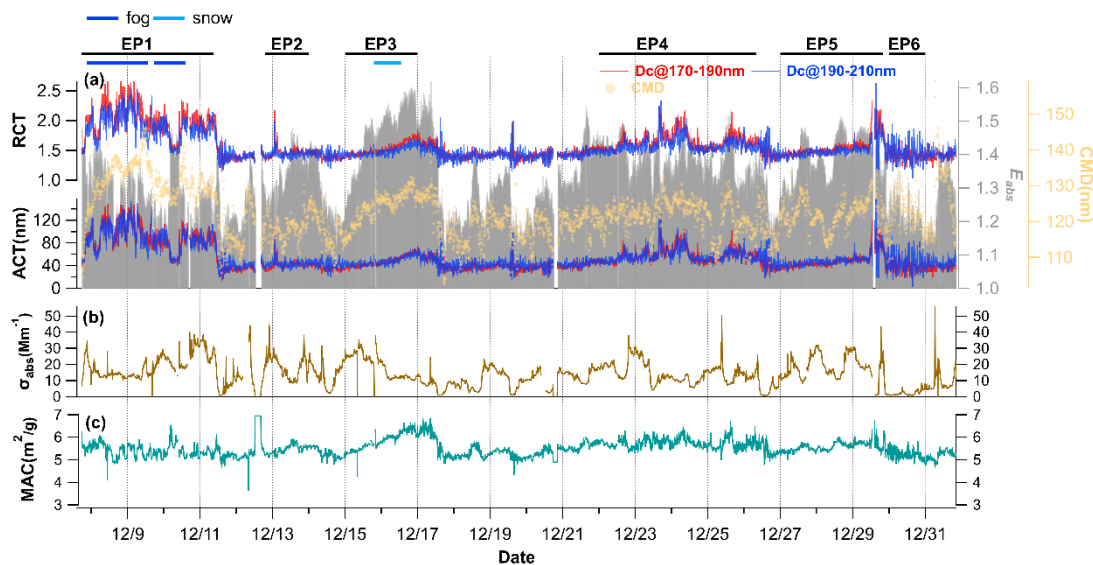
Figure 5: (a) Time series of the relative and absolute coating thicknesses of rBC-containing particles within the Dc range of 170-190 nm and 190-210 nm. The gray bars denote the absorption enhancement (Eabs) of rBC. The light yellow circles denote the size of the rBC core. Time series of the (b) absorption coefficient (σabs) and (c) mass absorption cross-section (MAC).

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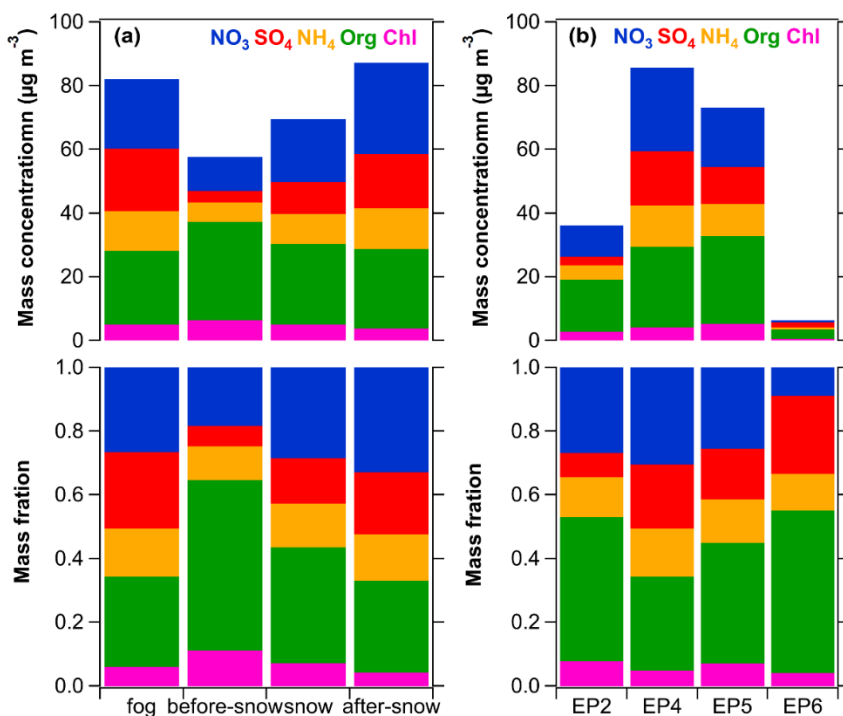
Figure 6: Changes in mass concentration and mass fractions of aerosol species during different periods.

Table 1: Brief summary of some of the experiments on the size and mixing state of rBC-containing particles.

rBC source	Location	Time	CMD (nm)	MMD (nm)	RCT	ACT (nm)	Reference
Urban emission (UE)	Beijing, China	Jun (Summer)		171	1.2(@Dc=180 nm)		Liu et al. (2020b)
	Shenzhen, China	Aug-Sep (Fall)		180*			Lan et al. (2013)
	Shenzhen, China	Oct-Nov (Fall)	110	210*			Huang et al. (2012)
	Shanghai, China	Dec (Winter)		200	2-8	50-300	Gong et al. (2016)
	Texas, US	Sep (Fall)		170		20(@Dc=200 nm)	Schwarz et al. (2008)
	Beijing, China	Nov-Dec (Winter)		195-211	2.5		Liu et al. (2019)
		May-June (Summer)		185	1.4		
	Beijing, China	winter		209			Wu et al. (2021)
		summer		167			
Rural emission (RE)	Shenzhen, China	Nov (Fall)	120	222			Huang et al. (2012)
	This study	Dec (Winter)	122	213	1.6 (@Dc=180 nm)	52.4 (@Dc=180 nm)	
Biomass burning (BB)	Laboratory			189	1.2-1.4(@Dc=200 nm)		Pan et al. (2017)
	Texas, US	Sep (Fall)	140	210*		65(@Dc=200 nm)	Schwarz et al. (2008)
Remote ocean (RO)	Finnish Arctic	Dec-Jan (winter)		194	2.0(@Dc=150~200 nm)		Raatikainen et al. (2015)
Background Continent (BC)	Paris, FR	Jan (Winter)		200		76(@Dc=200 nm)	Labrode et al. (2013)
	Texas, US	Sep (Fall)				48(@Dc=200nm)	Schwarz et al. (2008)

*The density of rBC is 2 g cm^{-3}

Table 2: Comparisons of Eabs in various field studies.

