

Comments from the editor:

The 2 reviewers that reviewed your original manuscript have had a chance to review the revised manuscript and your replies. I have also examined this discussion, and based on the latest reviewer comments I would like for you to address these latest comments before full publication is considered. Specifically, between the 2 reviewers there are 3 major comments that need to be carefully addressed:

Thanks for your careful examination of our manuscript and allowing us to further revise the manuscript. We highly appreciate your comments and the reviewers' suggestions, which undoubtedly improve our manuscript. Based on these comments and suggestions, we have made careful modifications to the original manuscript.

Major Comments:

1. (Reviewer 1) There are a lot of grammatical errors in this paper. I only list a few here but encourage the authors to carefully review and correct their work prior to resubmission.

We want to thank the reviewer for their careful reviewing of the manuscript. We have carefully checked the full manuscript and revised the possible grammatical errors and/or typos, including those mentioned by reviewer 1#.

2. (Reviewer 1) I suggest that the authors provide more context for what is in the SI. For instance, a lot of the SPAMS description is in the SI. The authors need to point this out in the main text as well.

We agree with the comment. We have provided more context for the SPAMS measurements in the main text as suggested. Please refer to the detailed response below or the section 2.4 of the revised manuscript.

3. (Reviewer 2) I do think that there is still some confusion as the role of ionization efficiency and the complexities in equating concentration to relative intensity in a mass spectrum. I do think this should be clarified before the paper is published.

Thanks for the comment. We have clarified the limitation of using the relative peak area as a proxy for the relative abundance of the measured chemical composition. Please refer to the detailed response below for more details.

In addition to these major comments, please be sure to address all of Reviewer 1's specific comments.

Thank you for reminding. We have adequately addressed all the reviewers' specific comments in the following text. Below we give a detailed response to each of the concerns raised by the reviewers. The reviewers' comments are in the black text, followed by our response in the blue text.

Due to the nature of the comments, once you resubmit I'll carefully review and make a decision. Most likely I won't need to solicit any further comments from the reviewers.

Thanks so much and look forward to your revised manuscript.

Most sincerely, Jason Surratt

Comments from Review #1

General:

This paper compares single-particle measurements of particle size, composition, and morphology of in-cloud and interstitial particles collected at a rural site in China. This paper mainly comments on the role of aqueous chemistry in forming organic shells and the observation of more branched soot particles in cloud. The revision is much clearer and improved. I recommend publication after consideration of my comments.

Major Comments:

1. There are a lot of grammatical errors in this paper. I only list a few here but encourage the authors to carefully review and correct their work prior to submission.

Thanks for the comment. We have carefully checked and revised the full manuscript and supporting materials, including errors mentioned in the specific comments.

2. I suggest that the authors provide more context for what is in the SI. For instance, a lot of the SPAMS description is in the SI. The authors need to point this out in the main text as well.

Thanks for the suggestions. We have added a detailed description of the performance of the SPAMS in the section 2.4. Please refer to the detailed response below or the section 2.4 of the revised manuscript.

Specific Comments:

Abstract:

Line 33 and throughout the paper: “mineral” should be changed to “mineral dust”

It has been changed accordingly.

Lines 33-34: I still have some reservations about the “aged mixture” label for the most important particle type for RES. It is a non-descript particle class. I suggest that the authors re-think this classification to appeal to a broader audience.

Thanks for the helpful suggestion. In this study, “aged mixture” refers to the particles containing S-rich or OM and two or more refractory components. We have changed the word “aged mixture” to “aged refractory mixture”, which may be appropriate to represent aged particles containing two or more refractory components.

Line 38: I suggest that authors give a very quick definition of the D_f to appeal to a broader audience.

Thanks for the suggestion, the definition of the D_f has been added. Fractal dimension (D_f), a morphologic parameter to represent the branching degree of particles, for soot particles in the RES (1.82 ± 0.12) is lower than that in the INT (2.11 ± 0.09), which indicates that in-cloud processes may result in less compact soot.

Introduction:

1. Line 46: remove “at a certain supersaturation”

It has been removed accordingly.

2. Line 49: change “coagulation” to “coalescence”

It has been changed accordingly.

3. I think what is missing is some context for cloud droplet formation and evaporation cycles. I suggest describing the number of cloud/evaporation cycles aerosols go through

in their lifetime and that these processes critically shape particle size, morphology, and composition with impacts on clouds and direct radiative forcing.

Thanks for the suggestion. The number of cloud/evaporation cycles experienced by aerosols in their lifetime is quite uncertain, which is affected by atmospheric convection, pressure, water vapor content, aerosol particle size and other factors, so it is difficult to evaluate. In addition, the influence of the change of chemical and physical properties caused by cloud processes on the aerosol and radiative forcing after evaporation has also been summarized, please refer to lines 48-54:

On the other hand, in-cloud processes, including the formation of sulfate, nitrate, and water-soluble organics, and the physical processes such as collision and coalescence, would substantially change the physical and chemical properties of the activated particles (Kim et al., 2019; Ma et al., 2013; Roth et al., 2016; Wu et al., 2013). Given that the morphology and mixing state are vital in determining the optical properties of particles (Adachi et al., 2010; Wu et al., 2018), changes of these properties upon in-cloud processes would further affect the subsequent atmospheric processes (e.g., cloud activation, heterogeneous reactions) and radiative forcing of particles after droplet evaporation.

4. Line 68: incomplete sentence

Thanks for pointing out this. The sentence has been changed to “In particular, physical properties play a leading role in cloud activation of inorganic/organic mixed particles (Topping et al., 2007).”. Please refer to lines 67-68.

5. Lines 81-87: make it clear what D_f impacts: size? Radiative impacts?

Thanks for the comment. The impact of D_f on the size and radiation forcing has been described here. Please refer to lines 81-84:

Fractal dimension (D_f) is widely used to indicate the extent of branching of soot (Brasil

et al., 1999), with densely packed or compacted soot particles having higher D_f than chain-like branched clusters or open structures. When the branched soot particles become compact, their size will decrease, but the scattering cross-section will be greater (Radney et al., 2014; Zhang and Mao, 2020).

Radney, J. G., You, R., Ma, X., Conny, J. M., Zachariah, M. R., Hodges, J. T., and Zangmeister, C. D.: Dependence of Soot Optical Properties on Particle Morphology: Measurements and Model Comparisons, *Environmental Science & Technology*, 48, 3169-3176, 10.1021/es4041804, 2014.

Zhang, X., and Mao, M.: Radiative properties of coated black carbon aerosols impacted by their microphysics, *Journal of Quantitative Spectroscopy & Radiative Transfer*, 241, 106718, 10.1016/j.jqsrt.2019.106718, 2020.

6. Line 83: I think the authors mean aqueous processing

Thanks for pointing this out, and “water processing” has been changed to “aqueous processing”.

Methods:

1. Lines 126-127: I think the authors mean “intensity of elements including carbon and heavier elements”. As written, this would imply that they cannot detect carbon with their EDS.

Thanks for the comment. The sentence has been revised to “The EDS is coupled with TEM to detect the intensity of elements including carbon and heavier elements ($Z \geq 6$).”

2. Line 133: what are particles with rim?

Just like the particles in Figure 1e and 1f, there is a ring outside. That is the rim.

3. Line 140 and throughout: change “S-rich” to “S-rich particles”

It has been changed accordingly.

4. Line 148: please also cite [Moffet et al., 2008]

It has been cited.

5. Line 156: are particles focused by an aerodynamic lens or nozzle inlet?

An aerodynamic lens. The description has been revised to “Particles entering SPAMS were first focused into a beam of particles through an aerodynamic lens, and then their flight velocities were determined by two continuous diode Nd:YAG laser beams (532 nm).”

6. Line 156: the authors should clarify here that the sizing region measures the terminal velocity of the particles. These velocities are converted to vacuum aerodynamic diameter (not vacuum dynamic size) via calibration with polystyrene spheres of known size.

We agree with the comment. It has been revised as suggested. Please refer to the lines 158-161:

Particles entering the SPAMS were first focused into a beam of particles through an aerodynamic lens, and then their flight velocities were determined by two continuous diode Nd:YAG laser beams (532 nm). Polystyrene spheres of known size were used as a standard substance to calibrate the vacuum dynamic size (d_{va}) of particles.

7. Line 158: the authors should specify that ions are separated and analyzed using a dual polarity time-of-flight mass analyzer.

It has been supplemented accordingly.

8. Line 159: change “mass spectrometry” to “mass spectra”

It has been changed accordingly.

9. Lines 159-161: please cite [Bhave et al., 2002; Gross et al., 2000]

They have been cited.

10. Section 2.4: a lot of detail is in the SI. The authors need to at least mention that the particle analysis methods, calibration methods, and particle type characteristics can be found in the SI.

Thanks for pointing out this. Such information has been included in the main text: “A detailed description of particle analysis methods and particle type characteristics can be found in the supporting information.” Please refer to lines 168-169.

Results:

1. Lines 192: does the high abundance of aged mixture particles in the RES indicate a lot of collision/coalescence of droplets with refractory material?

Thanks for the comment. It is hard to confirm from the data collected in the present study. These particles may also be activated to droplets since they are internally mixed with hygroscopic materials such as sulfate.

2. Line 203: is an incomplete sentence

Thanks for pointing out this. The sentence, “And the samples of cloud event #2 sampled at noon”, has been changed to “And the samples of cloud event #2 were collected at noon.” Please refer to line 212.

3. Lines 235-243: I suggest moving this text to section 3.4

It has been revised as suggested.

4. Lines 249-253: The authors cannot completely rule out that larger S-OM particles were more likely to be activated based on their size alone.

Thanks for the comment. Indeed, our data cannot completely rule out the activation of larger S-OM particles. Instead, we discussed the possibility that such core-shell mixing structure in the RES may also be explained by the primary activation of S-OM particles with larger sizes. As shown in the main text, the fraction of OM-containing particles increases from 33% in the INT to 60% in the RES. It is unlikely due to the favorable activation of S-OM or aged refractory mixture, since mixing with OM generally lower the hygroscopicity of inorganic-dominant particles (e.g., S-rich) (Brooks et al., 2004; Pierce et al., 2012). OM coating at the same site has been shown to inhibit the CCN activation of soot-containing particles (Zhang et al., 2017a). Besides, Evidence from the collocated SPAMS shows that the ratios of relative peak area between organics and sulfate are similar between the INT and particles before the cloud event, whereas they are higher in the RES (Table S2). This is corresponding to the production of oxidized organics during in-cloud processes (Zhang et al., 2017b), consistent with the TEM-EDS results. And thus, we suspect that it is most probably attributed to the in-cloud formation of OM on the surface of some S-rich particles, shifting the dominant particle type from S-rich to S-OM particles.

5. Line 258: Please clarify this sentence. It seems that the authors are implying that they do not have OM mixed with sulfate even though they just mentioned S-OM particles.

Thanks for pointing out this. The sentence has been changed to “Such a mixing structure is similar to those observed in the Arctic, background, or rural atmosphere (Hiranuma et al., 2013; Li et al., 2016; Yu et al., 2019), but is different from other findings in polluted areas, where OM-containing particles mainly existed in homogenous-like and thinly coated structures (Li et al., 2016).”. Please refer to lines 257-260.

6. Lines 268-270: this sentence needs to be rephrased. I do not understand it.

Thanks for the comment, and the sentence has been revised to “Droplets are expected to dissolve more volatile organic compounds (Chakraborty et al., 2016) with higher O/C, and the release of these compounds during droplet evaporation would result in underestimation of O/C in the RES.”

7. Lines 277-279: did the SPAMS see higher peak areas of m/z +43, which was found to be indicative of SOA and associated with high concentrations of O_3 (see [X Qin et al., 2012]).

Thanks for the comments. We did find a higher relative peak area of m/z $43C_2H_3O^+$ in the RES than that in the INT during cloud event #2 with higher concentration of O_3 . Such information has also been included in the discussion:

“Consistently, the relative peak area of m/z $43C_2H_3O^+$ in the RES is higher than that in the INT during cloud event #2 (Figure S7), indicative of the favorable formation of oxidized organic compounds (Qin et al., 2012; Zhang et al., 2017b).”

Qin, X., Pratt, K. A., Shields, L. G., Toner, S. M., and Prather, K. A.: Seasonal comparisons of single-particle chemical mixing state in Riverside, CA, *Atmospheric Environment*, 59, 587-596, 10.1016/j.atmosenv.2012.05.032, 2012.

Zhang, G., Lin, Q., Peng, L., Yang, Y., Fu, Y., Bi, X., Li, M., Chen, D., Chen, J., Cai, Z., Wang, X., Peng, P., Sheng, G., and Zhou, Z.: Insight into the in-cloud formation

of oxalate based on in situ measurement by single particle mass spectrometry, Atmospheric Chemistry and Physics, 17, 13891-13901, 10.5194/acp-17-13891-2017, 2017b.

Figures

1. I suggest adding an asterisk by elements that were considered background (e.g., Si and Cu).

Thanks for the comment. We add the asterisk on the background elements including C, Si, and Cu.

