

The authors appreciate the reviewers for reviewing our manuscript and providing constructive comments. As suggested, we carefully revised the manuscript thoroughly according to the valuable advices, as well as the typographical, grammatical, and bibliographical errors. Listed below are our point-by-point responses in blue to the review's comments (in italic). The figures added in the reply is represented by 'Figure', which is distinguished from 'Fig.' in the manuscript.

## Anonymous Referee #1

*This paper reports multimethod determination of the below-cloud wet scavenging coefficients of aerosols in Beijing, China. The analysis and interpretation of the results are overall fair. The paper presents very useful information regarding the wet deposition of aerosol. However, some additional information is still necessary for the readers to better understand this work.*

**[Response]:** Thanks for your suggestions and we have added the necessary information marked in the blue in the manuscript, such as section 2.2.3 (the description of modeling calculation), section 3.1 (the detailed introduction of this rain event) and section 3.2 (the detailed introduction of Fig. 3).

### *Specific comments:*

*1. The modeling analysis presented in this study is subject to some uncertainties. For example, the washout was only parameterized for precipitation intensity, the aerosol species and so on. The coagulation kernel ( $E$ ) was assumed to be a constant. What is the effect of this assumption on the modeling results? The authors should estimate the uncertainties of their modeling analysis, or at least state clearly the model configuration and limitation so that the readers can judge by themselves.*

**[Response]:** In the NAQPMS model, the below-cloud scavenging module from Comprehensive Air Quality Model with Extensions (CAMx) v4.42 was employed to calculate the below-cloud wet scavenging process and the wet scavenging coefficient was briefly described as follows:

$$K = \frac{4.2 \times 10^{-7} \times E \times P}{d_p} \quad (11)$$

where  $d_p$  is the mean rain drop size and related to precipitation intensity. The collision efficiency  $E$  is a function of aerosol particle size and mainly considers Brownian diffusion, interception and inertial impaction as shown in Figure. 1. According to Figure 1,  $E$  depends on the particle size, i.e. decreasing sharply as the diameter smaller than 400 nm, and slightly increasing from  $10^{-4}$  to  $10^{-3}$  when the particle size between 400 nm-2  $\mu\text{m}$ , after that it increasing very quickly. Thus, choose the  $E$  for different particle size will cause uncertainties in BWSC of one to two orders of magnitude for particle size in range of 0.01-2.5  $\mu\text{m}$ . NAQPMS used in this study assumed SNA resides in fine mode size range (0.1-2.5  $\mu\text{m}$ ) and the geometric mean diameter of 0.5  $\mu\text{m}$  was used in the calculation of  $E$ .

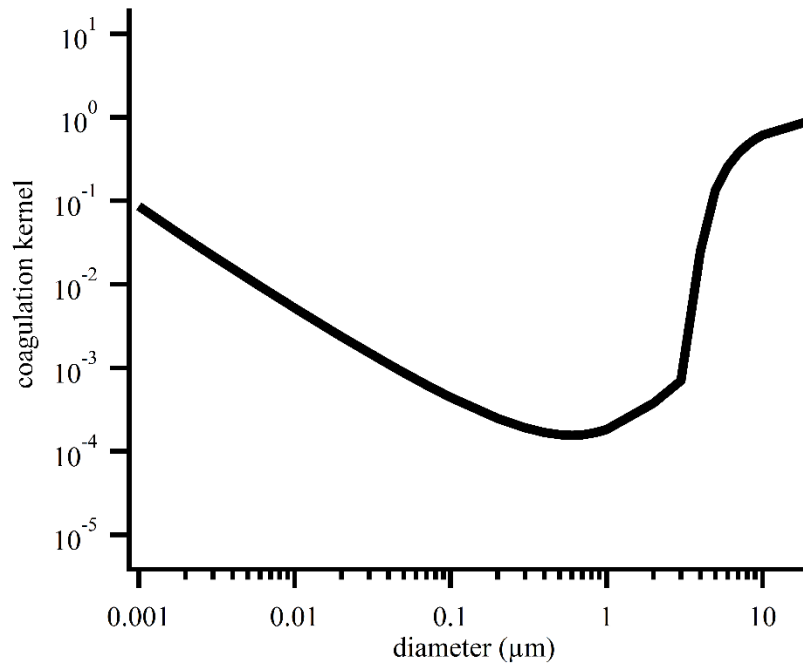


Figure 1. Size-resolved coagulation kernel based on the Brownian diffusion, interception and inertial impaction