Method	Location	Time	E_{abs} (λ nm)	Reference
TD+PAX+HR-AMS+AE33	Gucheng, China (rural)	Dec 2019-Jan 2020	1.32 ± 0.15 (870)	(Sun et al., 2021)
TD+PAX+SP2+AMS	California, USA (urban)	Jan-Feb 2013	1.21 ± 0.09 (870)	(Zhang et al., 2016)
SP2+CPMA+DMA	Beijing, China (Urban)	May-June 2018	1.15 (550)	(Liu et al., 2020)
SP2+PAX+ACSM	Xi'an, China (urban)	Dec 2012-Jan 2013	1.8 (870)	(Wang et al., 2014)
AE33+ACSM+OC/EC	Paris, France (suburban)	Mar 2014-Mar 2017	1.53 ± 0.93 (880)	(Zhang et al., 2018a)
TD+BBCES+IS	Shouxian, China (rural)	Jun-Jul 2016	2.3 ± 0.9 (532)	(Xu et al., 2018)
AFD	Yuncheng, China (rural)	Jun-Jul 2014	2.25 ± 0.55 (678)	(Cui et al., 2016)
SP2	Gucheng, China (rural)	Dec 2019	1.33 ± 0.57 (880)	This work
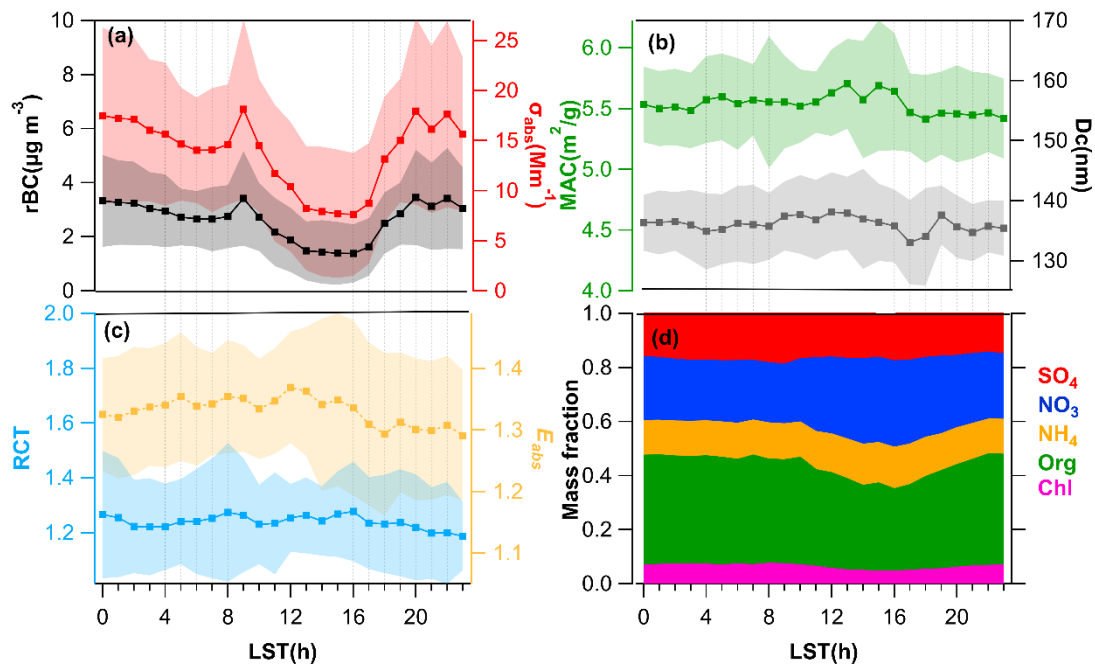
Figure 7: Diurnal variation in (a) *r*BC mass loading and absorption coefficients (σ_{abs}) of *r*BC-containing particles, (b) the mass absorption cross-section (MAC) of *r*BC-containing particles and the MMD, (c) relative coating thickness (RCT@*Dc*=180 nm), and absorption enhancement (E_{abs}) of *r*BC. The shaded areas denote the standard deviation. (d) Mass fraction of aerosol species.
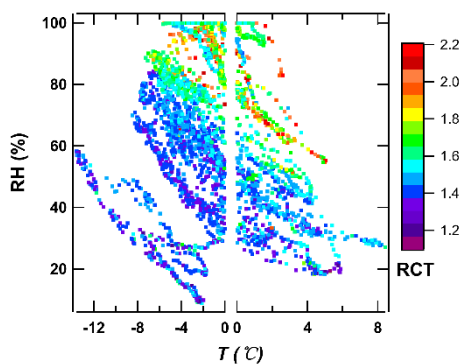
Figure 8: RCT as a function of temperature and RH.
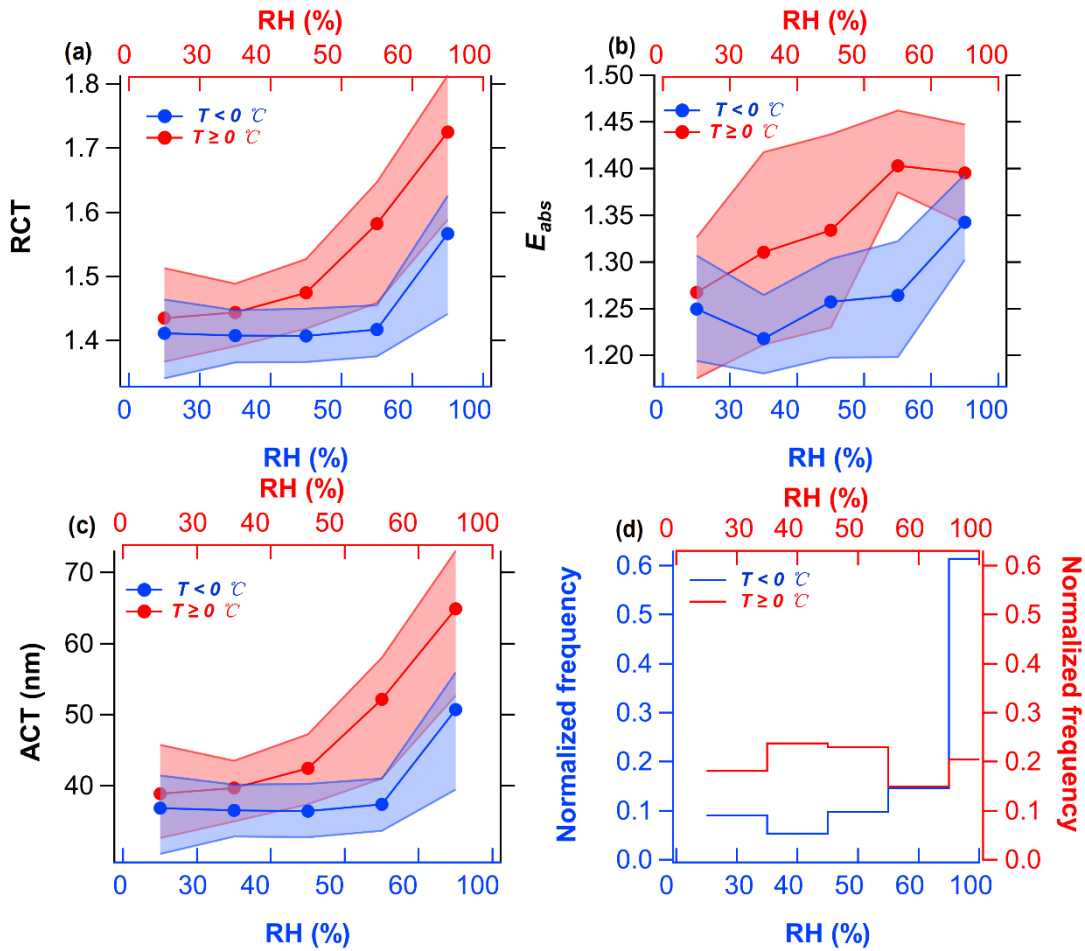
Figure 9: (a-c) RCT (Dc@180 nm), Eabs, and ACT (Dc@180 nm) dependence on RH under the temperature regimes of -14 ℃< T < 0 ℃ and 0 ℃< T < 8 ℃. Circles represent the average values. The upper and lower boundaries of the shaded areas represent the 75th and 25th percentiles of data points in each bin. (d) The normalized frequency of data points in each bin under different temperature ranges.

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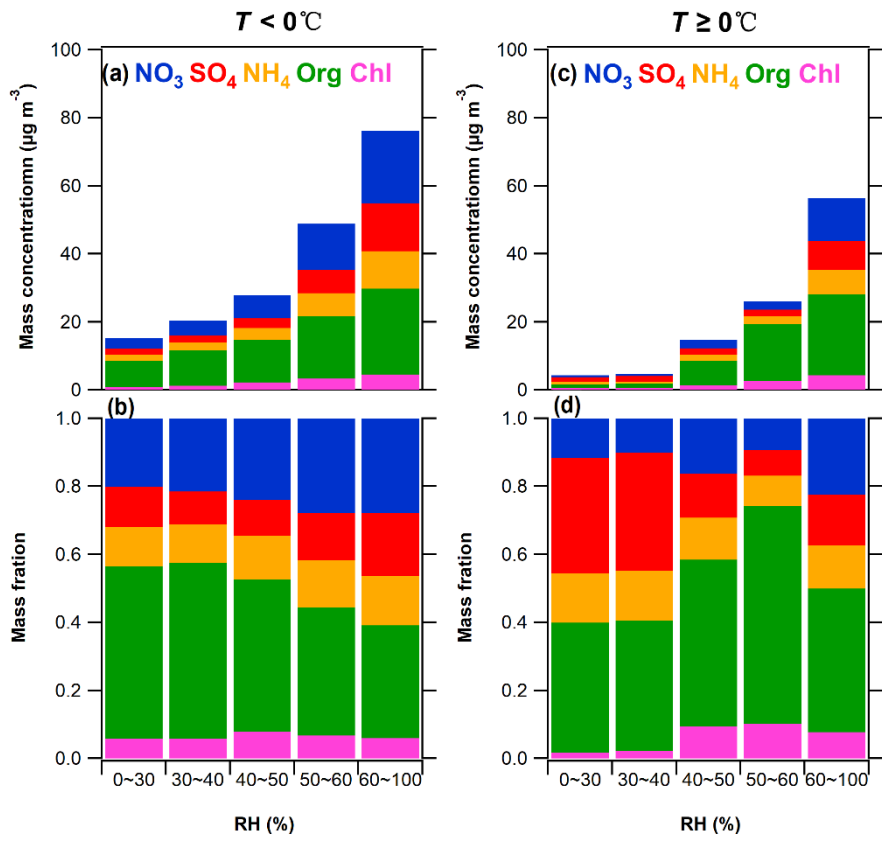
745 **Figure 10: Mass concentration and mass fractions of aerosol species dependence on RH at different temperature ranges. The left panel denotes the situation of -14 ℃< T < 0 ℃. The right panel denotes the situation of 0 ℃< T < 8 ℃.**