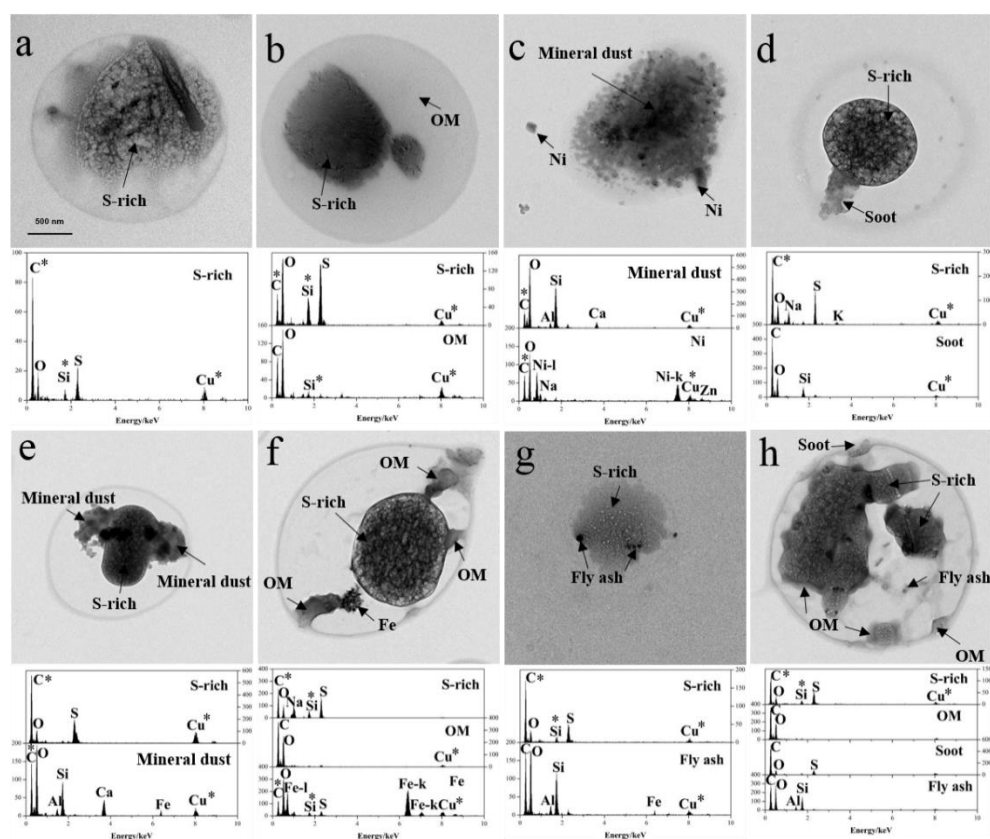


Figure 1. TEM images and EDS spectra of individual RES and INT particles with different particle types: (a) S-rich; (b) S-OM; (c) refractory; (d) aged soot; (e) aged mineral dust; (f) aged metal; (g) aged fly ash; (h) aged refractory mixture. Asterisk (*) represents the background element.

SI

1. Section 3: what software was used to import SPAMS spectra into Matlab? How many particles were analyzed?

The information on particle sizes and mass spectra is imported into the Matlab for subsequent analysis using the FATES toolkit (Sultana et al., 2017). A total of 117,436 particles from the SPAMS were analyzed. Such information has been included in section 3 of SI as suggested.

Sultana, C. M., Cornwell, G. C., Rodriguez, P., and Prather, K. A.: FATES: a flexible analysis toolkit for the exploration of single-particle mass spectrometer data, *Atmos. Meas. Tech.*, 10, 1323-1334, doi:10.5194/amt-10-1323-2017, 2017.

2. Section 3 also needs more citations for the characterization of these particle types. I suggest citing [Denkenberger et al., 2007; X Qin et al., 2012; X Y Qin and Prather, 2006] for the EC and HMOC particle types. I suggest [Silva et al., 2000] for the dust particle type. I suggest [Gaston et al., 2011] for the sea salt particle type. I suggest [Angelino et al., 2001; Pratt et al., 2009] for the amine particle type.

We agree with the comments and have cited these references as suggested.

References:

Angelino, S., D. T. Suess, and K. A. Prather (2001), Formation of aerosol particles from reactions of secondary and tertiary alkylamines: Characterization by aerosol time-of-flight mass spectrometry, *Environ. Sci. Tech.*, 35(15), 3130-3138.

Bhave, P. V., J. O. Allen, B. D. Morrical, D. P. Fergenson, G. R. Cass, and K. A. Prather (2002), A field-based approach for determining ATOFMS instrument sensitivities to ammonium and nitrate, *Environ. Sci. Tech.*, 36(22), 4868-4879.

Denkenberger, K. A., R. C. Moffet, J. C. Holecek, T. P. Rebotier, and K. A. Prather (2007), Real-time, single-particle measurements of oligomers in aged ambient aerosol particles, *Environ. Sci. Tech.*, 41(15), 5439-5446.

Gaston, C. J., H. Furutani, S. A. Guazzotti, K. R. Coffee, T. S. Bates, P. K. Quinn, L. I. Aluwihare, B. G. Mitchell, and K. A. Prather (2011), Unique ocean-derived particles serve as a proxy for changes in ocean chemistry, *J. Geophys. Res.-[Atmos.]*, 116, D18310, doi:18310.11029/12010JD015289.

Gross, D. S., M. E. Galli, P. J. Silva, and K. A. Prather (2000), Relative sensitivity factors for alkali metal and ammonium cations in single particle aerosol time-of-flight mass spectra, *Anal. Chem.*, 72(2), 416-422.

Moffet, R. C., et al. (2008), Characterization of aerosols containing Zn, Pb, and Cl from an industrial region of Mexico City, *Environ. Sci. Tech.*, 42(19), 7091-7097.

Pratt, K. A., L. E. Hatch, and K. A. Prather (2009), Seasonal volatility dependence of ambient particle phase amines, *Environ. Sci. Tech.*, 43(14), 5276-5281.

Qin, X., K. A. Pratt, L. G. Shields, S. M. Toner, and K. A. Prather (2012), Seasonal comparisons of single-particle chemical mixing state in Riverside, CA, *Atmos. Environ.*, 59, 587-596.

Qin, X. Y., and K. A. Prather (2006), Impact of biomass emissions on particle chemistry during the California Regional Particulate Air Quality Study, *International Journal of Mass Spectrometry*, 258(1-3), 142-150.

Silva, P. J., R. A. Carlin, and K. A. Prather (2000), Single particle analysis of suspended soil dust from Southern California, *Atmos. Environ.*, 34, 1811-1820.

Comments from Review #2

Thank you for responding to the comments! I do think that there is still some confusion as the role of ionization efficiency and the complexities in equating concentration to relative intensity in a mass spectrum. I do think this should be clarified before the paper is published.

We agree with the comment. To make it clear, we have clarified the limitation of using relative peak area as a proxy for the relative abundance of the measured species in Section 2.4:

“The relative peak area of characteristic peaks for each species in the mass spectra is generally applied to indicate its relative abundance in the particle (Bhave et al., 2002; Gross et al., 2000). However, it is still challenging to provide quantitative information on chemical compositions, mainly attributed to the different ionization efficiency and the complex matrix effects for various types of particles.”

Bhave, P. V., Allen, J. O., Morrical, B. D., Fergenson, D. P., Cass, G. R., and Prather, K. A.: A field-based approach for determining ATOFMS instrument sensitivities to ammonium and nitrate, *Environmental Science & Technology*, 36, 4868-4879, 10.1021/es015823i, 2002.

Gross, D. S., Galli, M. E., Silva, P. J., and Prather, K. A.: Relative sensitivity factors for alkali metal and ammonium cations in single particle aerosol time-of-flight mass spectra, *Analytical Chemistry*, 72, 416-422, 10.1021/ac990434g, 2000.

1 **Impact of in-cloud aqueous processes on the chemical**
2 **compositions and morphology of individual atmospheric**
3 **aerosols**

4 Yuzhen Fu^{1, 2}, Qin hao Lin^{1, #}, Guohua Zhang^{1, 3, *}, Yuxiang Yang^{1, 2}, Yiping Yang^{2, 4},
5 Xiufeng Lian^{1, 2}, Long Peng^{1, 2}, Feng Jiang^{1, 2, ##}, Xinhui Bi^{1, 3, *}, Lei Li⁵, Yuanyuan
6 Wang⁶, Duohong Chen⁷, Jie Ou⁸, Xinming Wang^{1, 3}, Ping'an Peng^{1, 3}, Jianxi Zhu⁴,
7 Guoying Sheng¹

8 ¹ State Key Laboratory of Organic Geochemistry and Guangdong Key Laboratory of Environmental
9 Protection and Resources Utilization, Guangzhou Institute of Geochemistry, Chinese Academy of
10 Sciences, Guangzhou 510640, PR China

11 ² University of Chinese Academy of Sciences, Beijing 100049, PR China

12 ³ Guangdong-Hong Kong-Macao Joint Laboratory for Environmental Pollution and Control, Guangzhou
13 510640, PR China

14 ⁴ CAS Key Laboratory of Mineralogy and Metallogeny & Guangdong Provincial Key Laboratory of
15 Mineral Physics and Materials, Guangzhou Institute of Geochemistry, CAS, Guangzhou 510640, PR
16 China

17 ⁵ Institute of Mass Spectrometer and Atmosphere Environment, Jinan University, Guangzhou 510632,
18 PR China

19 ⁶ Department of Atmospheric Science, School of Earth Science, Zhejiang University, Hangzhou 310027,
20 PR China

21 ⁷ State Environmental Protection Key Laboratory of Regional Air Quality Monitoring, Guangdong
22 Environmental Monitoring Center, Guangzhou 510308, PR China

23 ⁸ Shaoguan Environmental Monitoring Center, Shaoguan 512026, PR China

24 [#] now at: Guangdong Key Laboratory of Environmental Catalysis and Health Risk Control, Guangzhou
25 Key Laboratory Environmental Catalysis and Pollution Control, School of Environmental Science and
26 Engineering, Institute of Environmental Health and Pollution Control, Guangdong University of
27 Technology, Guangzhou 510006, PR China.

28 ^{##} now at: Institute of Meteorology and Climate Research, Karlsruhe Institute of Technology, Eggenstein-
29 Leopoldshafen 76344, Germany.

30 **Correspondence to:* Guohua Zhang (zhanggh@gig.ac.cn) and Xinhui Bi (bixh@gig.ac.cn)

31 **Abstract.** The composition, morphology, and mixing structure of individual cloud residues (RES) and interstitial
32 particles (INT) at a mountain-top site were investigated. Eight types of particles were identified, including sulfate-
33 rich (S-rich), S-organic matter (OM), aged soot, aged mineral dust, aged fly ash, aged metal, refractory, and aged
34 refractory mixture. A shift of dominant particle types from S-rich (29%) and aged soot (27%) in the INT to S-OM
35 (24%) and aged refractory mixture (22%) in the RES is observed. In particular, particles with organic shells are
36 enriched in the RES (30%) relative to the INT (12%). Our results highlight that the formation of more oxidized
37 organic matter in the cloud contributes to the existence of organic shells after cloud processing. Fractal dimension
38 (D_f), a morphologic parameter to represent the branching degree of particles, for soot particles in the RES ($1.82 \pm$
39 0.12) is lower than that in the INT (2.11 ± 0.09), which indicates that in-cloud processes may result in less compact
40 soot. This research emphasizes the role of in-cloud processes on the chemistry and microphysical properties of
41 individual particles. Given that organic coatings may determine the particle hygroscopicity, activation ability, and
42 heterogeneous chemical reactivity, the increase of OM-shelled particles upon in-cloud processes should have
43 considerable implications.

44 1 Introduction

45 Aerosol-cloud interaction is regarded as one of the most significant sources of uncertainty in assessing the
46 radiative forcing of aerosols so far (IPCC, 2013). On the one hand, aerosols can participate in the formation of
47 cloud droplets, which is primarily influenced by their chemical composition and size (Fan et al., 2016; Maskey et
48 al., 2017; Ogawa et al., 2016; Raymond and Pandis, 2002; Zelenyuk et al., 2010). On the other hand, in-cloud
49 processes, including the formation of sulfate, nitrate, and water-soluble organics, and the physical processes such
50 as collision and **coalescence**, would substantially change the physical and chemical properties of the activated
51 particles (Kim et al., 2019; Ma et al., 2013; Roth et al., 2016; Wu et al., 2013). Given that the morphology and
52 mixing state are vital in determining the optical properties of particles (Adachi et al., 2010; Wu et al., 2018),
53 changes of these properties upon in-cloud processes would further affect the subsequent atmospheric processes
54 (e.g., cloud activation, heterogeneous reactions) and radiative forcing of particles after droplet evaporation.

55 Understanding the morphology and mixing state of particles upon in-cloud processes is of considerable
56 significance to improve the knowledge of aerosol-cloud interactions. For instance, Zelenyuk et al. (2010) found
57 that both cloud droplet residues (RES) and interstitial particles (INT, or unactivated particles in the cloud) are
58 mainly composed of organics, sulfate, biomass burning particles, and processed sea salt at the North Slope of
59 Alaska. Kamphus et al. (2010) observed that 92% of RES are particles containing sulfates, organics, and nitrate
60 at the Jungfrauoch (Swiss Alps). At Mt. Tai, Liu et al. (2018b) observed that the main particle types are S
61 (sulfate)-soot (36%), S-fly ash/metal-soot (26%), and S-rich (24%) for RES and S-rich (61%), S-soot (15%) and
62 soot (15%) for INT. These results indicate that both RES and INT present complex mixtures, and carbonaceous
63 matters (i.e., organic materials (OM) and soot) are critical materials in the cloud mass.