We revised the model configure as “*The below-cloud scavenging module from Comprehensive Air Quality Model with Extensions (CAMx) v4.42 was employed to calculate the below-cloud wet scavenging process and the wet scavenging coefficient was briefly described as follows (Environ, 2005):*

$$K = \frac{4.2 \times 10^{-7} \times E \times P}{d_p} \quad (11)$$

where  $d_p$  is the mean rain drop size and related to precipitation intensity. The collision efficiency  $E$  is a function of aerosol particle size and mainly considers Brownian diffusion, interception and inertial impaction. NAQPMS used in this study assumed SNA resides in fine mode size range (0.1-2.5  $\mu\text{m}$ ) and the geometric mean diameter of 0.5  $\mu\text{m}$  was used in the calculation of  $E$ .”

And we also added the more description of NAQPMS in section 2.2.3 as “In this study, a three-dimensional regional model, the Nested Air Quality Prediction Modeling System (NAQPMS) was adopted to calculate the aerosol scavenging coefficient. The NAQPMS, developed by IAP, is a fully modularized chemical transport model describing regional and urban-scale air pollution (Wang et al., 2001). The meteorological condition is driven by Weather Research and Forecasting (WRF) model. The NAQPMS consists of modules used for horizontal and vertical advection (Walcek and Aleksic, 1998), diffusion (Byun and Dennis, 1995), dry and wet deposition (Zhang et al., 2003; Stockwell et al., 1990), gaseous phase, aqueous phase, and heterogeneous atmospheric chemical reactions (Zaveri and Peters, 1999; Stockwell et al., 1990; Li et al., 2012). Carbon-Bond Mechanism Z (CBM-Z) and aerosol thermodynamic equilibrium partition model (ISORROPIAII.7) have been used to calculate the gas and inorganic aerosol process. The cloud-process and aqueous chemistry module from Community Multi-scale Air Quality (CMAQ) modeling system v4.7 have been coupled in model by Ge et al. (2014). More details can be found in Li et al. (2016, 2017a). The NAQPMS has been widely used in prediction of acid rain, dust and secondary pollutions and can also reproduce well the physical and chemical evolution of reactive pollutants by solving the mass balance equations in terrain-following coordinates (Chen et al., 2019; Yang et al., 2019). It has been applied in Ministry of Ecology and Environment and local Environmental Protection Bureau such as Beijing, Shanghai, Guangzhou and Nanjing, etc. The NAQPMS also made great contribution to air quality assurance during the major activities (Wang et al., 2001; Wang et al., 2014d; Wu et al., 2010).”

2. A brief introduction to Fig.3 should be provided, which should be much helpful for

*the readers to better understand this study.*

**[Response]:** We added more introductions of the symbols in the caption to Fig. 3 as “*The top and bottom of the boxes represent the 75<sup>th</sup> and 25<sup>th</sup> percentiles, and central lines mean the median BWSCs. The whiskers represent maximum and minimum BWSCs, respectively.*”

Besides, for better understanding of multimethod, we have revised the symbols for multimethod and unified the expressions in the manuscript as: “*The theoretical estimated scavenging coefficients are labeled T. The field observations estimated by Eq (3) and (9) are labeled O1 and O2, respectively. The updated estimated method by Eq (10) is labeled as O2'. The modeling results are labeled M. And in following comments, we use the updated symbols instead. It will not be repeated later.*”

We also added the detailed description for Fig. 3 in section 3.2 as “*The observed O1 by SMPS, which mostly covers the range of Aitken and accumulation mode aerosols (0.014-0.74  $\mu\text{m}$ ), are much lower than the other two measurements. The observed BWSCs by original O2 are larger than the updated O2' method. However, O2' ( $5.7 \times 10^{-5}$ ,  $8.9 \times 10^{-5}$  and  $5.4 \times 10^{-5} \text{ s}^{-1}$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ ) is much closer to the results of O1 ( $\sim 10^{-5} \text{ s}^{-1}$  for particle size in the range of 0.014-10.35  $\mu\text{m}$ ).*

3. *There are only two cases in the paper, more field measurement estimation could decrease the influence produced by accidental elements, and give more convincing results.*

**[Response]:** We agree with the reviewer’s comment as well. In order to make up the bias from the limited rain events, we added the nine rain events at the same sampling site in IAP in summer of 2014 to compare the estimation methods of BWSCs of different precipitation with the same method O2’. Our results found that there is a strong exponential power relationship between the BWSCs and precipitation intensity both in the summer of 2014 and the rainfall event in winter of APHH-Beijing campaign with the coefficients of determination for SNA are over than 0.68. It indicated that rainfall event in winter of APHH-Beijing campaign also obeys the general wet scavenging rule in Beijing.