Supplementary

Table S1: Abbreviations and symbols used in this paper

Abbreviation/symbols	Full name/explanation
SP2	Single particle soot photometer (DMT Inc.)
HR-AMS	Aerodyne high–resolution time–of–flight aerosol mass spectrometer (Aerodyne Research Inc.)
DMA	Differential mobility analyzer (TSI Inc.)
CPC	Condensation particle counter (TSI Inc.)
rBC	Refractory black carbon measured by SP2 through laser-induced incandescence method
NR-PM1	Non-refractory submicron
Dc	Mass equivalent diameter of rBC core
Dp	Diameter of rBC-containing particles including rBC core and coatings
MMD	Mass median diameter
CMD	Count median diameter
RCT	Relative coating thickness of rBC-containing particles (Dp/Dc)
ACT	Absolute coating thickness of rBC-containing particles ((Dp-Dc)/2)
LEO	Leading-edge-only fitting method
E_{abs}	Absorption enhancement of rBC-containing particles compared to bare rBC
MAC	mass absorption cross-section of rBC-containing particles
σ_{abs}	absorption coefficient of rBC-containing particles

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Table S2: Representative cases of different episodes in Gucheng.

Selected cases	Month/Day/time	rBC mass loading ($\mu g\ m^{-3}$)	MMD (nm)	CMD (nm)	RCT(ACT) (Dc@170-190nm)	E_{abs}	MAC (m^2/g)	RH (%)	T (°C)	Wind speed ($m\ s^{-1}$)
Fog	12/7/21:10-12/9/13:30 12/9/17:30-12/10/14:20	3.1 ± 0.9	220	131	2.0 ± 0.3 (92.7 ± 23.8)	1.2 ± 0.1	5.4	100	-3.3 ± 1.1	0.63 ± 0.72
Before-snow	12/15/00:00-12/15/18:50	4.7 ± 0.7	213	120	1.4 ± 0.1 (39.9 ± 3.6)	1.2 ± 0.1	5.4	61.0 ± 10.1	-0.2 ± 2.5	0.6 ± 0.3
Snow	12/15/19:00-12/16/13:00	2.8 ± 1.1	210	126	1.5 ± 0.1 (46.9 ± 4.5)	1.3 ± 0.0	5.9	97.0 ± 4.9	0.1 ± 0.3	0.7 ± 0.3
After-snow	12/16/13:10-12/17/00:00	1.9 ± 0.2	206	129	1.7 ± 0.1 (61.3 ± 4.7)	1.4 ± 0.0	6.3	96.0 ± 1.9	0.7 ± 0.6	0.9 ± 0.5
EP2	12/12/19:00-12/14/00:00	3.4 ± 1.4	203	118	1.5 ± 0.1 (42.3 ± 9.4)	1.2 ± 0.1	5.4	50.4 ± 12.5	-0.4 ± 4.2	0.8 ± 0.5
EP4	12/22/00:00-12/26/08:00	1.2 ± 1.2	202	116	1.7 ± 0.1 (58.6 ± 12.7)	1.3 ± 0.1	5.7	81.0 ± 13.1	-1.8 ± 2.5	1.0 ± 0.8
EP5	12/27/00:00-12/29/20:00	3.4 ± 1.3	211	122	1.6 ± 0.2 (48.9 ± 14.1)	1.3 ± 0.1	5.6	62.4 ± 14.7	-1.4 ± 3.5	1.1 ± 0.7
EP6	12/30/00:00-12/31/00:00	0.5 ± 0.3	216	122	1.4 ± 0.1 (37.0 ± 9.5)	1.1 ± 0.0	5.2	22.7 ± 7.7	-4.8 ± 2.3	2.3 ± 1.2

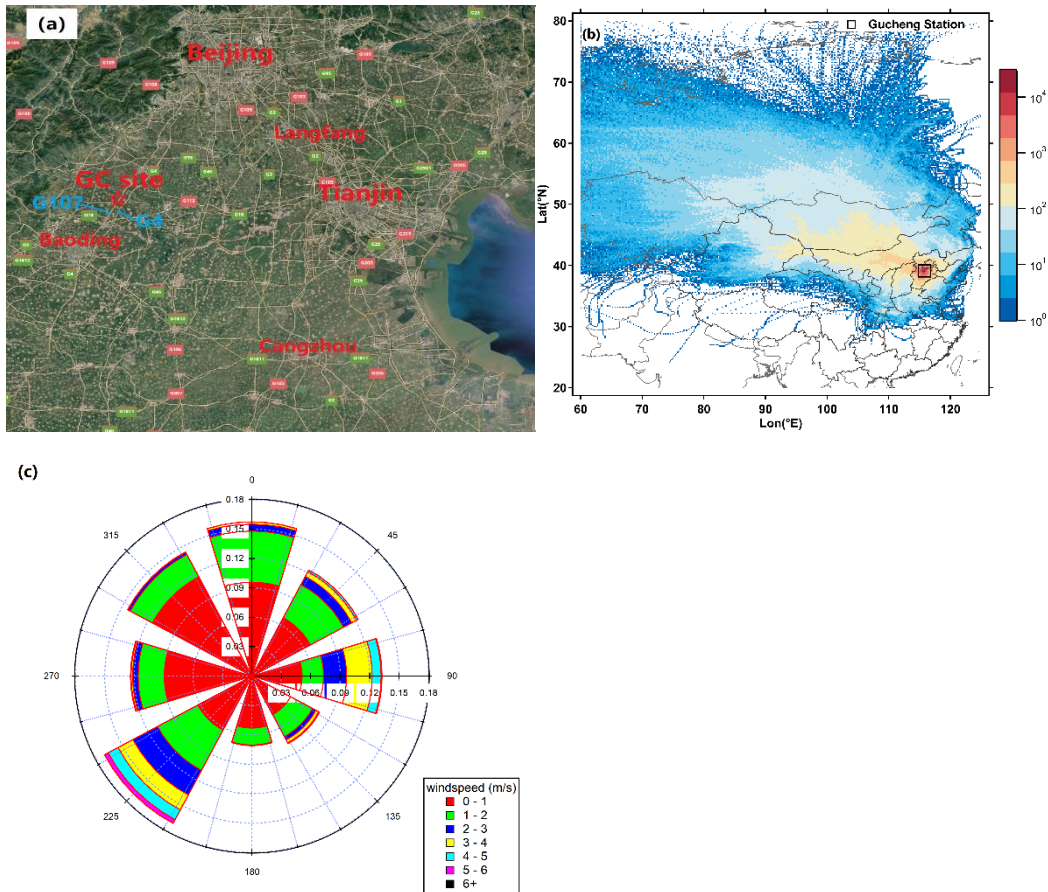



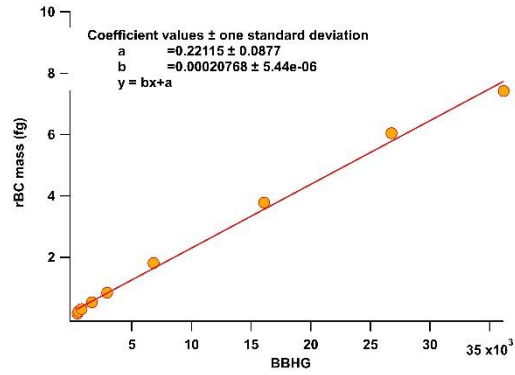
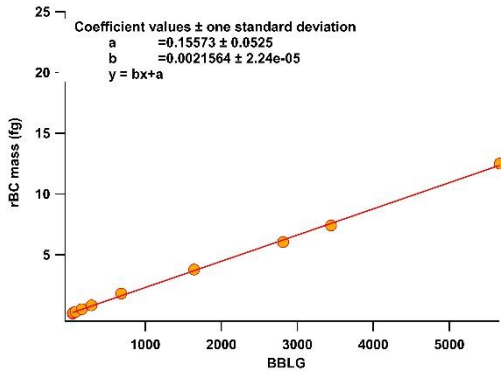
Figure S1: (a) Map of the experimental site (GC) that is located about 120 kilometers southwest of Beijing, adjacent to National Highway 107 (G107) and Beijing-Hong Kong-Macau Expressway (G4); the map was taken from Google Maps (©Google Maps 2021, https://www.google.com/maps, last access: 15 March 2021). (b) The sampling site was influenced by the historical air mass during the observation period, back trajectory analysis calculated by NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT). (c) The wind rose shows the frequency distribution of every 10 minutes averaged wind direction and speed throughout the experimental period.