64 While extensive studies are reporting the extent of aqueous phase processing on the modification of aerosol
65 bulk (e.g., mass) and/or chemical (e.g., mixing state, hygroscopicity) properties (Chakraborty et al., 2016; Ervens
66 et al., 2011), the influence of in-cloud processes on the physical properties (e.g., shape, mixing structure) of
67 individual particles is still ambiguous. In particular, **physical properties play a leading role in the cloud activation
68 of inorganic/organic mixed particles** (Topping et al., 2007). A hydrophobic organic-rich coating will form on a
69 hygroscopic particle core if liquid-liquid phase separation occurs (Song et al., 2013). Besides, the distribution of
70 organics and its association with other aerosol types is also crucial for the accurate calculation of its radiative
71 effects (Zhu et al., 2017). However, to what extent in-cloud processes play a role in reshaping the distribution of
72 organic and inorganic compositions remains unknown, although such coating structures have been identified in
73 ambient aerosols (Adachi and Buseck, 2008; Li and Shao, 2010; Yu et al., 2019). Considering that **secondary
74 formation during** in-cloud processes contributes to a substantial fraction (up to 60%) of organic aerosols (Ervens
75 et al., 2011; Liu et al., 2012; Myriokefalitakis et al., 2011; Spracklen et al., 2011), the influence of this process in
76 atmospheric chemistry cannot be neglected.

77 For another type of carbonaceous material (i.e., soot), there is extensive evidence **showing** that the absorption
78 and cloud activation of soot-containing particles can be significantly affected by coatings (Adachi et al., 2010;
79 Wu et al., 2018; Moffet and Prather, 2009). The critical factors to **accurately predict** such impact include the

80 amount and nature of the coating material, the exact particle morphology, and the size distribution (Qiu et al.,
81 2012; Radney et al., 2014). Fractal dimension (D_f) is widely used to indicate the extent of branching of soot (Brasil
82 et al., 1999), with densely packed or compacted soot particles having higher D_f than chain-like branched clusters
83 or open structures. When the branched soot particles become compact, their size will decrease, but the scattering
84 cross-section will be greater (Radney et al., 2014; Zhang and Mao, 2020). While some studies have found that
85 soot restructuring occurs after aqueous processing (Bhandari et al., 2019; Ma et al., 2013; Mikhailov et al., 2006),
86 or being coated by OM (Spencer and Prather, 2006) and sulfate (Zhang et al., 2008), Khalizov et al. (2013)
87 suggested that soot with thin organic coating did not become more compact under high humidity. Besides, the
88 morphology and mixing structure of soot involving the formation of organics upon cloud processing is also poorly
89 constrained.

90 To further improve our understanding of the morphology and mixing structures between the various
91 components within individual RES and INT, we conducted a 25-day field observation of cloud events at a
92 background site in southern China. A transmission electron microscope (TEM) combined with energy-dispersive
93 X-ray spectrometry (EDS) was used to analyze the chemical composition, size, morphology, and mixing structure
94 of individual RES and INT. Previously, the chemical composition and mixing state of RES at the same site have
95 been investigated with a single particle aerosol mass spectrometer (SPAMS) (Lin et al., 2017; Zhang et al., 2017a).
96 Herein, we focus on the mixing structure (e.g., chemical compositions and morphology) of individual particles,
97 in particular, OM-containing particles. Meanwhile, particle types and mixing state of RES and INT are also
98 discussed. The difference between the mixing structure of RES and INT may indicate the impact of in-cloud
99 aqueous processes.

100 **2 Materials and Methods**

101 **2.1 Sampling site**

102 Sampling was conducted at the top of Mt. Tianjing (112°53'56" E, 24°41'56" N; 1690 m above sea level) in
103 southern China from 18 May to 11 June 2017. The sampling site is located in a natural preserve, and it is almost
104 unaffected by local anthropogenic sources. It is about 50 km and 350 km away from the north of the Pearl River
105 Delta (PRD) region and the South China Sea, respectively.

106 **2.2 Collection of RES and INT**

107 A cloud event was identified with visibility below a threshold of 3 km and relative humidity (RH) above a
108 threshold of 95%, using a ground-based counterflow virtual impactor (GCVI, model 1205, Brechtel Mfg. Inc.,
109 USA). The GCVI was automatically triggered when there was a cloud event, whereas it was not allowed to sample
110 when a precipitation sensor detected rain or snow. Then cloud droplets were introduced into the GCVI, followed
111 by removing water in an evaporation chamber (40 °C) to obtain RES. The sampling process might experience
112 some particle loss due to the evaporation of highly volatile substances. The droplet cut size, at which the
113 transmission efficiency of CVI is 50%, was set at a size larger than 7.5 μm (Shingler et al., 2012). INT was

114 sampled using another inlet (PM_{2.5} cyclone inlet, with a flowrate of 5 lpm), followed by passing through a silica
115 gel diffusion dryer.

116 A DKL-2 sampler (Genstar Electronic Technology Co., Ltd., China) was used to collect RES and INT on copper
117 grids coated with carbon film with an airflow of 1 L min⁻¹. The collection efficiency of the sampler is 50% at a
118 particle size of 80 nm, assuming the particle density is 2 g cm⁻³. To avoid particle overlapping, the sampling
119 duration was set within 10 minutes. All samples were placed in a sealed plastic sample box and stored in a
120 desiccator at room temperature for subsequent analysis.

121 The information about cloud events and samples are summarized in Table 1. We focused on three cloud events
122 (#1, #2, and #3), with a duration of 14, 34, and 47 hours, respectively. RES and INT samples from these cloud
123 events were analyzed, with INT not available for the cloud event #1. To minimize the influence of rapid change
124 of cloud condition, all the samples were collected during the stable and mature periods (Visibility < 100 m).

125 2.3 TEM analysis of RES and INT

126 Chemical composition, size, and morphology of individual RES and INT were characterized by a TEM (FEI
127 Talos F200S) operated at 200 kV. TEM/EDS is a very effective tool to analyze the microscopic characteristics of
128 individual particles. The resolution of images between 1 μm and 100 nm can be magnified from 7,000 to 36,000
129 fold, which depended on the size of particles. The EDS is coupled with TEM to detect the intensity of elements
130 including carbon and heavier elements (Z ≥ 6). The produced X-rays signal in the EDS system is detected by a
131 silicon (Si) drift detector (SDD), and thus Si is not considered in the discussion. Cu is also not considered due to
132 the interference from the copper grids. In the TEM vacuum chamber, some volatile substances (e.g., ammonium
133 nitrate (NH₄NO₃) and volatile organic matter) would be lost. Moreover, volatile materials are often sensitive to
134 strong electron beams. Due to the analysis error of volatile materials, TEM/EDS studies typically focus on
135 refractory compositions. Using an image analysis software (ImageJ), the equivalent circle diameters (ECD) of all
136 particles can be obtained from the scanned images from the TEM. For particles with rim, only the nucleus is
137 counted, because the rims contain only a small amount of OM. Overall, 780 particles, including RES and INT,
138 were analyzed.

139 Base on various element spectra, RES and INT were mainly classified as sulfate-rich (S-rich), carbonaceous
140 material, mineral dust, metal, and fly ash (Li et al., 2016; Twohy and Anderson, 2008). Elemental compositions
141 of S-rich particles were dominated by S and O, and some of them were associated with minor N, K and Na. Low
142 intensity of N could be due to the evaporation of ammonia nitrate under the high energy electron beam (Smith et
143 al., 2012). This led to the bubbly appearance of S-rich particles. In this case, S-rich particles represented secondary
144 inorganic particles. The elemental compositions of carbonaceous materials were characteristics of abundant C and
145 minor O. Carbonaceous materials were divided into soot and OM according to different morphology. Soot was
146 composed of tens to hundreds of carbon spheres ranging from 21 to 108 nm in diameter (average diameter was
147 47.7 nm), which often displayed botryoidal aggregates. OM did not have a chain-like structure, which generally
148 exhibited amorphous state and spherical or irregular shapes. Mineral dust particles were consisted of Si, Al, Ca,
149 O and minor Fe. Mineral dust was mainly clay, feldspar, calcite and gypsum, usually showing irregular shapes.
150 Metal particles were represented as Fe, Zn, Ti, Mn, or Ni. Metal particles were characteristic of spherical,

151 rectangular or irregular morphologies. They were largely from natural dust and industrial combustion (Moffet et
152 al., 2008; Silva et al., 2000; Ye et al., 2018). The presence of spherical metal particles indicated that they
153 experienced melting at high temperature (Giere et al., 2003; Giere et al., 2006). Fly ash particles mainly contained
154 Si, Al and O. Fly ash particles tended to be spherical in morphology and they were generally produced from the
155 process of coal combustion (Chen et al., 2012; Henry and Knapp, 1980).

156 2.4 SPAMS analysis of RES and INT

157 A SPAMS (Hexin Analytical Instrument Co., Ltd., Guangzhou, China) was used to analyze the chemical
158 composition and size distribution of individual particles in real-time. Particles entering the SPAMS were first
159 focused into a beam of particles through an aerodynamic lens, and then their flight velocities were determined by
160 two continuous diode Nd:YAG laser beams (532 nm). Polystyrene spheres of known size were used as a standard
161 substance to calibrate the vacuum dynamic size (d_{va}) of particles. Next, the pulsed laser (266 nm) was precisely
162 triggered to ionize the target particle according to the intrinsic velocity of each particle, and the positive and
163 negative ions are separated and analyzed using a dual polarity time-of-flight mass analyzer. Finally, we obtained
164 the information of individual particles, including d_{va} and the positive and negative ion mass spectra. The relative
165 peak area of characteristic peaks for each species in the mass spectra is generally applied to indicate its relative
166 abundance in the particle (Bhave et al., 2002; Gross et al., 2000). However, it is still challenging to provide
167 quantitative information on chemical compositions, mainly attributed to the different ionization efficiency and the
168 complex matrix effects for various types of particles. A detailed description of particle analysis methods and
169 particle type characteristics can be found in the supporting information.

170 2.5 Calculating morphology parameters of soot

171 The fractal dimension of soot is characterized in the following statistical scaling law (Brasil et al., 1999; Köylü
172 et al., 1995):

$$173 \quad N = k_g \left(\frac{2R_g}{d_p} \right)^{D_f}$$

174 where N is the number of monomers within a certain soot aggregate, k_g is the fractal pre-factor, R_g is the radius of
175 gyration, d_p is the diameter of the monomer, and D_f is the mass fractal dimension. R_g can be obtained by using a
176 simple relationship between R_g and L_{max} , the maximum length of the soot aggregate (Brasil et al., 1999):

$$177 \quad L_{max}/2R_g = 1.50 \pm 0.05$$

178 And, the number of monomers, N , can be calculated by a power-law correlation of projected area of monomer
179 and aggregate:

$$180 \quad N = k_a \left(\frac{A_a}{A_p} \right)^\alpha$$

181 where k_a is a constant, A_a and A_p are the projected area of aggregate and monomer, respectively, and α is an
182 empirical projected area exponent. The value of k_a and α depends on the degree of monomer overlap (δ) in the
183 aggregate (Oh and Sorensen, 1997), and δ can be determined by:

$$184 \quad \delta = \frac{2a}{l}$$

185 where a is monomer radius, and l is the center distance of adjacent monomers. The values of parameters including
186 a , l , A_a , A_p , L_{max} , and d_p can be obtained by analyzing TEM images. Then D_f can be calculated by the above four
187 formulas.

188 3 Results and Discussion

189 3.1 Particle type and mixing state of RES and INT

190 According to mixing state, RES and INT were divided into the following eight types (Figure 1): S-rich, S-OM,
191 refractory (soot/mineral dust/metal/fly ash), aged soot (S/OM-soot), aged mineral dust (S/OM-mineral dust), aged
192 metal (S/OM-metal), aged fly ash (S/OM-fly ash), and aged refractory mixture (S/OM-soot/mineral dust/metal/fly
193 ash). S-rich or OM, generally considered to be aged since they are mainly secondarily produced in the atmosphere,
194 are internally mixed with refractory materials (soot/mineral dust/metal/fly ash) (Canagaratna et al., 2007; Huang
195 et al., 2012; Jiang et al., 2019). Such internally mixed S/OM-refractory particles are named as aged refractory
196 particles herein. Aged particle types containing two or more refractory components are named as “aged refractory
197 mixture”. It is worth noting that refractory are refractory particles without S-rich and OM.

198 Figure 2 shows the number fraction of different particle types in the RES and INT during cloud events #2 and
199 #3. S-rich, S-OM, aged soot, and aged refractory mixture particles are dominant particle types. The most abundant
200 particles in the RES are aged refractory mixture (23%), followed by S-OM (22%), aged soot (20%), S-rich (16%),
201 aged metal (9%), aged fly ash (5%), aged mineral dust (4%), and refractory (1%). Differently, INT is
202 predominated by S-rich (29%), aged soot (27%), S-OM (15%), aged refractory mixture (10%), and the lesser
203 percentage of aged fly ash (8%), refractory (5%), aged mineral dust (4%), and aged metal (2%) were also observed.
204 Among three cloud events, the RES are dominated by S-OM in cloud event #1 and #2 and aged refractory mixture
205 particles in cloud event #3 (Figure 3). It is also shown that the RES and INT analyzed by TEM/EDS can represent
206 their compositions throughout cloud events #2 and #3, since such compositions were relatively stable throughout
207 these periods (Figure S3).