Although we cannot obtain the SMPS, SPAMS and POPC data in the summer of

2014 to compare the BWSCs under the multimethod, the same method the O2' has been employed to estimate the BWSCs in multi-event. Still, the multimethod to determinate the BWSCs of one rain event and even for multimethod of different rainfall events are needed for the future research.

The authors appreciate the reviewers for reviewing our manuscript and providing constructive comments. As suggested, we carefully revised the manuscript thoroughly according to the valuable advices, as well as the typographical, grammatical, and bibliographical errors. Listed below are our point-by-point responses in blue to the review's comments (in italic). The figures added in the reply is represented by 'Figure', which is distinguished from 'Fig.' in the manuscript.

## **Anonymous Referee #2**

*The manuscript 'Multimethod determination of the below-cloud wet scavenging coefficients of aerosols in Beijing, China' written by Danhui Xu investigated wet scavenging process during APHH-Beijing. The authors tried to clarify well-known but not well-understood wet deposition process based on theoretical measure, field observation, and numerical model. The findings through this study is essential to understand the wet deposition process and also can contribute to refine the wet deposition scheme in CTMs and reduce its uncertainty. Although I would like to recommend to publish this manuscript, the following comments should be addressed with in-depth discussion for furthermore understanding.*

**[Response]:** We thank the reviewer for giving the positive comments. With response to the comments, the revised manuscript with in-depth discussion is much easier to understand for the readers, especially in section 2.2.3 (the description of modeling calculation), section 3.1 (the detailed introduction of this rain event) and section 3.2 (the detailed introduction of Fig. 3). And we also added the observation data list, the introduction of the nine precipitations in summer of Beijing, 2014, and more abbreviations in the supplementary file.

### **Major comments:**

*1. P8, L192-193 (Symbols used in this manuscript):*

*I could follow the results and discussion section; however, it will be straightforward to use T, F (or O), and M to indicate theory, field observation, and modeling calculation, and these expressions are easy to catch discussion. Or, please include the rationale to use same M for field observation.*

**[Response]:** For better understanding of multimethod, we have revised the symbols for multimethod and unified the expressions in the manuscript as: “*The theoretical estimated scavenging coefficients are labeled T. The field observations estimated by Eq (3) and (9) are labeled O1 and O2, respectively. The updated estimated method by Eq (10) is labeled as O2'. The modeling results are labeled M.*” And in following comments, we use the updated symbols instead. It will not be repeated later.

2. P9, L220-227 (Discussion on BWSCs in Figure 3):

*More discussion is required on the following two points.*

*- What is M1 difference calculated by SMPS, POPC, and SPAMS? The particle size should be referred, though this can be partly covered in Figure 4. Please explicitly define the box plot shown in Figure 3. What percentiles (and/or average) are used to draw it?*

**[Response]:** The M1, now is called O1, calculated by the number concentrations collected from SMPA, SPAMS and POPC, respectively. The SMPS mainly measure particle number concentration from 14 to 740 nm. The SPAMS mainly measure the various chemical compositions with particle diameters ranging from 0.2 to 2.5  $\mu\text{m}$ , while the POPC mainly focus on the coarse diameter of 0.4-10.35  $\mu\text{m}$  (Table. s1). For readers' convenience, we added the observation instruments message in Fig. 4 caption. And we also added the description as “*The observed O1 by SMPS, which cover the range of Aitken and accumulation mode aerosols (0.014-0.74  $\mu\text{m}$ ), are much lower than the other two measurements (0.2-2.5  $\mu\text{m}$  for SPAMS and 0.4-10.35  $\mu\text{m}$  for POPC, respectively).*”