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Figure S2: The calibration coefficient values for broadband high gain (BBHG) and broadband low gain (BBLG) of incandescence signals.
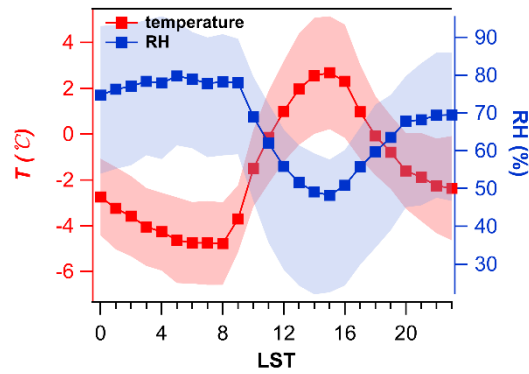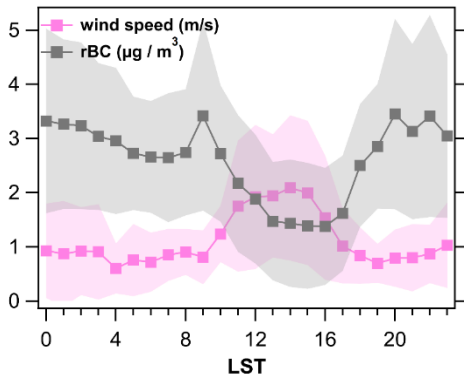
Figure S3: Diurnal variation in rBC mass loading, wind speed, temperature, and RH. The shaded areas denote the
765 **standard deviation.**
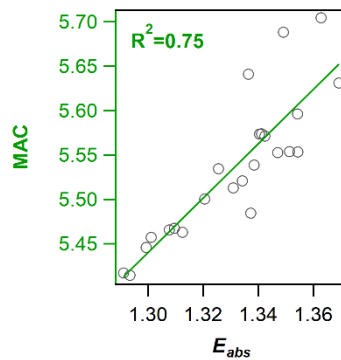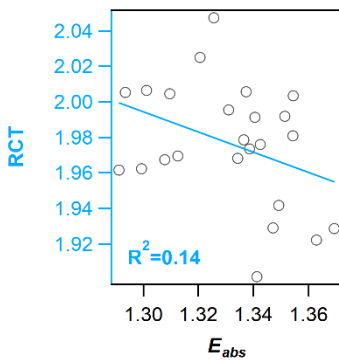
Figure S4: Correlation of E_{abs} with RCT and MAC in a diurnal cycle pattern.

Reply to the comments of anonymous reviewers on manuscript Entitled " Mixing state of refractory black carbon in fog and haze at rural sites in winter on the North China Plain"

We appreciate very much the patient and insightful comments and recommendations of the reviewers in improving this paper and our future research. We have revised the manuscript accordingly. Here, we will respond in black to all the comments one by one as follows.

Response to Reviewer #1

specific comments:

1. The Wegener-Bergeron-Findeisen process, or being called "cold-rain process" is used to describe ice crystal growth in a mixed phase clouds environment where it is a subsaturated environment for liquid water but a supersaturated environment for ice. As an assumption, ice crystals or tiny black carbon core can grow faster if the number density is much small compared to liquid water. Generally, it may a good explanation for the favorable formation of thickly coated rBC-containing particles. If possible, I would courage the authors to collect more evidence to have more solid conclusion.

Reply: Thanks for your reminder. After lots of studies, we found that it is hard to connect the observed changes in the size distributions of rBC particles near the surface level to the process in the cloud. The mechanism of wet scavenging and its effect on the distribution of aerosol particle size is complex (Sun et al., 2019; Zhao et al., 2020c), and we will investigate in more depth how wet scavenging affects rBC particle size in a future study. Here, a simple conclusion was obtained that the snow with relatively weak intensity (0.23 mm/h) may preferentially remove the small core size of rBC. We rewrite this section as:

The number size distribution followed the lognormal distribution and peaked at 122 nm (CMD) (Fig. 4a). Because most rBC particles (by number) emitted from urban areas were below the SP2 detection limit of 70 nm, the comparison of the number size distribution between urban and rural areas was meaningless (Schwarz et al., 2008). There was snowfall with the intensity of 0.23 mm/h during the observation period, and the mass and number size distributions of the rBC core mass equivalent diameter (Dc) during the snow event are illustrated in Fig. 4b-c. During before-snow, snow, and after-snow periods, the MMD was 213 nm, 210 nm, and 206 nm, decreasing by 3.3%, and the CMD was 120 nm, 126 nm, and 129 nm, an increase of 7.5%, respectively. We found that the number size distribution during the pre-snow and after-snow periods was consistent with a log-normal distribution. CMD of fresh BC usually ranges from 50 to 80 nm (Bond et al., 2013), so we extrapolated the fitting curves to 10 nm (Fig. 4c). The difference in areas between the two log-normal fit curves at Dc less than 121nm was 0.23, that is, the amount of rBC with Dc less than 121 nm decreased by 28.4% after snow. According to Fig. 3, the number

concentration of *r*BC with *Dc* below 100 nm decreased significantly while the number concentration of *r*BC with *Dc* about 200 nm decreased slightly during the snow case. This result is similar to the observation in Beijing that the snow with a precipitation intensity of 0.37 mm reduced the volume concentration of the small particle size (< 100 nm) fraction in the accumulation mode (Su et al., 2016). Previous studies found that wet deposition is the main mechanism for removing *r*BC-containing particles (Taylor et al., 2014). Wet removal plays an important role in determining the size distribution of *r*BC during vertical transport in the free troposphere (Moteki et al., 2012), and rainfalls, as a means of wet removal, preferentially remove large *r*BC-containing particles, resulting in a decrease in MMD after rain events (Liu et al., 2020b; Wang et al., 2018). However, the snow with relatively weak intensity may preferentially remove the small core size of *r*BC. Although the CMD increased moderately, the effect of removing rBC particles with a small core size (Dc) by snow was insignificant on the MMD (Liu et al., 2020a). The mechanism of wet scavenging and its effect of the distribution of aerosol particle size is complex (Zhao et al., 2020c; Sun et al., 2019) and we will investigate in more depth how wet scavenging affects *r*BC particle size in a future study. (line 331-358).

2. The basic information of instrumentation of HR-AMS is missing. Please add more introduction about the measurement of chemical composition. Note that HR-AMS provides the mass concentration of non-refractory species in PM1, and *r*BC measured by SP2 normally have MED peak at 200 nm. Therefore, it should be much careful about the relationship between the coating thickness of rBC and chemical composition in section 3.5.

Reply: Thanks for the reminder. We added the introduction of HR-AMS in the manuscript (line 125-130): A high–resolution time–of–flight aerosol mass spectrometer (HR-AMS, Aerodyne Research Inc., USA) was used to measure the non-refractory submicron (NR-PM1) aerosol species (organics, sulfate, nitrate, ammonium, and chloride). More details on this instrument can be found in previous studies (Jimenez, 2003; Kumar et al., 2016). Ambient relative humidity varied from 9% to 100% during the observation period, with an average relative humidity of 66%. And in section 3.5, five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< *T* < 0 ℃), and subcooled condition (0 ℃< *T* < 8 ℃)) to investigate the dependence of the mixing state of *r*BC on temperature, RH, and NR-PM1. We rediscussed this section as: The factors that affect the aging degree of *r*BC-containing particles include photochemical reactions, chemical coatings, and the meteorology of the environment (Laborde et al., 2013; Saathoff et al., 2003; Schnaiter, 2005). Here, we select *r*BC-containing particles with a *Dc* size of 170-190 nm. Fig. 8 shows RCT as a function of temperature and RH. Temperature and RH ranged from -14 to 8 ℃ and from 9 to 100%, respectively. In general, the changes of RH were mainly controlled by diurnal variations in temperature (Fig. S3). The higher the RH, the greater the RCT at the same temperature. At temperatures lower than -8 ℃, the RH was lower than 60%, the RCT was smaller than 1.5. However, at the intermediate temperature regime (-7 – 4 ℃), the RH required when the RCT exceeds 1.6 decreases with the increase of temperature. Larger RCT (RCT > 2) occurred in the temperature range of - 4 – 4 ℃ when the RH was higher than 50%. Indicating that lower RH (~60%)

required for *r*BC to form thick coatings under the subcooled condition (0 °C< T < 8 °C), while higher RH (~80%) required for *r*BC to form thick coatings under the supercooled condition (-14 °C< T < 0 °C).