208 The different air masses are expected to affect the distribution of particle types. The distribution of several types
209 of particles in the RES was observed to be divergent in different cloud events, corresponding to different air
210 masses, as shown in Figure 3 and Figure 4. The number fraction of OM-containing particles was the highest (81%)
211 in cloud event #2, which might be partly attributed to the higher concentration of O_3 during cloud event #2 (Table
212 S1). And the samples of cloud event #2 were collected at noon. Higher solar radiation during the sampling time
213 might also promote heterogeneous photochemical oxidation reactions during the cloud process and increased the
214 generation of OM within cloud droplets (Xu et al., 2017). Aged metal particles accounted for a similar percentage

215 (7-12%) for three cloud events. The proportion of aged **mineral dust** during cloud event #1 (14%) was nearly four
216 times those in the other two cloud events. Aged fly ash particles had the highest proportion (10%) in cloud event
217 #3 compared with the other two cloud events, most probably influenced by the different air masses (Figure 4).
218 Aged **mineral dust** particles of cloud event #1 may be influenced by the long-distance transportation of dust from
219 Southeast Asia (Salam et al., 2003). Clearly, aged fly ash particles of cloud event #3 are associated with the air
220 masses from the PRD region with a dense distribution of industrial facilities there (Cao et al., 2006).

221 **3.2 The morphology and mixing structure of carbonaceous particles**

222 OM-containing particles, including all of S-OM particles, part of aged refractory (S-OM/OM-refractory) and
223 **aged refractory mixture** (S-OM/OM-soot/**mineral dust**/metal/fly ash) particles, accounted for 60% of RES and 33%
224 of INT during cloud events #2 and #3. According to the mixing structures between OM and other materials (Figure
225 5), OM-containing particles are classified into the following five categories: thinly coated (Figure 5b), core-shell
226 (Figure 5c), embedded (Figure 5d), attached (Figure 5e), and homogenous-like (Figure 5f) structures (Li et al.,
227 2016). A particle is classified as a thinly coated structure when wrapped with a thin layer of OM. The thickness
228 of the OM layer of thinly coated particles ranges from 12 to 150 nm. Generally, the shapes of OM-containing
229 particles with the thinly coated structure are elliptical or irregular. The difference between the core-shell structure
230 and thinly coated structure is the relative thickness of OM: Core-shell structure possessed thicker organics than
231 thinly coated structure. The thickness of the shell varies from 86 to 2110 nm, and the ratio of the projected area
232 of the shell to particle ranges from 0.20 to 0.97. Moreover, OM-containing particles with a core-shell structure
233 are round. Embedded or attached structure for the OM-containing particles refers to the relative distribution of
234 OM, i.e., embedded in or attached to other materials (e.g., sulfate). Well mixed OM-containing particles with no
235 identifiable boundary between organic and non-organic matter were identified with a homogenous-like structure.

236 The first most abundant particles are thinly coated geometry, comprising 53% of RES and 59% of INT during
237 cloud event #2 and #3. The second are core-shell particles for RES and attached particles for INT. The percentage
238 of core-shell particles in the RES is almost 2.5 times that in the INT (27% vs. 12%). Embedded and homogenous-
239 like particles account for minor proportions (< 4%) for both RES and INT.

240 Soot-containing particles, including all of the aged soot particles (S/OM-soot) and part of refractory
241 (soot/**mineral dust**/metal/fly ash) and **aged refractory mixture** particles (S/OM-soot/**mineral dust**/metal/fly ash),
242 account for 36% of RES and 39% of INT during cloud event #2 and #3, respectively. The fraction is consistent
243 with the range of those (< 30% – ~60%) observed at the same site by SPAMS (Zhang et al., 2017a). Most of the
244 soot particles are observed to distribute around the periphery of particles (Figure S4).

245 **3.3 In-cloud formation of OM**

246 It can be seen from Figure 2 that a shift of dominant particle types from S-rich (29%) and aged soot (27%) in
247 the INT to the **aged refractory mixture** (23%) and S-OM (22%) in the RES. In particular, the fraction of OM-
248 containing particles increases from 33% in the INT to 60% in the RES. It is unlikely due to the favorable activation
249 of S-OM or **aged refractory mixture**, since mixing with OM generally lower the hygroscopicity of inorganic-
250 dominant particles (e.g., S-rich) (Brooks et al., 2004; Pierce et al., 2012). OM coating at the same site has been

251 shown to inhibit the CCN activation of soot-containing particles (Zhang et al., 2017a). Instead, it is most probably
252 attributed to the in-cloud formation of OM on the surface of some S-rich particles, shifting the dominant particle
253 type from S-rich to S-OM particles. It can be supported by the relatively larger median size of S-OM particles
254 (0.76 μm) than S-rich particles (0.56 μm) (Figure S5), since in-cloud formation of OM is expected to enlarge the
255 original S-rich particles (Pierce et al., 2012).

256 In addition, the fraction of OM-containing particles with core-shell mixing structure in the RES is almost 2.5
257 times that in the INT (Figure 5a). Such a mixing structure is similar to those observed in the Arctic, background,
258 or rural atmosphere (Hiranuma et al., 2013; Li et al., 2016; Yu et al., 2019), but is different from other findings in
259 polluted areas, where OM-containing particles mainly existed in homogenous-like and thinly coated structures
260 (Li et al., 2016). It is also consistent with several laboratory simulations demonstrating that reactive uptake of
261 volatile organic compounds (VOCs) on inorganic sulfate and heterogeneous and multiphase reactions between
262 these species would lead to a core-shell morphology (e.g., Riva et al., 2019; Zhang et al., 2018a; Zhang et al.,
263 2019). Recently, Gorkowski et al. (2020) came up with a particle morphology prediction framework developed
264 for mixtures of organic aerosol based on the measurements from aerosol optical tweezers experiments and
265 literature data, and they hypothesized the core-shell morphology dominated by secondary organic aerosols (SOA)
266 in the shell phase.

267 Moreover, we estimated the O/C ratio of coating and shell within OM-containing particles. It should be noted
268 that the O/C ratio of organic coating and shell is underestimated herein due to the copper grid evenly covered by
269 carbon film. Moreover, while some loss of volatile organic compounds during the TEM/EDS analysis may affect
270 the O/C of particles, the relatively higher O/C ratio for the RES is still affirmative. Droplets are expected to
271 dissolve more volatile organic compounds (Chakraborty et al., 2016) with higher O/C. The release of these
272 compounds during droplet evaporation would result in underestimating of O/C in the RES. We found that the
273 average value of the O/C ratio of RES is higher than that of INT, and the average value of the O/C ratio of RES
274 with a core-shell structure is 0.23, which is two times that with a thinly coated structure (0.11) (Table 2), indicating
275 that these RES with core-shell particles are more oxidized. At the same site, we have previously observed
276 enhanced aqueous SOA products, such as oxalate in the cloud (Zhang et al., 2017b). The higher O/C ratio of core-
277 shell particles is also consistent with current studies reporting more oxidized organic species in cloud/fog residues
278 (Brege et al., 2018; Chakraborty et al., 2016; Zhang et al., 2017b). With high levels of VOCs at the sampling site
279 (Lv et al., 2019), the prevalent formation of aqueous SOA through the uptake of VOCs in cloud droplets would
280 be expected (Kim et al., 2019; Liu et al., 2018a). The contribution from photochemical processes may also be
281 reflected by the association of the highest fraction (81%) of OM-containing particles with a higher concentration
282 of O_3 during cloud event #2 (Table S1). Consistently, the relative peak area of m/z 43 $\text{C}_2\text{H}_3\text{O}^+$ in the RES is higher
283 than that in the INT during cloud event #2 (Figure S7), indicative of the favorable formation of oxidized organic
284 compounds (Qin et al., 2012; Zhang et al., 2017b).

285 However, one may expect that such a core-shell mixing structure in the RES can also be explained by the
286 primary activation of S-OM particles with larger sizes. Unfortunately, no sample before the cloud events is
287 available for TEM/EDS measurements. However, with evidence from the collocated SPAMS, we show that this
288 is not convincing. As shown in Table S2, the ratios of relative peak area between organics and sulfate are similar

289 between the INT and particles before cloud event, whereas they are higher in the RES. This is corresponding to
290 the production of oxidized organics during in-cloud processes (Zhang et al., 2017b), consistent with the TEM/EDS
291 results.

292 3.4 The D_f of soot in the RES and INT

293 Figure 6 shows the D_f of soot within RES and INT of cloud event #2 and #3. The result shows that the D_f of
294 soot is smaller in the RES (1.82 ± 0.12) than in the INT (2.11 ± 0.09), which means that soot is more branched in
295 the RES. It is noted that 62.5% of all soot-containing particles with clear boundaries are included in the D_f
296 calculation since thick coating around soot might make the boundary of monomers not clear enough (Bhandari et
297 al., 2019). The obtained D_f are close to those ($1.83 - 2.16$) reported at a background site (Wang et al., 2017). The
298 D_f of soot in the RES and INT likely represents partly coated soot (1.82 ± 0.05) (Yuan et al., 2019) and embedded
299 soot (2.16 ± 0.05) (Wang et al., 2017), respectively. In addition to emission sources and coating processes, high
300 relative humidity (RH) during nighttime is a critical factor to increase the compactness of soot (Yuan et al., 2019).

301 While some previous studies demonstrated that soot aggregates tend to be more compact (with larger D_f) after
302 aging or cloud processing (Adachi and Buseck, 2013; Moffet and Prather, 2009; Wu et al., 2018), our results
303 suggest that in-cloud processes may result in more branched soot, as shown in Figure 6. Considering that D_f is
304 controlled mainly by emission sources, combustion conditions, and aging processes (Adachi et al., 2007), we
305 propose three possible explanations for the lower D_f of soot in the RES than that in the INT. The first and the most
306 likely reason is that some of the soot aggregates are immediately encapsulated by non-volatile materials (such as
307 organic matter) after emission by combustion sources. These coatings fill the spaces between the branches of soot
308 aggregates, which inhibits the relatively large deformation and reconfiguration of the soot aggregates during
309 transport and activation into cloud droplets (Zhang et al., 2018b). Differently, soot aggregates may shrink easily
310 and become more compact during the long-distance transport if the soot aggregates are emitted without non-
311 volatile coatings (Adachi and Buseck, 2013). We show that soot aggregates have higher D_f and lower average
312 ECD in the INT (247 nm) than in the RES (266 nm), which means that larger, less dense soot particles are easier
313 to act as CCN. This is consistent with a study reporting that small particles are more compact than large particles
314 (Adachi et al., 2014). The second is that water-soluble substances within aerosols will be miscible after activating
315 to cloud droplets (Gorkowski et al., 2020). The coating materials of soot may be released, which makes soot more
316 branched in the droplets and the following-up droplet evaporation. The third possible explanation is that different
317 combustion materials and combustion conditions produce soot-containing particles with different mixing states
318 and morphology (China et al., 2014; Khalizov et al., 2013; Liu et al., 2017; Zhang et al., 2018b).

319 This result contrasts with the current study reporting that soot sampled after cloud droplet evaporating is more
320 compact than freshly emitted and interstitial soot (Bhandari et al., 2019). Our observations at the background site
321 show that the majority of soot aggregates in both RES and INT (~80%) are located in off-center positions (Figure
322 S4), having less compact shapes even after being coated. This is quite different from the core-shell model currently
323 used in the climate models (Bond and Bergstrom, 2006; Wu et al., 2018). Through theoretical calculation, Adachi
324 et al. (2010) suggested that absorption cross-sections could be reduced by 20-30% with off-center positions of
325 soot relative to center positions. This means that the models based on core-shell assumption may overestimate the
326 absorption of soot-containing particles after cloud processing.

327 4 Conclusion and atmospheric implications

328 The result highlights the different morphology and mixing structures of activated and interstitial particles,
329 which may imply the substantial role of in-cloud aqueous processes in reshaping the activated particles. While Yu
330 et al. (2019) considered organic coatings on sulfate in the Arctic as a result of the increase of SOA following
331 particle aging and growth during transport, our data further imply a specific role of in-cloud processes in the
332 coating on sulfate. The prevalence of OM shelled particles after cloud processing also supports a current laboratory
333 observation depicting that rapid film formation and fast heterogeneous oxidation can provide an efficient way of
334 converting water-insoluble organic films into more water-soluble components in aerosols or cloud droplets
335 (Aumann and Tabazadeh, 2008).

336 Gorkowski et al. (2020) suggested that mixing structures of OM-containing particles **is** related to the oxidation
337 degree of OM. We also show that OM shells formed **in cloud droplets** have a higher degree of oxidation. Such a
338 chemical and morphological modification of aerosol particles may influence species diffusivities from the interior
339 to the surface region of the shell and gas-particle partitioning between the shell and gas (Liu et al., 2016; Shiraiwa
340 et al., 2013). Such a reshaping may also have an influence on aerosol hygroscopicity. Extrapolating the linear
341 relationship between the O/C ratio and the hygroscopicity parameter (κ_{org}) indicates that $\kappa_{\text{org-shell}}$ is about 1.4 times
342 $\kappa_{\text{org-coating}}$ (Jimenez et al., 2009; Lambe et al., 2011). In addition, the formation of the organic film could result in
343 a change of surface tension and thus affect the critical supersaturation required for particle activation (Ovadnevaite
344 et al., 2017). The heterogeneous ice nucleation potential may be suppressed for mineral particles when coated by
345 OM (Möhler et al., 2008). Given the critical contribution of in-cloud aqueous SOA, several mixing structures of
346 OM-containing aerosols upon in-cloud processes may have substantial implications in modeling the direct and
347 indirect radiative forcing of aerosols (Scott et al., 2014; Zhu et al., 2017).