In Fig. 3, we have added the detailed introduction as: “*Fig 3. Box and whisker plots of the multimethod estimation of the BWSCs. The top and bottom of the boxes represent the 75<sup>th</sup> and 25<sup>th</sup> percentiles, and central lines mean the median BWSCs. The whiskers represent maximum and minimum BWSCs, respectively.*”

*- M2 and M2' can generally agree with M1; however, the significant point should be the distinction of chemical species. From M1 of SPAMS, it seems no difference on SNA whereas M2 and M2' calculated larger BWSC for S. What are reasons to these results of SNA?*

**[Response]:** There are two main reasons. One is the selection of the time. As we mentioned in section 3.1, due to the light rain intensity in early stage (no more than 0.5 mm from 7:00 AM to 16:00 PM, hourly rainfall data have been added in the revised Figure 1), the data from 16:29 PM on November 20<sup>th</sup> is taken as the beginning time of the rainfall event in calculating the O1. The data from 7:00 AM to 16:00 PM was represented as “before rainfall events” and calculated the mean value for SNA as well as the aerosol numbers. However, the data from SPAMS, 3 hours data from 7:00 AM -10:00 AM were missing. The missing data may result some uncertainties in estimating the BWSC using O1. In general, the median BWSCs of NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> in different particles sizes are  $4.6 \times 10^{-5}$ ,  $4.4 \times 10^{-5}$  and  $5.0 \times 10^{-5}$  s<sup>-1</sup> in O1 of SPAMS, respectively. However, for PM<sub>2.5</sub> (total aerosols with particle size smaller than 2.5 μm) of BWSCs estimated by O2’ are  $5.7 \times 10^{-5}$ ,  $8.9 \times 10^{-5}$  and  $5.4 \times 10^{-5}$  s<sup>-1</sup>, respectively, which indicated larger BWSC for SO<sub>4</sub><sup>2-</sup>. In spite of this, the two methods estimated similar BWSC for NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>.

Another one is the different sampling site of SPAMS with the other analyzers (SMPS, POPC, etc). The SPAMS is deployed in the China National Environmental Monitoring Centre (CNEMC), which is located in the northeast, 8 km away from the IAP sampling site. This site is a typical suburban site and mainly affected by residential source. (Figure. 2). Before this rainfall event, Beijing was occurring an air pollution case. The measurement in IAP site found that the NOR/SOR (oxidation ratios of NO<sub>x</sub>/oxidation ratios of SO<sub>2</sub>) decreased to 0.84, while the SO<sub>4</sub><sup>2-</sup>/NO<sub>3</sub><sup>-</sup> increased to 0.38, which implicated a special increasing of SO<sub>4</sub><sup>2-</sup> in this event. The especially high SO<sub>4</sub><sup>2-</sup> concentration is mainly due to speeding up of SO<sub>2</sub> oxidation inducing by the NH<sub>4</sub>NO<sub>3</sub>, which drives the increasing of aerosol water content and trigger the positive feedback between AWC and aerosol secondary aerosol formation (Ge et al., 2014). The high proportion of S in aerosols were much easier to be scavenged by the rainfall, as the data of NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> decreased from 50.1, 70.6 and 25.3 mg L<sup>-1</sup> to 28.5, 25.2 and 10.3 mg L<sup>-1</sup> (a reduction of 43.2, 64.3 and 59.5%) in the rainwater shown in Figure. s2. Therefore, the much higher BWSCs of SO<sub>4</sub><sup>2-</sup> estimated by O2’ in IAP site was reasonable. We added the sampling site



information in section 2.1 as “It’s deployed during the measuring time in China National Environmental Monitoring Centre (CNEMC), which is located in the northeast, 8 km away from the IAP sampling site. This site is a typical suburban site and mainly affected by residential source.”