840   In order to exclude the influence of special cases on the general results, data from fog days and snow periods were excluded to further investigate the aging of *r*BC. Based on the normalized frequency of data points under different RH and temperature ranges (Fig. 9d), five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 °C< T < 0 °C), and subcooled condition (0 °C< T < 8 °C)) to investigate the dependence of the mixing state of *r*BC on temperature and RH. The mean RCT,

845   E_{abs}, ACT, and aerosol species as a function of RH for 2 different temperatures regimes were shown in Fig. 9 and Fig. 10. The average RCT increased from ~1.4 to 1.6 and from ~1.4 to 1.7 and the average ACT increased from ~37 nm to 51 nm and from ~39 to 65 nm at the GC site as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The coating thickness of *r*BC particles was thicker under subcooled conditions than under supercooled conditions. This indicates that a suitable high temperature is conducive to the aging of *r*BC

850   particles. This variation was also supported by the gradual increases in mass concentration and mass fractions of secondary components as a function of RH in the environment. Under subcooled conditions, the coating thickness of *r*BC started to increase significantly at RH >40%, whereas this phenomenon did not occur until RH >60% under supercooled conditions. This shows that the RH required to form a thicker coating of *r*BC particles was lower under subcooled conditions. The high mass fraction of secondary components formed by liquid phase and heterogeneous

855   reactions in the environment under high RH and subcooled conditions were favorable to form coatings of *r*BC particles (Collier et al., 2018; Wu et al., 2016).

   The RH dependence of average E_{abs} was shown in Fig. 9b. The RH dependence of E_{abs} was likely caused by the RCT as supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH in the ambient (Fig. 10a-b). The average E_{abs} increased from ~ 1.2 to 1.3 and from 1.3 to 1.4 as RH increased from 0 to 100%

860   under the supercooled condition and subcooled condition, respectively. The E_{abs} under subcooled conditions was greater than that under supercooled conditions. The average E_{abs} remained almost constant after RH exceeded 60% under subcooled conditions. We suspected that the contribution of the aqueous-phase formation of secondary components to E_{abs} might be limited under high RH (Sun et al., 2021). (line 494-535)

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 3. There are lots of symbols and abbreviations in the manuscript, please summarize them in a table list.

Reply: Thanks for the reminder. We summarized symbols and abbreviations in Table S1 of the Supplement.

Table S1. Abbreviations and symbols used in this paper

Abbreviation/symbols	Full name/explanation
SP2	Single particle soot photometer (DMT Inc.)
HR-AMS	Aerodyne high–resolution time–of–flight aerosol mass spectrometer (Aerodyne Research Inc.)
DMA	Differential mobility analyzer (TSI Inc.)
CPC	Condensation particle counter (TSI Inc.)
rBC	Refractory black carbon measured by SP2 through laser-induced incandescence method
NR-PM1	Non-refractory submicron
Dc	Mass equivalent diameter of rBC core
Dp	Diameter of rBC-containing particles including rBC core and coatings
MMD	Mass median diameter
CMD	Count median diameter
RCT	Relative coating thickness of rBC-containing particles (Dp/Dc)
ACT	Absolute coating thickness of rBC-containing particles (($Dp-Dc$)/2)
LEO	Leading-edge-only fitting method
E_{abs}	Absorption enhancement of rBC-containing particles compared to bare rBC
MAC	mass absorption cross-section of rBC-containing particles
σ_{abs}	absorption coefficient of rBC-containing particles

880 4. line 120: "nonrefractory"is not a word, please change to "non-refractory"

Reply: Thanks for the reminder. We corrected the spelling. (line 126)

5. Line 424, The expression "in California 2013 during winter" seems wrong. Please check the grammar thoroughly the
885 manuscript.

Reply: Tanks for the reminder. We have modified the expression at two locations, "in California during winter in 2013" and "in Beijing 2018 during summer" (line 430 and line 432): Therefore, we compared the differences in E_{abs} between the GC site and other regions. A moderate E_{abs} (1.21 ± 0.09) at 870 nm was found in California during winter in 2013 which
890 was close to the present study (Zhang et al., 2016). The E_{abs} were slightly larger than the value simulated using core-shell in Beijing during summer in 2008 at 550 nm (1.15) (Liu et al., 2020b).

895 **Response to Reviewer #2**

Specific comments:

Line 76-78, this is not a complete sentence.

900 Reply: Thanks for the reminder. The sentence "Generally, a thicker coating of rBC–containing particles with increased hygroscopic properties and the ability to act as CCN under severe pollution conditions" has been changed to "Generally, rBC-containing particles with thicker coatings have higher hygroscopic properties and can act as CCN under severe pollution conditions." (line 79-81)

905 Line 79, "adsorption" should be "absorption"

Reply: We have corrected the word "absorption" (line 81).

Line 83-84, regarding the absorbing BC aerosols along with the high emissions over North China Plain, a reference could be mentioned here, Yang et al. (2016, doi: 10.1002/2016JD024938).

Reply: Thanks for the reminder. We have added this reference in the manuscript to support the point of "The North China 910 Plain is one of the BC hotspots with intensive anthropogenic emissions" (line 86-87): The North China Plain is one of the BC hotspots with intensive anthropogenic emissions (Cao et al., 2006; Yang et al., 2016).

Line 88, I would suggest using "During recent years" instead of "In recent years"

Reply: We have modified this expression in line 92: During recent years, an increasing number of observations have used SP2 to observe the mixing state of black carbon in China (Wang et al., 2014; Gong et al., 2016; Liu et al., 915 2020b; Pan et al., 2017).

Line 188, I wonder why the authors assumed 0 for the absorption part in the refractive index after coating.

Reply: We assumed a coating refractive index $n_s = 1.48 + 0i$. The absorption part in the refractive index is 0 because any coating material found in the atmosphere is nonabsorbing at the wavelength of 1064 nm. More details can be found in the reference Taylor et al. (2015, 10.5194/amt-8-1701-2015). We cite this literature in our study (line 920 185-187) Using a core refractive index $n_c = 2.26 + 1.26i$ and a coating refractive index $n_s = 1.48 + 0i$, we input the LEO fitted scattering signal and rBC core size into Mie calculations (Taylor et al., 2015).

Section 2.3, One one hand, it might be briefly mention that the HYSPLIT has been widely used for dispersion and trajectory analysis by providing some references, like the recent study by Fan et al. (2021a,b, doi: 10.1007/s11869-021-01023-9, doi:

 On the other hand, the potential uncertainties in the trajectory analysis might be worthy to briefly mentioned, which is highly associated with the accuracy in meteorology simulations.

Reply: Thanks for the reminder. We have added the suggestions mentioned by the reviewer: "The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model developed by NCEP (National Center for Environmental Prediction) and NCAR (National Center for Atmospheric Research) was widely used to simulate the air footprint area for dispersion and trajectory analysis, which was based on the Lagrangian transport model (https://ready.arl.noaa.gov/HYSPLIT_traj.php, last access: February 28th, 2020) (Cohen et al., 2015;Fan et al., 2021a;Fan et al., 2021b). The potential uncertainty of the HYSPLIT model is related to the meteorological assimilation data sets that drive the model simulation (Su et al., 2015)." (line 222-228)

Line 255, I would suggest changing "Liu et al., (2019)" to "Liu et al. (2019)"

Reply: Thanks for the reminder. We have changed it. (line 264)

Line 330-340, Another potential mechanism is the competition of hygroscopic growth of aerosol particles and wet scavenging. As indicated by Sun et al. (2019, doi: 10.1029/2019EA000717) and Zhao et al. (2020, doi: 10.3390/atmos11090906), aerosol mass concentration increases with precipitation when precipitation is weak and decreases with precipitation when precipitation is heavy. The snowfall case is similar to weak (or middle) precipitation case with relatively weak wet scavegnging capability, making the hygroscopic growth of aerosols essential and further resulting in less small particles and more large particles.