348

349 *Data availability.* Data are available on request from Guohua Zhang (zhanggh@gig.ac.cn) and Xinhui Bi
350 (bixh@gig.ac.cn).

351 *Author contribution.* GHZ and XHB designed the research (with input from XMW and GYS). YZF, GHZ, and
352 XHB analyzed the data, and wrote the manuscript. YZF, XFL, YXY, FJ, and QHL conducted sampling work
353 under the guidance of GHZ, XHB and XMW. LL, DHC and JO had an active role in supporting the sampling
354 work. YZF performed the laboratory analysis of individual particles by TEM/EDS, with support from YPY and
355 JXZ. All authors contributed to the discussions of the results and refinement of the manuscript.

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

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646 **Table 1. The information of cloud events and samples, including starting and ending time of each cloud event, the number and type of analyzed particles, the mean value**
 647 **of visibility and number concentration of RES or INT during sampling time.**
 648

Cloud event	Starting Time*	Ending Time*	Particles	Type	Visibility/m	Number Concentration/cm ⁻³
Cloud #1	2017/5/20 18:19	2017/5/21 8:34	190	RES	66	195
Cloud #2	2017/5/23 20:35	2017/5/25 6:35	161	INT	50	99
			162	RES	88	299
Cloud #3	2017/6/8 18:30	2017/6/10 17:30	132	INT	44	996
			135	RES	33	111

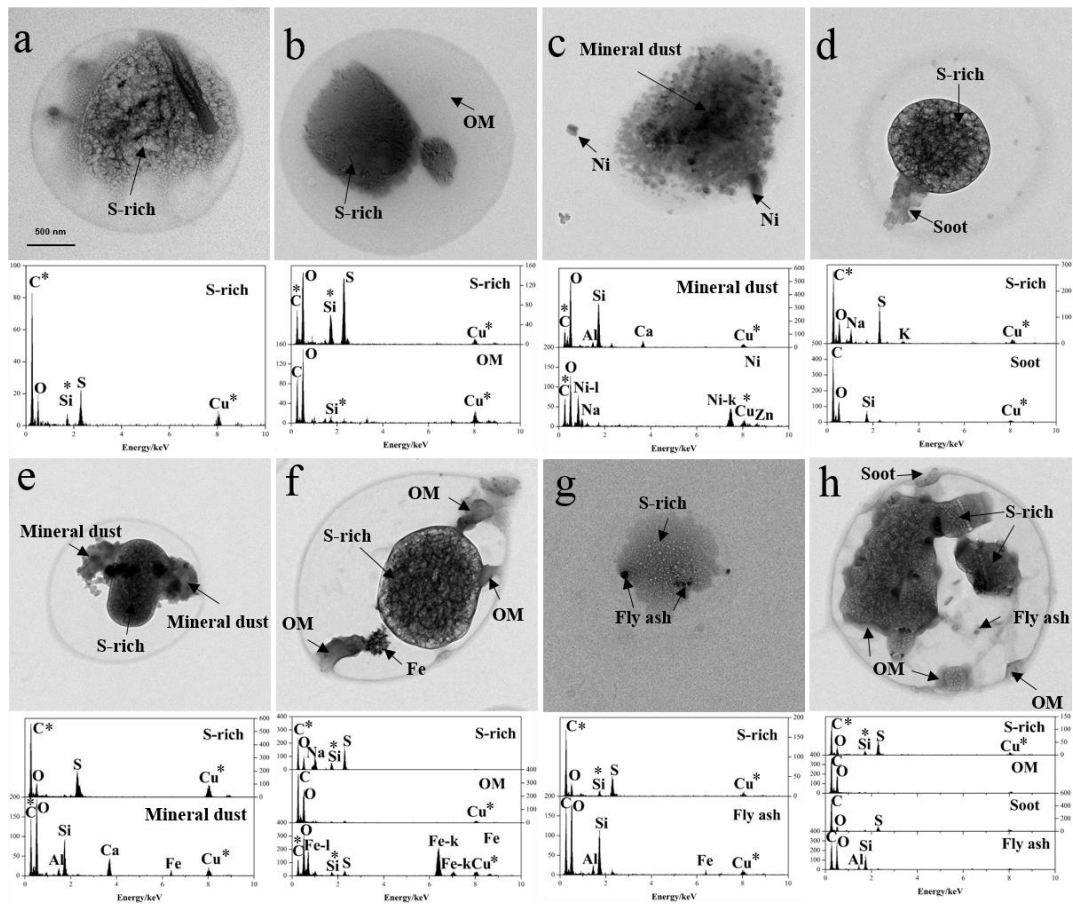
649 * Local time, i.e., Chinese Standard Time, UTC+8.

650 **Table 2. The average value of O/C ratio of OM-containing particles with thinly coated and core-shell mixing structures.**

651

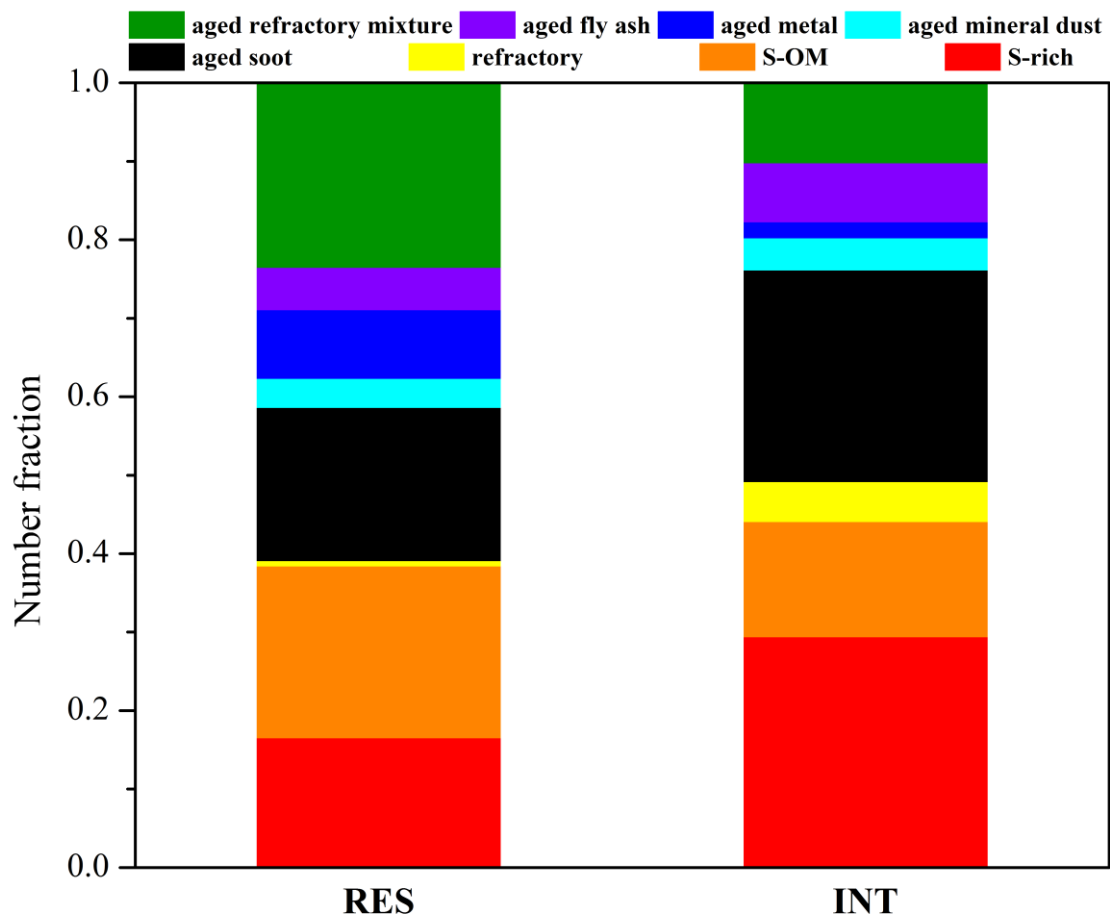
Type	thinly coated	core-shell
RES	0.11	0.23
INT	0.08	0.06

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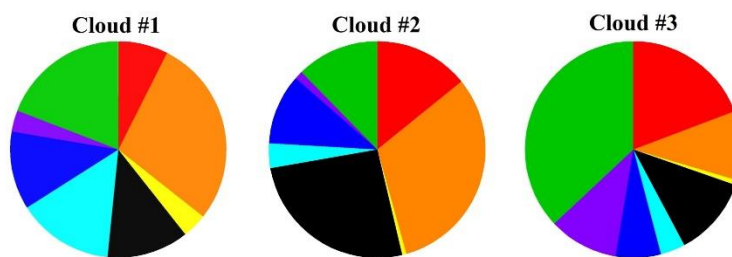
654 Figure 1. TEM images and EDS spectra of individual RES and INT particles with different particle types: (a)
 655 S-rich; (b) S-OM; (c) refractory; (d) aged soot; (e) aged mineral dust; (f) aged metal; (g) aged fly ash; (h) aged
 656 refractory mixture. Asterisk (*) represents the background element.



657

658 Figure 2. Number fractions of different particle types in the RES and INT of cloud event #2 and #3 measured

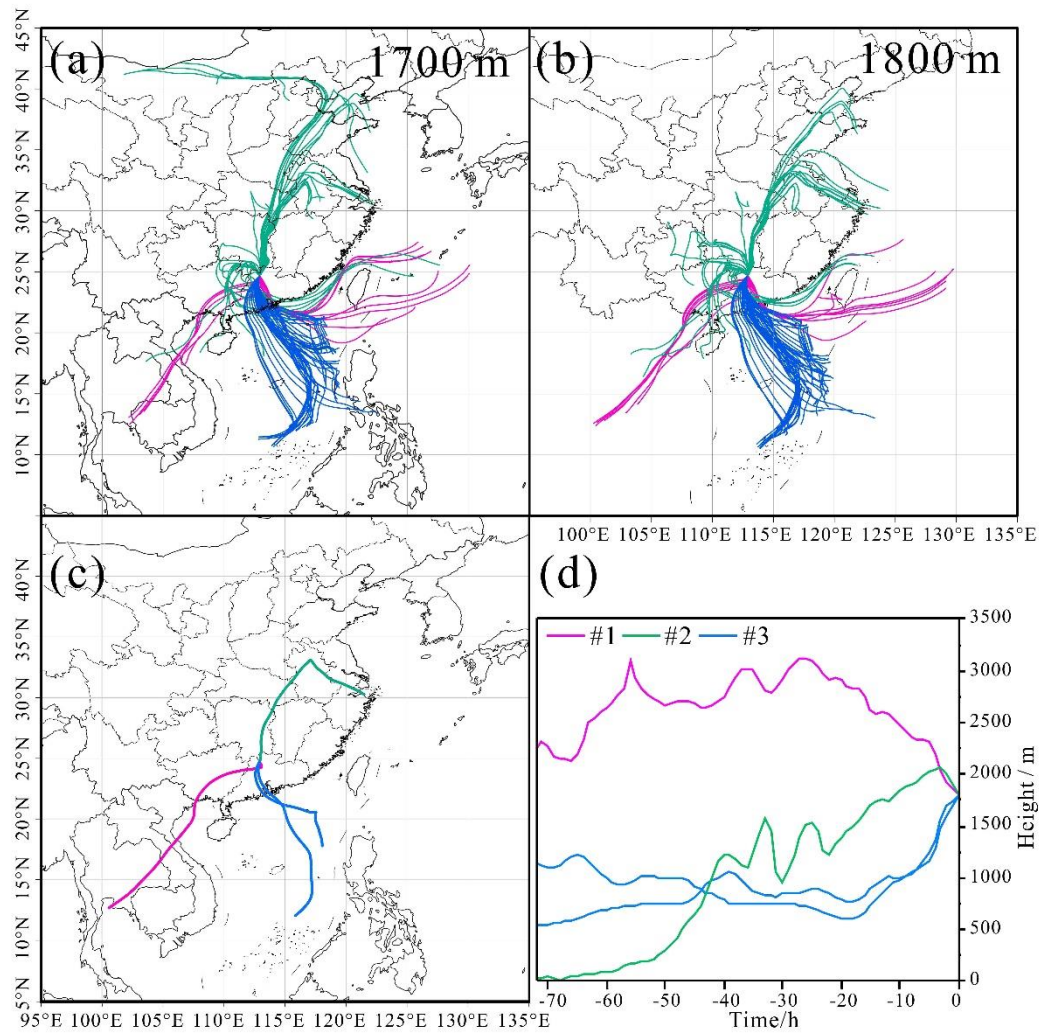
659 by TEM/EDS.