Reply: Thanks for your advice. In our study, the MMD and CMD are both an indication of the core size (Dc) of the rBC. We used the SP2 (Single-particle soot photometer) to measure the rBC, which were dried before they enter SP2, so the variation in the core size of the rBC is independent of hygroscopic growth, theoretically. While the aqueous phase processing or long-time aging processes can change the mixing state of rBC from externally irregular to internally compact and spherical shape (Pagels et al., 2009). The method of classifying the intensity of rainfall in the literature you have provided is very instructive to us. And we found that our result is similar to the observation in Beijing that the snow with a precipitation intensity of 0.37 mm reduced the volume concentration of the small particle size (< 100 nm) fraction in the accumulation mode (Su et al., 2016). So, in our study, a simple conclusion was obtained that the snow with relatively weak intensity (0.23 mm/h) may preferentially remove the small core size of rBC. The mechanism of wet scavenging and its effect on the distribution of aerosol particle size is complex (Zhao et al., 2020c; Sun et al., 2019) and we will investigate in more depth how wet scavenging affects rBC particle size in a future study. More details can be shown in the manuscript: The number size distribution followed the lognormal distribution and peaked at 122 nm (CMD) (Fig. 4a). Because most rBC particles (by number) emitted from urban areas were below the SP2 detection limit of 70 nm, the comparison of the number size distribution between urban and rural areas was meaningless (Schwarz et al., 2008). There was snowfall with the intensity of 0.23 mm/h during the observation period, and the mass and number size distributions of the rBC core mass equivalent diameter (Dc) during the snow event are illustrated in Fig. 4b-c. During before-snow, snow, and after-snow periods, the MMD was 213 nm, 210 nm, and 206 nm, decreasing by 3.3%, and the CMD was 120 nm, 126 nm, and 129 nm, an increase of 7.5%,

respectively. We found that the number size distribution during the pre-snow and after-snow periods was consistent with a log-normal distribution. CMD of fresh BC usually ranges from 50 to 80 nm (Bond et al., 2013), so we extrapolated the fitting curves to 10 nm (Fig. 4c). The difference in areas between the two log-normal fit curves at Dc less than 121nm was 0.23, that is, the amount of rBC with Dc less than 121 nm decreased by 28.4% after snow. According to Fig. 3, the number concentration of rBC with Dc below 100 nm decreased significantly while the number concentration of rBC with Dc about 200 nm decreased slightly during the snow case. This result is similar to the observation in Beijing that the snow with a precipitation intensity of 0.37 mm reduced the volume concentration of the small particle size (< 100 nm) fraction in the accumulation mode (Su et al., 2016). Previous studies found that wet deposition is the main mechanism for removing rBC-containing particles (Taylor et al., 2014). Wet removal plays an important role in determining the size distribution of rBC during vertical transport in the free troposphere (Moteki et al., 2012), and rainfalls, as a means of wet removal, preferentially remove large rBC-containing particles, resulting in a decrease in MMD after rain events (Liu et al., 2020b; Wang et al., 2018). However, the snow with relatively weak intensity may preferentially remove the small core size of rBC. Although the CMD increased moderately, the effect of removing rBC particles with a small core size (Dc) by snow was insignificant on the MMD (Liu et al., 2020a). The mechanism of wet scavenging and its effect of the distribution of aerosol particle size is complex (Zhao et al., 2020c; Sun et al., 2019) and we will investigate in more depth how wet scavenging affects rBC particle size in a future study. (line 331-358).

Line 367, "are" -> "were"

Reply: We have corrected the state. (line 378) Laborde et al. (2013) reported that rBC-containing particles emitted by traffic were essentially uncoated with a median coating thickness of 2±10 nm at a core size of 180-220 nm, the median ACT of rBC-containing particles emitted by biomass burning was approximately 20-40 nm, and the median ACT of rBC-containing particles in continental aged air masses reached ~80 nm.

Line 424-425, what do the authors mean "in California 2013 during winter"? Do they mean "in California during winter in 2013"? Also, "in Beijing 2018 during summer".

Reply: We have modified the expression at two locations. (line 430 and line 432): Therefore, we compared the differences in E_{abs} between the GC site and other regions. A moderate E_{abs} (1.21 ± 0.09) at 870 nm was found in California during winter in 2013 which was close to the present study (Zhang et al., 2016). The E_{abs} were slightly larger than the value simulated using core-shell in Beijing during summer in 2008 at 550 nm (1.15) (Liu et al., 2020b).

Line 449-451, current location is suggested to add here.

Reply: We added the study location "GC site" to the manuscript. (line 457) The mean rBC mass loading increased during the evening, with a peak (3.5 µg m^{-3}) at 20:00 LST and a minimum (1.4 µg m^{-3}) at 16:00 LST at the GC site, which was later than that in Beijing (Liu et al., 2020).

Line 518-519, I do not understand how the authors conclude that "-4~ 0 ℃ may be the most conducive temperature range" based on descriptions before this sentence. Please explain.

Reply: The conclusion was rediscussed. five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< T < 0 ℃), and subcooled condition (0 ℃< T < 8 ℃)) to investigate the dependence of the mixing state of rBC on temperature, RH, and NR-PM1. We concluded that relatively warm and high RH environments (RH> 50%, -4 ℃< T < 4 ℃) were more favorable to rBC aging than dry and cold environments (RH< 60%, T < -8 ℃). We rediscussed this section as: The factors that affect the aging degree of rBC-containing particles include photochemical reactions, chemical coatings, and the meteorology of the environment (Laborde et al., 2013; Saathoff et al., 2003; Schnaiter, 2005). Here, we select rBC-containing particles with a Dc size of 170-190 nm. Fig. 8 shows RCT as a function of temperature and RH. Temperature and RH ranged from -14 to 8 ℃ and from 9 to 100%, respectively. In general, the changes of RH were mainly controlled by diurnal variations in temperature (Fig. S3). The higher the RH, the greater the RCT at the same temperature. At temperatures lower than -8 ℃, the RH was lower than 60%, the RCT was smaller than 1.5. However, at the intermediate temperature regime (-7 – 4 ℃), the RH required when the RCT exceeds 1.6 decreases with the increase of temperature. Larger RCT (RCT > 2) occurred in the temperature range of -4 – 4 ℃ when the RH was higher than 50%. Indicating that lower RH (~60%) required for rBC to form thick coatings under the subcooled condition (0 ℃< T < 8 ℃), while higher RH (~80%) required for rBC to form thick coatings under the supercooled condition (-14 ℃< T < 0 ℃).

In order to exclude the influence of special cases on the general results, data from fog days and snow periods were excluded to further investigate the aging of rBC. Based on the normalized frequency of data points under different RH and temperature ranges (Fig. 9d), five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< T < 0 ℃), and subcooled condition (0 ℃< T < 8 ℃)) to investigate the dependence of the mixing state of rBC on temperature and RH. The mean RCT, E_{abs}, ACT, and aerosol species as a function of RH for 2 different temperatures regimes were shown in Fig. 9 and Fig. 10. The average RCT increased from ~1.4 to 1.6 and from ~1.4 to 1.7 and the average ACT increased from ~37 nm to 51 nm and from ~39 to 65 nm at the GC site as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The coating thickness of rBC particles was thicker under subcooled conditions than under supercooled conditions. This indicates that a suitable high temperature is conducive to the aging of rBC particles. This variation was also supported by the gradual increases in mass concentration and mass fractions of secondary components as a function of RH in the environment. Under subcooled conditions, the coating thickness of rBC started to increase significantly at RH >40%, whereas this phenomenon did not occur until RH >60% under supercooled conditions. This shows that the RH required to form a thicker coating of rBC particles was lower under subcooled conditions. The high mass fraction of secondary components formed by liquid phase and heterogeneous reactions in the environment under high RH and subcooled conditions were favorable to form coatings of rBC particles (Collier et al., 2018; Wu et al., 2016).

The RH dependence of average E_{abs} was shown in Fig. 9b. The RH dependence of E_{abs} was likely caused by the RCT as

1025   supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH
in the ambient (Fig. 10a-b). The average E_{abs} increased from ~ 1.2 to 1.3 and from 1.3 to 1.4 as RH increased from 0 to 100%
under the supercooled condition and subcooled condition, respectively. The E_{abs} under subcooled conditions was greater
than that under supercooled conditions. The average E_{abs} remained almost constant after RH exceeded 60% under subcooled
conditions. We suspected that the contribution of the aqueous-phase formation of secondary components to E_{abs} might be

1030   limited under high RH (Sun et al., 2021). (line 494-535)

Response to Reviewer #3

Major Comments:

Mentions about the impacts of droplet collision and WBF process

1035   In this study, air near the surface level was sampled for the SP2. It is hard to connect the observed changes in the size
distributions of rBC particles near the surface level to the process in cloud. Authors referenced the papers with respect this
topic (e.g., Ding et al., 2019). Ding et al. (2019) conducted the observation in cloud at a high-altitude site, indicating their
results were based on the direct evidence from in-situ characterization of in-cloud scavenging of rBC particles. Ground-level
measurements can be affected by not only the wet deposition process but also the horizontal advection. Authors need to clarify

1040   the differences between this and referred previous studies. So, this description especially in Abstract and Conclusion seems
overstating the observational results, and should be removed if not further verified.