660 ■ S-rich ■ S-OM ■ refractory ■ aged soot ■ aged mineral dust ■ aged metal ■ aged fly ash ■ aged refractory mixture

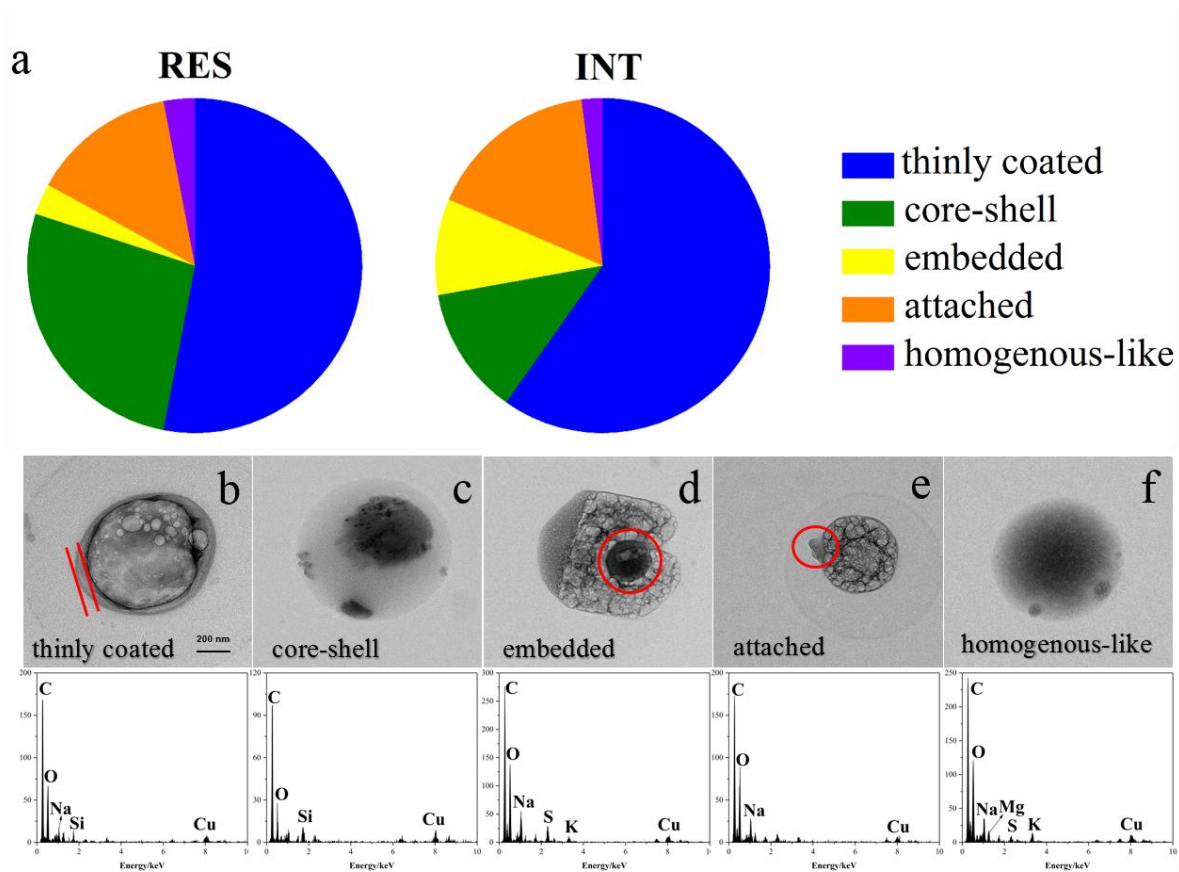
661 **Figure 3. Number fraction of different particle types in the RES during three cloud events measured by**

662 **TEM/EDS.**



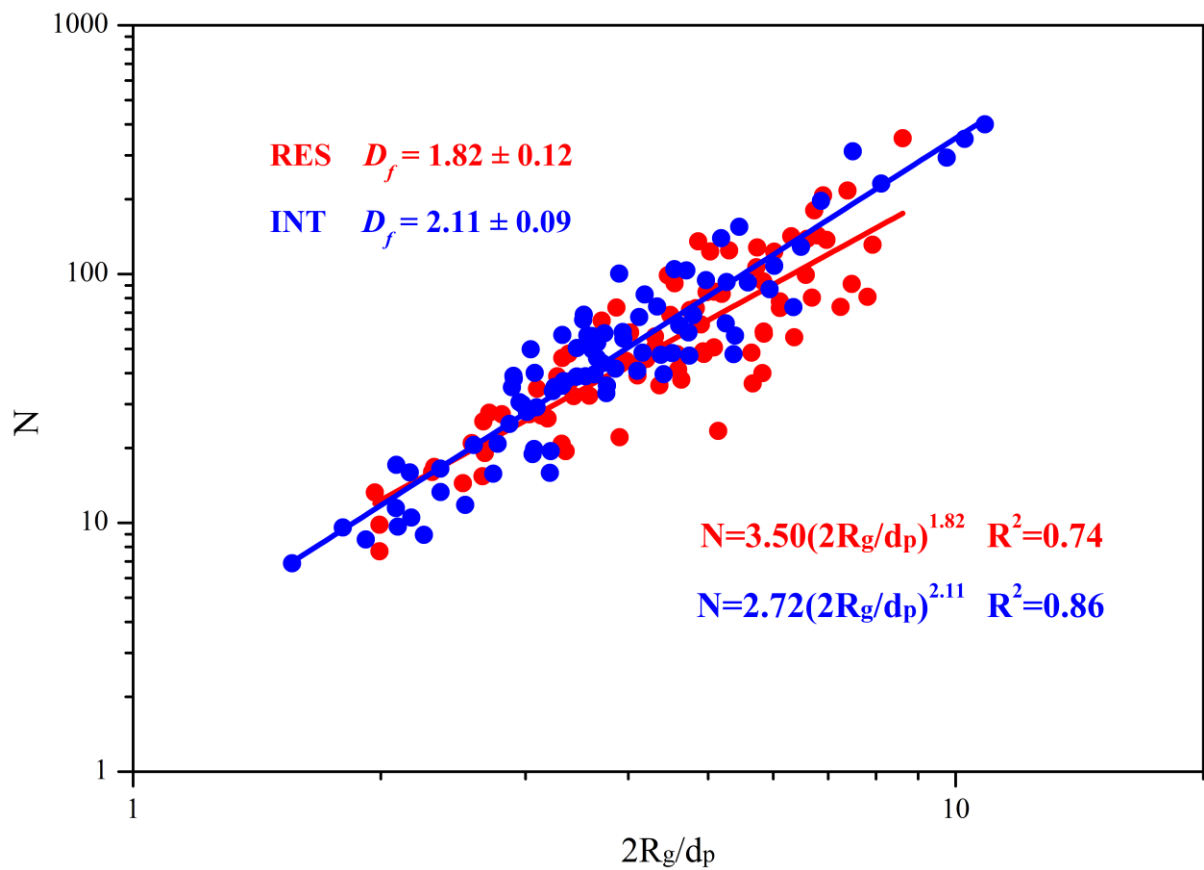
663

664 **Figure 4.** HYSPLIT back trajectories (72 h) for air masses arriving at our sampling site at the height of 1700
 665 m (a) and 1800 m (b) hourly during the three cloud events. The HYSPLIT back trajectories at the height of
 666 1800 m during sampling periods (c) and heights (above sea level) of the air masses during transport (d). The
 667 horizontal axis represents several time points (0-72 h) before the time point input into the HYSPLIT model.



668

669 **Figure 5.** Number fractions of OM-containing particles with different mixing structures in the RES and INT (a) and
 670 typical TEM images and corresponding EDS spectra of OM: thinly coated (b); core-shell (c); embedded
 671 (e); homogenous-like (f) during cloud event #2 and #3.



672
 673
 674

Figure 6. Fractal dimensions of soot in the RES and INT during cloud event #2 and #3.

Supplement of

Impact of in-cloud aqueous processes on the chemical compositions and morphology of individual atmospheric aerosols

Yuzhen Fu et al.

*Correspondence to: Guohua Zhang (zhanggh@gig.ac.cn) and Xinhui Bi (bixh@gig.ac.cn)

1 **1 Air mass backward trajectories and meteorology conditions**

2 The backward trajectory and the height (above sea level) of air masses during sampling were calculated
3 by the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model
4 (<http://ready.arl.noaa.gov>). During three cloud events, the sampling site was greatly influenced by air
5 masses from Southeast Asia, northern China and the South China Sea. Compared with the cloud event
6 #1, the air masses of cloud event #2 and #3 passed through a relatively low path on the way to the
7 sampling site. Thus, the air masses of cloud event #2 and #3 were affected more by the ground
8 anthropogenic emissions. The ambient temperature at the sampling station varied from 12.1 to 18.6 °C
9 during three cloud events. All samples were collected during the stable period of cloud events, when the
10 mass concentration of PM_{2.5} was less than 5 µg m⁻³ and visibility was less than 100 m. The concentrations
11 of PM_{2.5} during cloud event #1 were lower than those during cloud event #2 and #3. Consistently, the
12 mean concentrations of O₃, SO₂ and NO_x were higher in the cloud event #2 and #3 (Table S1).

13 **2 The size distribution of RES and INT**

14 In this study, a PM_{2.5} cyclone inlet and a GCVI (ground-based counterflow virtual impactor) inlet were
15 used to collect INT and RES, which is similar to Cozic et al. (2007). Additionally, the particle size in this
16 study refers to as ECD (equivalent circle diameter) obtained from TEM images, which is larger than ESD
17 (equivalent spherical diameter). Liu et al. (2018) showed that the ECD of individual dry particles on the
18 substrate is 0.4952 times that of the ESD.

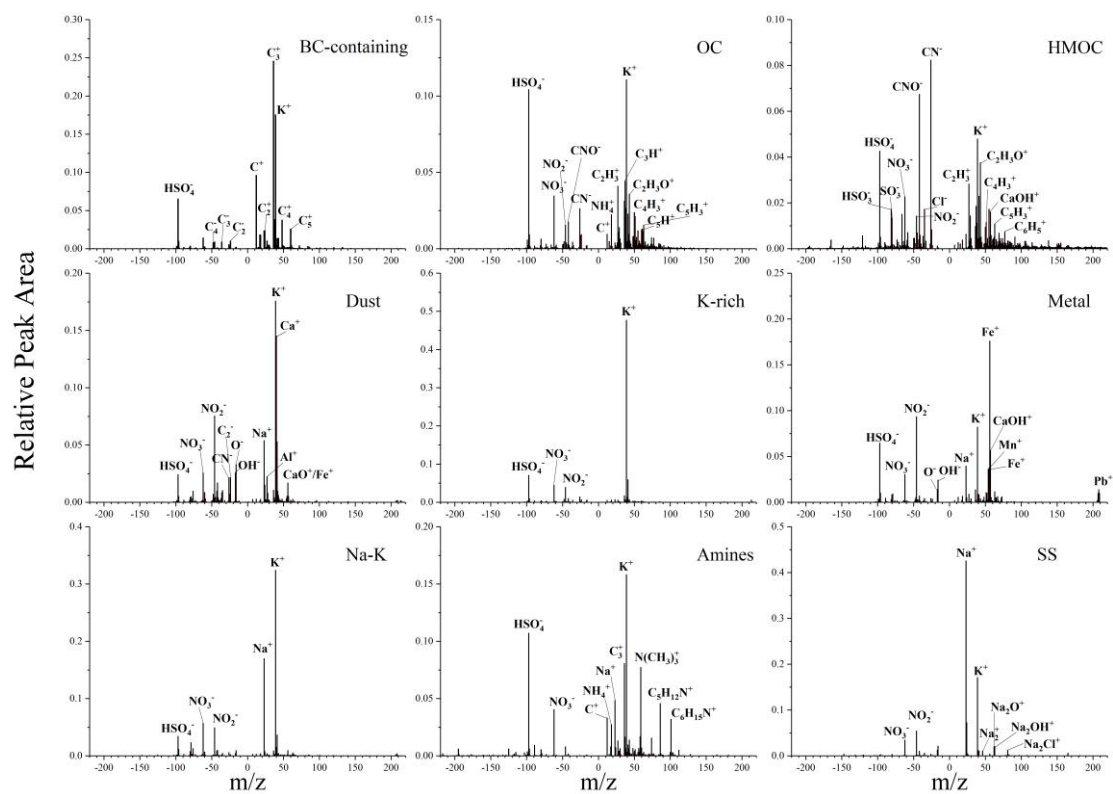
19 The size distribution data shows a higher median diameter of RES (1.20 µm) than INT (0.63 µm)
20 (Figure S2), which are higher than those (0.8 and 0.45 µm, respectively) at Mount Tai in northern China
21 (Li et al., 2011). This could be because Mount Tai is located in an industrial area, whereas our site
22 represents a background region mainly influenced by long-range transport. Additionally, the formation
23 of secondary compounds during cloud events increases the size of RES (Zhang et al., 2017).

24 The size distribution of different particle types revealed that S-rich and aged soot particles were
25 predominant in smaller size segments, and aged mixture particles in larger size segments (Figure S8).
26 Likewise, the size-resolved number fractions of different particle types from the results of the SPAMS
27 also showed that the BC-containing particles were mainly distributed between 0.1 and 1.3 µm,

28 representing ~80% of the submicron RES and ~73% of the submicron INT population, respectively
29 (Figure S9).

30 3 Identification of several types of particles within RES and INT measured by SPAMS

31 The information on particle sizes and mass spectra is imported into the Matlab for subsequent analysis
32 using the FATES toolkit (Sultana et al., 2017). A total of 117,436 particles from the SPAMS were
33 analyzed. All the particles with bipolar mass spectra and the size range of d_{va} 0.1–1.9 μm were classified
34 several clusters by an adaptive resonance theory neural network (ART-2a) with a learning rate of 0.05, a
35 vigilance factor of 0.8 and 20 iterations, and merged similar clusters manually. Ten characteristic particle
36 types (Figure S1) were obtained, including BC (black carbon)-containing, OC (organic carbon), HMOC
37 (highly molecular organic carbon), Dust, K-rich, Metal, Na-K, Amines, SS (sea salt) and Others. BC-
38 containing particles are characterized by elemental carbon cluster ions (m/z 12C^\pm , 24C_2^\pm , 36C_3^\pm , 48C_4^\pm , ...)
39 (Arndt et al., 2017). OC particles mainly contain fragment ions of organics (m/z $27\text{C}_2\text{H}_3^+$, $37\text{C}_3\text{H}^+$,
40 $43\text{C}_2\text{H}_3\text{O}^+$, -26CN^- , ...) (Denkenberger et al., 2007; Qin et al., 2012). The mass spectra of HMOC
41 particles show the presence of peaks of OC particles and some other organic peaks (such as m/z $77\text{C}_6\text{H}_5^+$,
42 $91\text{C}_7\text{H}_7^+$). Furthermore, HMOC particles are distinguished from OC particles by marked ion fragments
43 detected in range of $m/z > 100$ (Qin and Prather, 2006). Dust particles present significant ions at m/z
44 27Al^+ , 40Ca^+ and $56\text{CaO}^+/\text{Fe}^+$ (Silva et al., 2000). K-rich particles are identified according to the strong
45 signal at m/z 39K^+ only in positive mass spectra. Metal particles show the presence of metal ion peaks
46 (such as Fe^+ (m/z 54 and 56), Mn^+ (m/z 55), Pb^+ (m/z 206, 207 and 208)) in positive mass spectra. Na-K
47 particles are characterized by peaks at m/z 23Na^+ , 39K^+ , and less intense peaks at m/z -46NO_2^- , -62NO_3^- ,
48 -97HSO_4^- . The mass spectra of amines particles contain ions signals at m/z $59\text{N}(\text{CH}_3)_3^+$, $86\text{C}_3\text{H}_{12}\text{N}^+$,
49 $101\text{C}_6\text{H}_{15}\text{N}^+$ (Angelino et al., 2001; Pratt et al., 2009). SS particles are mainly composed of ions peaks
50 at m/z 23Na^+ , 46Na_2^+ , $62\text{Na}_2\text{O}^+$, $63\text{Na}_2\text{OH}^+$ and $81\text{Na}_2\text{Cl}^+$ (Gaston et al., 2011). Most particles are
51 observed to be internally mixed with sulfate and nitrate (m/z -46, -62, -97). Particles with inconspicuous
52 mass spectrum characteristics are named as others. Specific classification criteria were described in detail
53 elsewhere (Zhang et al., 2015).



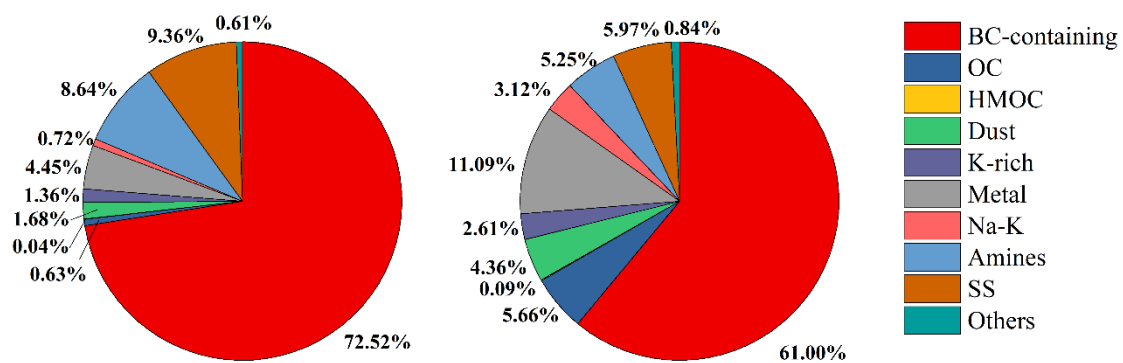
54

55 Figure S1. Average positive and negative mass spectra of the main particle types (i.e., BC-containing, OC,

56 HMOC, Dust, K-rich, Metal, Na-K, Amines, SS) measured by SPAMS.

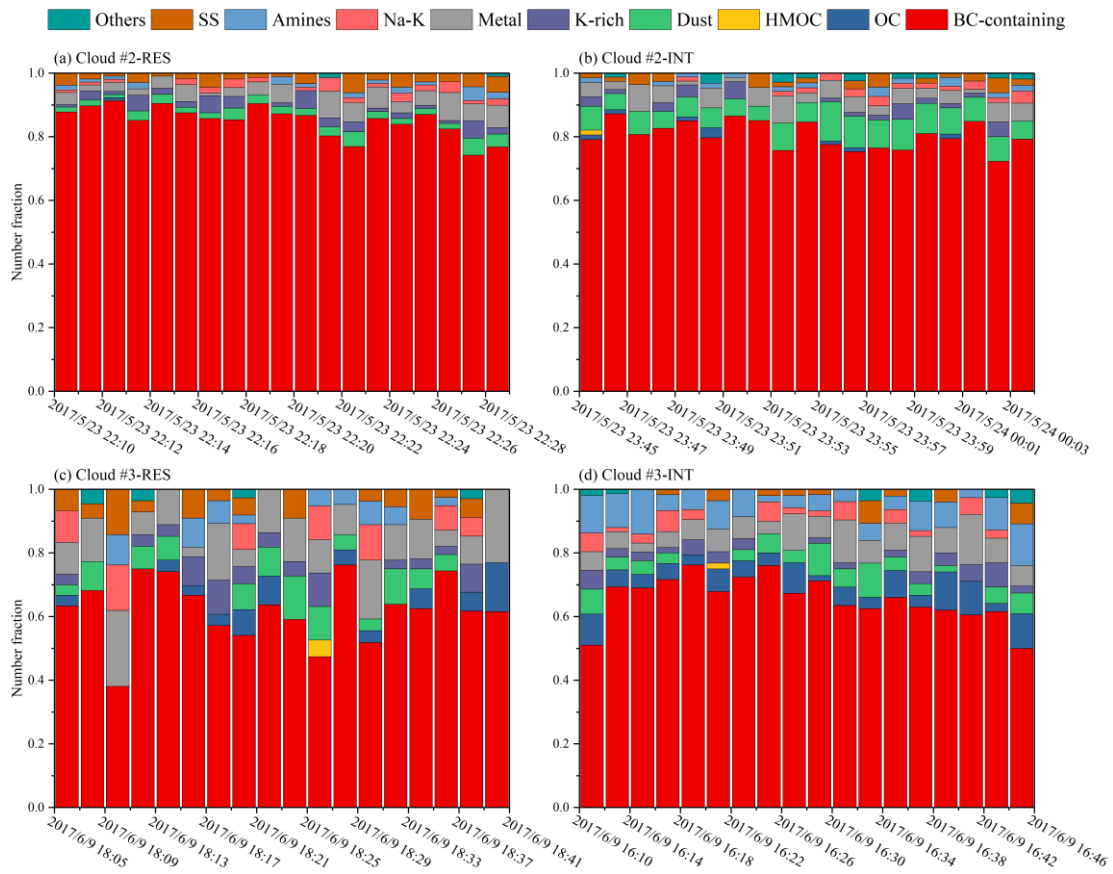
(a) Cloud #2-RES

(b) Cloud #3-RES



57

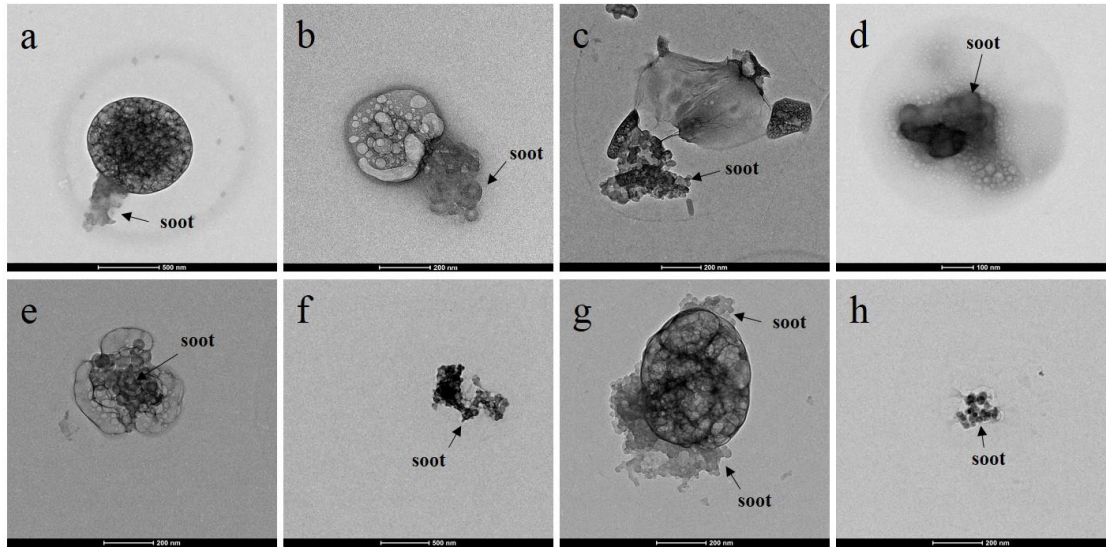
58 Figure S2. The chemical composition of RES measured by the SPAMS during cloud event #2 and #3.



59

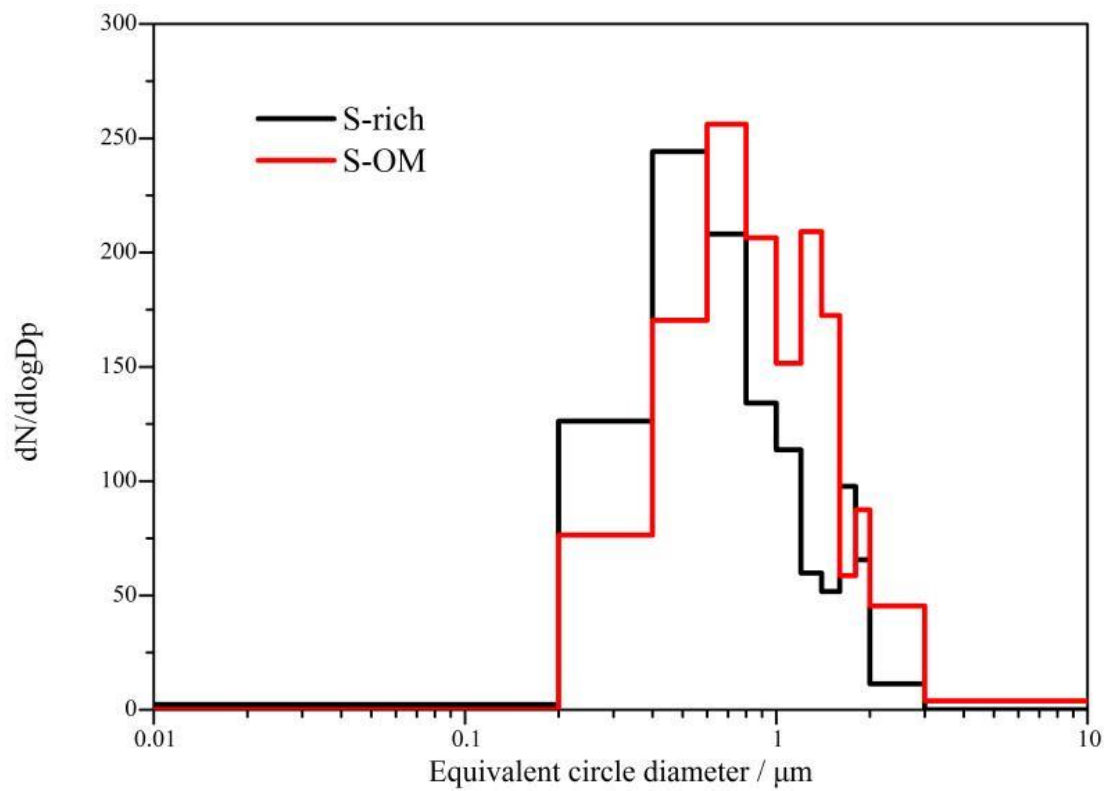
60 **Figure S3. Time series of the chemical composition of RES and INT measured by the SPAMS during cloud**