Reply: Thanks for the reminder. It is hard to connect the observed changes in the size distributions of rBC particles near the
surface level to the process in the cloud. The mechanism of wet scavenging and its effect on the distribution of aerosol
particle size is complex (Sun et al., 2019; Zhao et al., 2020c), and we will investigate in more depth how wet

1045   scavenging affects rBC particle size in a future study. Here, a simple conclusion was obtained that the snow with
relatively weak intensity (0.23 mm/h) may preferentially remove the small core size of rBC. We rewrite
this section as:

The number size distribution followed the lognormal distribution and peaked at 122 nm (CMD) (Fig. 4a). Because
most rBC particles (by number) emitted from urban areas were below the SP2 detection limit of 70 nm, the

1050   comparison of the number size distribution between urban and rural areas was meaningless (Schwarz et al., 2008).
There was snowfall with the intensity of 0.23 mm/h during the observation period, and the mass and number size
distributions of the rBC core mass equivalent diameter (Dc) during the snow event are illustrated in Fig. 4b-c.
During before-snow, snow, and after-snow periods, the MMD was 213 nm, 210 nm, and 206 nm, decreasing by
3.3%, and the CMD was 120 nm, 126 nm, and 129 nm, an increase of 7.5%, respectively. We found that the number

1055   size distribution during the pre-snow and after-snow periods was consistent with a log-normal distribution. CMD
of fresh BC usually ranges from 50 to 80 nm (Bond et al., 2013), so we extrapolated the fitting curves to 10 nm
(Fig. 4c). The difference in areas between the two log-normal fit curves at Dc less than 121nm was 0.23, that is,

the amount of rBC with Dc less than 121 nm decreased by 28.4% after snow. According to Fig. 3, the number concentration of rBC with Dc below 100 nm decreased significantly while the number concentration of rBC with Dc about 200 nm decreased slightly during the snow case. This result is similar to the observation in Beijing that the snow with a precipitation intensity of 0.37 mm reduced the volume concentration of the small particle size (< 100 nm) fraction in the accumulation mode (Su et al., 2016). Previous studies found that wet deposition is the main mechanism for removing rBC-containing particles (Taylor et al., 2014). Wet removal plays an important role in determining the size distribution of rBC during vertical transport in the free troposphere (Moteki et al., 2012), and rainfalls, as a means of wet removal, preferentially remove large rBC-containing particles, resulting in a decrease in MMD after rain events (Liu et al., 2020b; Wang et al., 2018). However, the snow with relatively weak intensity may preferentially remove the small core size of rBC. Although the CMD increased moderately, the effect of removing rBC particles with a small core size (Dc) by snow was insignificant on the MMD (Liu et al., 2020a). The mechanism of wet scavenging and its effect of the distribution of aerosol particle size is complex (Zhao et al., 2020c; Sun et al., 2019) and we will investigate in more depth how wet scavenging affects rBC particle size in a future study. (line 331-358).

Data analyses of the coating thickness of rBC particles

As authors indicated in P14 L. 380-383, and previous studies (Schwarz et al., 2008a; Zhao et al., 2020) suggested, the coating thickness of rBC-containing particles with the core diameter smaller than 140-150 nm is not quantitatively evaluated because the light scattering signals originated from such small rBC particles does not exceed the noise level. Descriptions on relative and absolute coating thickness (RCT and ACT, respectively) for the size range of core diameter from 70-500 nm in section 3.3.1, and figures 5 and 6 (also S4 and S5) included such misleading results, even though authors noted this point in the manuscript. These results should not be included in order to minimize the misinterpretation by readers. Please consider the necessary modification to prevent readers from misunderstanding the results provided in this study.

Reply: Thanks for the reminder. To avoid misleading the reader, we have removed the figures and descriptions that contain misleading information. We concentrate on RCT and ACT for rBC particles with Dc at 170-190 nm and 190-210 nm in the new manuscript.

The analyses of RH and temperature dependence of mixing state and light absorption properties.

Air temperature widely varied from < -8ºC to > 4 ºC, indicating that the parts of the data sets were obtained in the subcooled and supercooled condition at the same RH level. When RH dependence shown in Figure 9 is separately analyzed by the condition of the air temperature (maybe lower or higher than 0ºC), the discussion will be more insightful.

Reply: Good advice. In the new manuscript, five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< T < 0 ℃), and subcooled condition (0 ℃< T < 8 ℃)) to investigate the dependence of the mixing state of rBC on temperature, RH, and NR-PM1. We

1090   concluded that relative warm and high RH environments (RH> 50%, -4 ℃< T < 4 ℃) were more favorable to rBC ageing than dry and cold environments (RH< 60%, T < -8 ℃). We rediscussed this section as: The factors that affect the aging degree of rBC-containing particles include photochemical reactions, chemical coatings, and the meteorology of the environment (Laborde et al., 2013; Saathoff et al., 2003; Schnaiter, 2005). Here, we select rBC-containing particles with a Dc size of 170-190 nm. Fig. 8 shows RCT as a function of temperature and

1095   RH. Temperature and RH ranged from -14 to 8 ℃ and from 9 to 100%, respectively. In general, the changes of RH were mainly controlled by diurnal variations in temperature (Fig. S3). The higher the RH, the greater the RCT at the same temperature. At temperatures lower than -8 ℃, the RH was lower than 60%, the RCT was smaller than 1.5. However, at the intermediate temperature regime (-7 – 4 ℃), the RH required when the RCT exceeds 1.6 decreases with the increase of temperature. Larger RCT (RCT > 2) occurred in the temperature range of -

1100   4 – 4 ℃ when the RH was higher than 50%. Indicating that lower RH (~60%) required for rBC to form thick coatings under the subcooled condition (0 ℃< T < 8 ℃), while higher RH (~80%) required for rBC to form thick coatings under the supercooled condition (-14 ℃< T < 0 ℃).

In order to exclude the influence of special cases on the general results, data from fog days and snow periods were excluded to further investigate the aging of rBC. Based on the normalized frequency of data points under different

1105   RH and temperature ranges (Fig. 9d), five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< T < 0 ℃), and subcooled condition (0 ℃< T < 8 ℃)) to investigate the dependence of the mixing state of rBC on temperature and RH. The mean RCT, E_{abs}, ACT, and aerosol species as a function of RH for 2 different temperatures regimes were shown in Fig. 9 and Fig. 10. The average RCT increased from ~1.4 to 1.6 and from ~1.4 to 1.7 and the average ACT increased from ~37 nm to

1110   51 nm and from ~39 to 65 nm at the GC site as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The coating thickness of rBC particles was thicker under subcooled conditions than under supercooled conditions. This indicates that a suitable high temperature is conducive to the aging of rBC particles. This variation was also supported by the gradual increases in mass concentration and mass fractions of secondary components as a function of RH in the environment. Under subcooled conditions, the coating thickness

1115   of rBC started to increase significantly at RH >40%, whereas this phenomenon did not occur until RH >60% under supercooled conditions. This shows that the RH required to form a thicker coating of rBC particles was lower under subcooled conditions. The high mass fraction of secondary components formed by liquid phase and heterogeneous reactions in the environment under high RH and subcooled conditions were favorable to form coatings of rBC particles (Collier et al., 2018; Wu et al., 2016).

1120   The RH dependence of average E_{abs} was shown in Fig. 9b. The RH dependence of E_{abs} was likely caused by the RCT as supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH in the ambient (Fig. 10a-b). The average E_{abs} increased from ~ 1.2 to 1.3 and from 1.3 to 1.4 as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The E_{abs} under subcooled conditions was greater than that under supercooled conditions. The average E_{abs} remained almost constant after RH exceeded 60% under subcooled

1125   conditions. We suspected that the contribution of the aqueous-phase formation of secondary components to E_{abs} might be limited under high RH (Sun et al., 2021). (line 494-535)

Minor Comments:

1130   Reply: We corrected the expression. (line 41) The relationship between the microphysical properties of rBC and meteorological conditions was also studied. Relative warm and high RH environments (RH> 50%, -4 °C< $T < 4$ °C) were more favorable to rBC ageing than dry and cold environments (RH< 60%, $T < $ -8 °C).

P3 L79; adsorption efficiency must be absorption efficiency.

Reply: We have corrected the word "absorption" (line 81).