61 **events #2 and #3.**



62

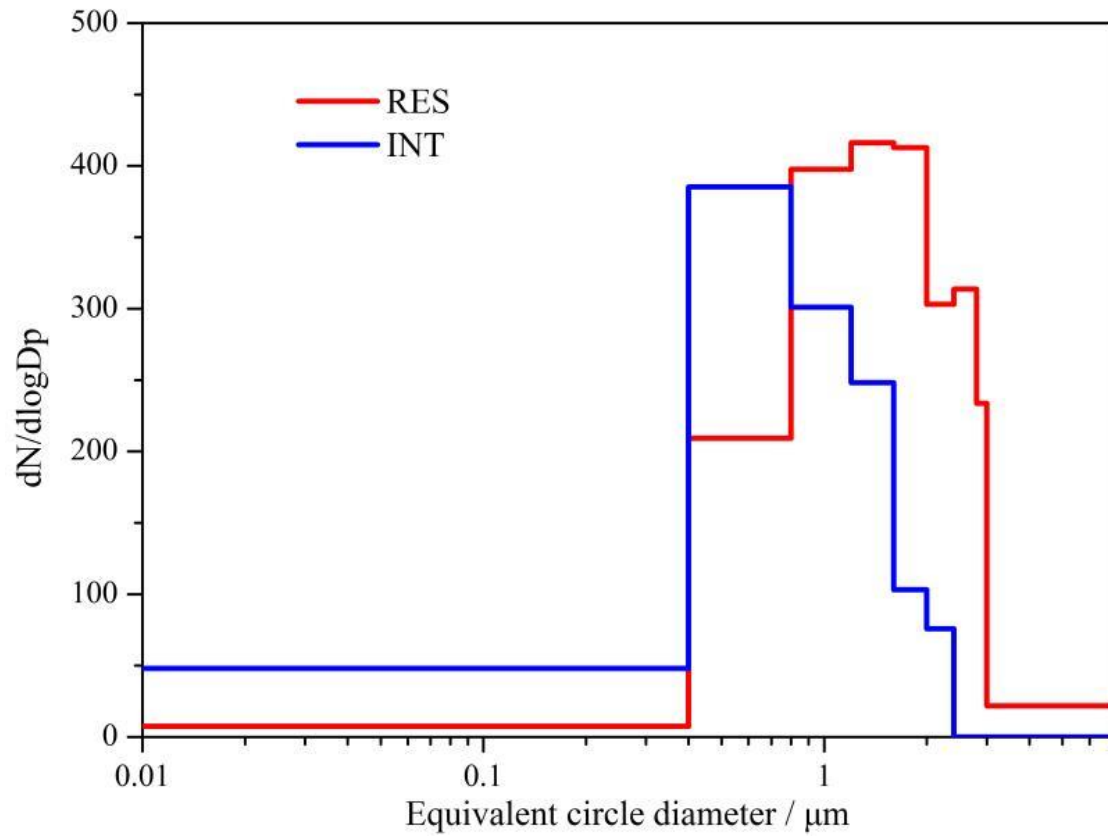
63 **Figure S4. Typical TEM images of soot particles in the RES (a-d) and INT (e-h).**



64

65 **Figure S5. The size distribution of S-rich and S-OM particles. There are few S-rich particles with the size of**

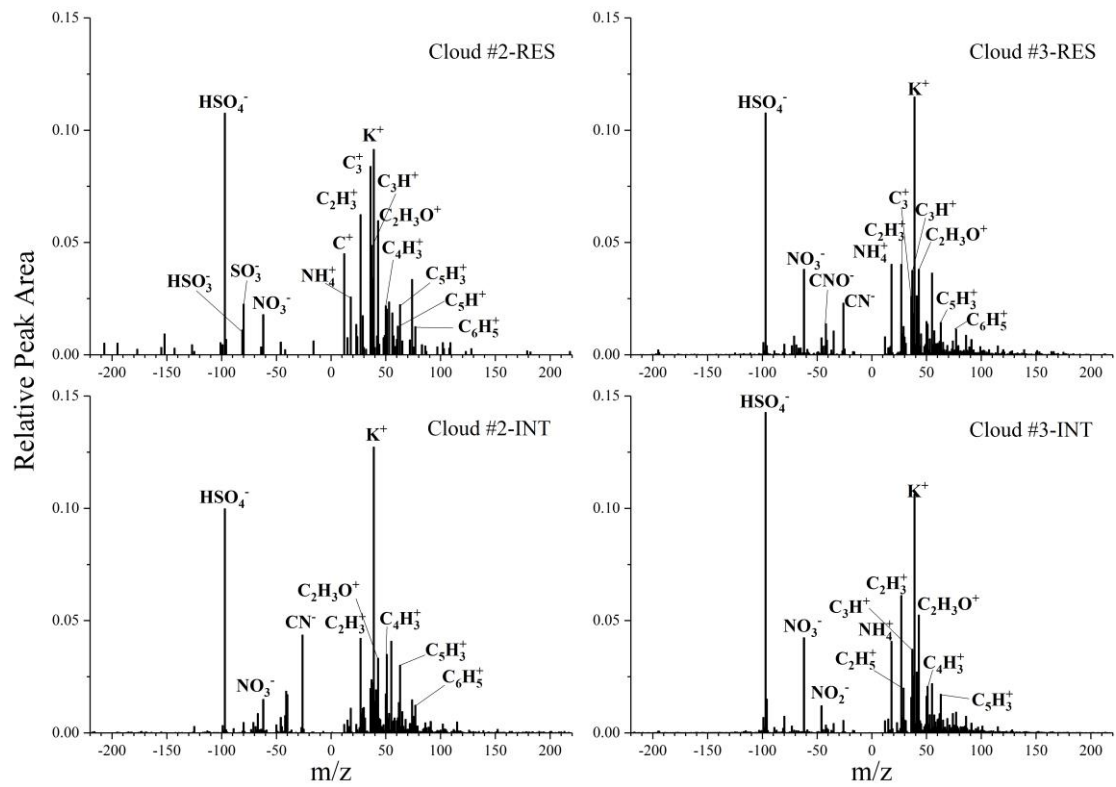
66 **less than 0.2 μm , and the median size are 0.56 μm and 0.76 μm for S-rich and S-OM particles, respectively.**



67

68 **Figure S6. Size distribution of RES and INT during cloud event #2 and #3. There are more INT particles**

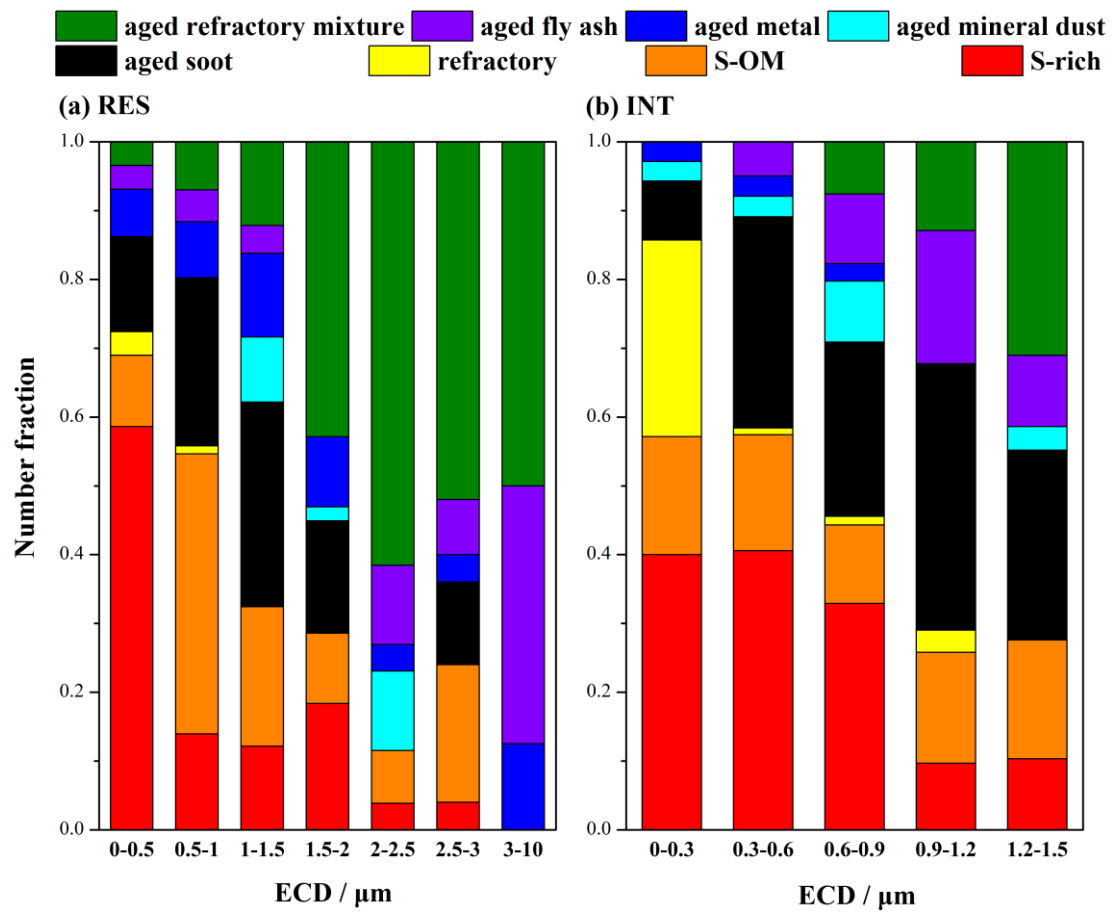
69 **when the size is less than 0.8 μm , and more RES particles when the size is larger than 0.8 μm .**



70

71 **Figure S7. Average positive and negative mass spectra of OM particles (OC and HMOC) of RES and INT**

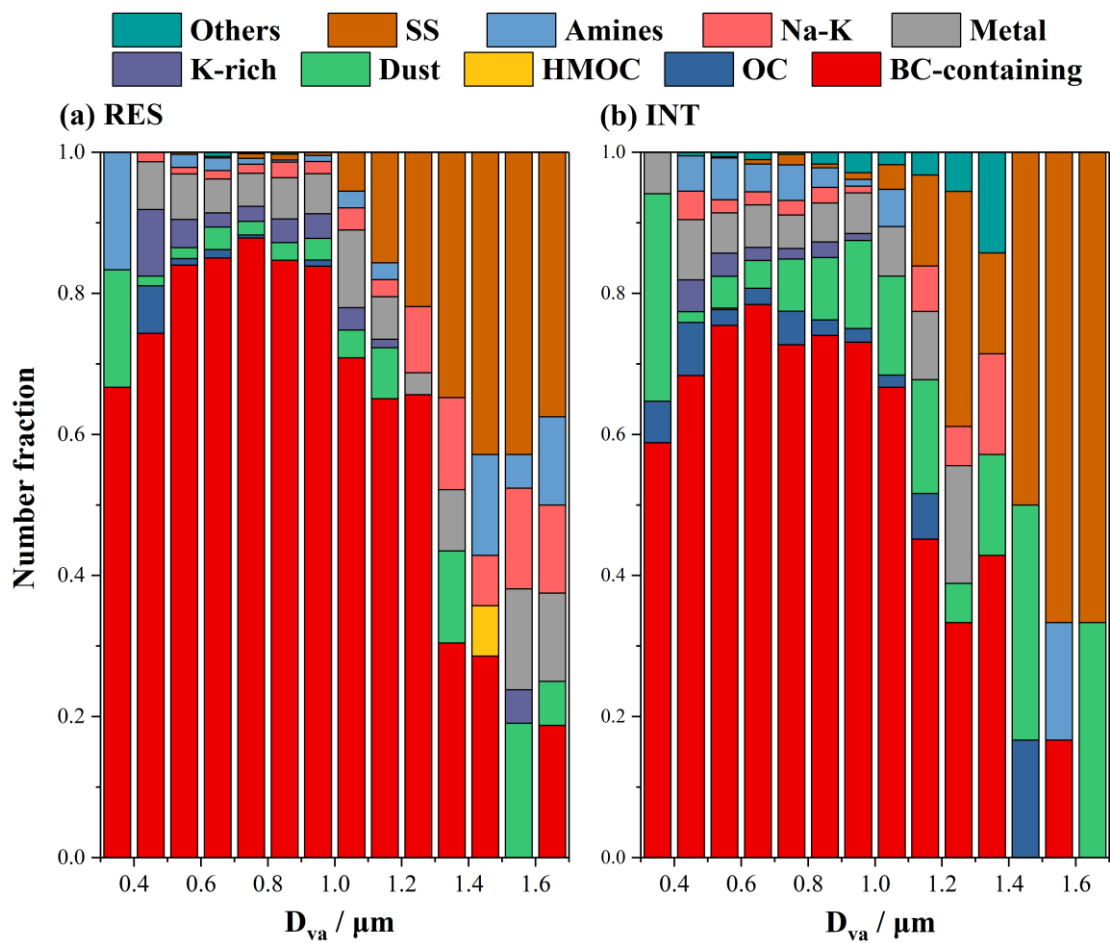
72 **particles measured by the SPAMS during cloud events #2 and #3.**



73

74

Figure S8. Size-resolved number fraction distributions of RES and INT by TEM/EDS.



75

76 Figure S9. Size-resolved number fraction distributions of RES and INT by the SPAMS.

77 **Table S1.** The concentration of NO_x, SO₂, O₃, PM₁₀ and PM_{2.5} during three cloud events.

78

cloud event	NO _x (ppb)	SO ₂ (ppb)	O ₃ (ppb)	PM ₁₀ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)
#1	2.6	0.4	30.5	3.6	1.1
#2	3.5	1.2	39.1	4.8	1.9
#3	4.3	0.6	34.4	11.4	4.7

79

80 **Table S2.** The ratios of relative peak area between organics (m/z 27, 29, 37, 43, 50, 51, 61, 63) and sulfate
81 (m/z -97) of OM particles (OC and HMOC) during in-cloud (RES and INT) and pre-cloud (Ambient)
82 periods.

83

84

	RES	INT	Ambient
Organics/Sulfates	1.68	1.57	1.59

85 **Table S3.** Morphological descriptors of soot particles within RES and INT.

86

parameters	A_p	d_p	L_{max}	N	D_f	k_g
RES	1658(175)	43(2)	255(12)	66(8)	1.82(0.12)	3.5(0.08)
INT	1842(133)	46(2)	316(16)	68(6)	2.11(0.09)	2.72(0.05)

87 A_p , mean projected area of the monomer; d_p , monomer diameter; L_{max} , maximum length of soot

88 aggregates; N , number of monomers in a soot aggregate; D_f , mass fractal dimension; k_g , structural

89 coefficient. In parentheses are the standard error of A_p , d_p , L_{max} , N , D_f and k_g .

90 **Table S4.** Overlap (δ), constant (k_a) and empirical exponent (α).

91

parameters	δ	k_a	α
RES	1.54	1.52	1.13
INT	1.4	1.44	1.11

92

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