1135   P4 L80; Does the thermal effect mean the direct effect of BC (heating the atmosphere)?

Reply: "the thermal effect" in the manuscript means the effect of heating the atmosphere caused by black carbon as an absorbing aerosol, that is, the direct effect of BC. We modified the sentence. line 83: Under high pollution and RH conditions, the absorption efficiency of black carbon was significantly increased. Reducing the emission of BC and related secondary coating precursors is an effective way to alleviate the direct radiation effect of BC in urban
1140   environments.

P4 L106; "relative, absolute" should be "relative and absolute".

Reply: We corrected the expression. line 109: (2) the relative and absolute coating thickness and optical properties of rBC-containing particles and their influencing factors

P5 section 2.1; Please describe the relative humidity conditions for the aerosol measurements, HR-AMS and SP2.

1145   Reply: Ambient relative humidity varied from 9% to 100% during the observation period, with an average relative humidity of 66% (line 128-129). The introduction of the HR-AMS was added in Sect 2.1 in the new manuscript (line 124-129): A high–resolution time–of–flight aerosol mass spectrometer (HR-AMS, Aerodyne Research Inc., USA) was used to measure the non-refractory submicron (NR-PM1) aerosol species (organics, sulfate, nitrate, ammonium, and chloride). More details on this instrument can be found in previous studies (Jimenez, 2003; Kumar
1150   et al., 2016). Ambient relative humidity varied from 9% to 100% during the observation period, with an average relative humidity of 66%. The detailed description of the SP2 was shown in Sect 2.2.1 in the manuscript.

P5 L126–127; To the best of my knowledge, "soft black carbon tube" is not widely used in the aerosol research community. Please describe the details about this product (manufacturer, what are the beneficial points to use this instead of
1155   other types of tubes, and impacts to the SP2 deployments (what is "black carbon" here, and does this make no contamination?))

Reply: Thanks for the reminder. The expression "soft black carbon tube" here is a direct translation from Chinese English. Tubes do not contain black carbon or elements that interfere with observation. We used this kind of tube in many *r*BC observations (Pan et al., 2017; Liu et al., 2020b; Liu et al., 2019b; Liu et al., 2020a). This kind of tube won't make any contamination. We corrected the "soft black carbon tube" to "soft tube". (line 134)

P5 L136; Only the detection of the incandescence signal cannot lead to derive the information about the boiling point temperature. In the SP2 community, the ratio of the signals observed at the detectors for narrow and broad wavelength bands is used for estimating the color temperature of thermal radiation from rBC-containing particles.

Reply: Thanks for the reminder. We corrected the expression as: "The ratio of the signals observed at the detectors for the narrowband and broadband is used for estimating the color temperature of thermal radiation from *r*BC–containing particles and, thereby, constrains particle composition." (line 143-146)

P7 section 2.2.3; Uncertainties to estimate the optical properties using the Mie theory is related to not only simple assumption of shell-core structure of rBC-containing particles but also spherical shape of rBC particles. This point should be included.

Reply: Thanks for the reminder. We corrected the expression as: "Simple assumptions of core-shell structure and ideal spheric shape of *r*BC particles, used in Mie theory to calculate optical properties, cause uncertainty due to the lack of observation of optical size information and morphology." (line 218-220)

P15 L399; "an uncoated rBC core diameter" is more simply *Dc*, isn't it?

Reply: Yes, "uncoated rBC core diameter" is *Dc*.

P15 L417; "refraction index" must be "refractive index".

Reply: Thanks for the reminder. We corrected the spelling. (line 425)

P16 L448; "clear noon-low and morning and evening-high pattern" is a lengthy phrase and hard to understand. Please rephrase this into more appropriate and easy-to-understand expression. Similar phrases are found in elsewhere (e.g., P17 L467). This also should be modified accordingly.

Reply: Thanks for the reminder. We modified the "clear noon-low and morning and evening-high pattern" and "day–high and night–low pattern" as "U shaped". (line 455, line 473) The temporal variation in *r*BC exhibited a clear "U" shaped pattern while the diurnal variation in wind speed was the opposite (Fig. S3). The temporal variation in RCT did not present a clear inverted "U" shaped pattern like Beijing during summer (Liu et al., 2020b).

P17 K 463; "an opposite trend" is not appropriate, because their relationships do not seem anticorrelated.

Reply: Thanks for the reminder. We have removed this sentence.

P18–19 section 3.5; The evaporation of aerosol components is discussed in relation to increases in the air temperature from around zero to higher. To the best of my knowledge, at least for ammonium nitrate, the temperature around zero is still favorable condition sustaining the existence of particulate phase nitrate. The discussions described here seems too simplistic. Please refer the studies about the researches on the equilibrium of submicron aerosols (e.g., Morino et al., 2006) and discuss more carefully this part.

Reply: Thanks for the reminder. In winter, nitrate is almost entirely present in the particulate form (Morino et al., 2006). We have removed this misleading expression. The conclusion was rediscussed. five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< $T < 0$ ℃), and subcooled condition (0 ℃< $T < 8$ ℃)) to investigate the dependence of the mixing state of rBC on temperature, RH, and NR-PM1. We rediscussed this section as: The factors that affect the aging degree of rBC-containing particles include photochemical reactions, chemical coatings, and the meteorology of the environment (Laborde et al., 2013; Saathoff et al., 2003; Schnaiter, 2005). Here, we select rBC-containing particles with a Dc size of 170-190 nm. Fig. 8 shows RCT as a function of temperature and RH. Temperature and RH ranged from -14 to 8 ℃ and from 9 to 100%, respectively. In general, the changes of RH were mainly controlled by diurnal variations in temperature (Fig. S3). The higher the RH, the greater the RCT at the same temperature. At temperatures lower than -8 ℃, the RH was lower than 60%, the RCT was smaller than 1.5. However, at the intermediate temperature regime (-7 – 4 ℃), the RH required when the RCT exceeds 1.6 decreases with the increase of temperature. Larger RCT (RCT > 2) occurred in the temperature range of - 4 – 4 ℃ when the RH was higher than 50%. Indicating that lower RH (~60%) required for rBC to form thick coatings under the subcooled condition (0 ℃< $T < 8$ ℃), while higher RH (~80%) required for rBC to form thick coatings under the supercooled condition (-14 ℃< $T < 0$ ℃).

In order to exclude the influence of special cases on the general results, data from fog days and snow periods were excluded to further investigate the aging of rBC. Based on the normalized frequency of data points under different RH and temperature ranges (Fig. 9d), five RH levels of 0%-30%, 30%-40%, 40%-50%, 50%-60%, and 60%-100% were selected in 2 different temperature ranges (supercooled condition (-14 ℃< $T < 0$ ℃), and subcooled condition (0 ℃< $T < 8$ ℃)) to investigate the dependence of the mixing state of rBC on temperature and RH. The mean RCT, E_{abs}, ACT, and aerosol species as a function of RH for 2 different temperatures regimes were shown in Fig. 9 and Fig. 10. The average RCT increased from ~1.4 to 1.6 and from ~1.4 to 1.7 and the average ACT increased from ~37 nm to 51 nm and from ~39 to 65 nm at the GC site as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The coating thickness of rBC particles was thicker under subcooled conditions than under supercooled conditions. This indicates that a suitable high temperature is conducive to the aging of rBC particles. This variation was also supported by the gradual increases in mass concentration and mass fractions of secondary components as a function of RH in the environment. Under subcooled conditions, the coating thickness of rBC started to increase significantly at RH >40%, whereas this phenomenon did not occur until RH >60% under

supercooled conditions. This shows that the RH required to form a thicker coating of rBC particles was lower under subcooled conditions. The high mass fraction of secondary components formed by liquid phase and heterogeneous reactions in the environment under high RH and subcooled conditions were favorable to form coatings of rBC particles (Collier et al., 2018; Wu et al., 2016).

The RH dependence of average E_{abs} was shown in Fig. 9b. The RH dependence of E_{abs} was likely caused by the RCT as supported by the gradual increase in the mass concentration and mass fraction of secondary components as a function of RH in the ambient (Fig. 10a-b). The average E_{abs} increased from ~ 1.2 to 1.3 and from 1.3 to 1.4 as RH increased from 0 to 100% under the supercooled condition and subcooled condition, respectively. The E_{abs} under subcooled conditions was greater than that under supercooled conditions. The average E_{abs} remained almost constant after RH exceeded 60% under subcooled conditions. We suspected that the contribution of the aqueous-phase formation of secondary components to E_{abs} might be limited under high RH (Sun et al., 2021). (line 494-535)

P19 L516; "u nder" (space between u and n) should be modified.

Reply: Thanks for the reminder. We corrected the spelling.

P20 L 558; Does "non-homogeneous" mean "heterogeneous"?

Reply: Thanks for the reminder. We have changed the presentation.

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