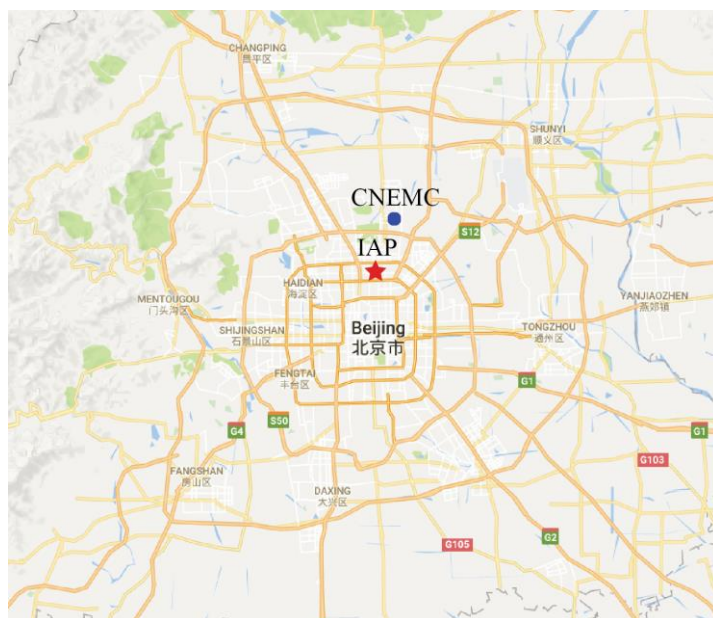


Figure.2 The sampling site in IAP, Beijing (marked in red) and CNEMC (marked in blue)

3. P10, L256-257 (Relationship of BWSC and precipitation shown in Figure 5):

Data in 2014 summertime is essential to see this relationship; hence detail descriptions of this summer campaign is needed (will be appropriate introduced in Section 2.1). When compared winter results with summer results, it should be noticed that BWSC of  $\text{NO}_3$  (up to  $10^{-3} \text{ sec}^{-1}$ ) is greater than that of  $\text{SO}_4$  and  $\text{NH}_4$  during summertime. This is reversed finding during wintertime, because BWSC of  $\text{SO}_4$  is greater than  $\text{NO}_3$ . What is reasons of these BWSC differences found in summer and winter?

**[Response]:** We added the detail descriptions of precipitation in summer of 2014 in supplementary file as “The Figure. s2 shows the average concentrations of SNA in summer of 2014 (Box and whisker plot) and this rain event in winter of APHH-Beijing campaign. The VWA concentrations are no more than 30, 40 and 15  $\text{mg L}^{-1}$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ , respectively, and decreased sharply during the beginning of rainfall and remained at low levels during the event. The in-cloud scavenging process is

*considered as the median value of the concentrations after accumulated precipitation exceeds 5 mm. These values were 2.75, 3.33, and 2.51 mg L<sup>-1</sup> for NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup>, respectively, in summer of 2014 (as shown in Figure. s2 marked in grey shadow) (Xu et al., 2017)."*

In normal case, NO<sub>3</sub><sup>-</sup> bearing compounds in coarse particles are much more easily scavenged than SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> (Xu et al., 2017) same with the fitting line as shown in previous results in summer of 2014 in Fig. 5. However, it is different in this rainfall event in winter of APHH-Beijing campaign. According to the calculation of O2', the larger difference of concentration between the beginning fraction and the later fraction in rainfall events, the more efficiently of the below-cloud scavenging will be estimated. In this rain event, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> decreased from 50.1, 70.6 and 25.3 mg L<sup>-1</sup> to 28.5, 25.2 and 10.3 mg L<sup>-1</sup> (a reduction of 43.2, 64.3 and 59.5%) in the rainwater (Figure. s2), with the largest ratio of reduction for SO<sub>4</sub><sup>2-</sup>. Before this rainfall event, Beijing was occurring an air pollution case. The measurement in IAP site found that high SO<sub>4</sub><sup>2-</sup> concentration in aerosols. For example, the NOR/SOR (oxidation ratios of NO<sub>x</sub>/oxidation ratios of SO<sub>2</sub>) decreased to 0.84, while the SO<sub>4</sub><sup>2-</sup>/NO<sub>3</sub><sup>-</sup> increased to 0.38 from pre-polluted period to pollution period. The especially high SO<sub>4</sub><sup>2-</sup> concentration is mainly due to speeding up of SO<sub>2</sub> oxidation inducing by the NH<sub>4</sub>NO<sub>3</sub>, which drive the increasing of aerosol water content and trigger the positive feedback between AWC and aerosol secondary aerosol formation (Ge et al., 2014). The high proportion of S in aerosols were much more easier to be scavenged by the rainfall. Therefore, the much higher BWSCs of SO<sub>4</sub><sup>2-</sup> estimated by O2' in this event was reasonable.

However, due to the uncertainties of limited rainfall event during this campaign, the comparison of the multi-method estimation of BWSC needed more strong evidence as well as more field measurement data in various polluted conditions in the future.

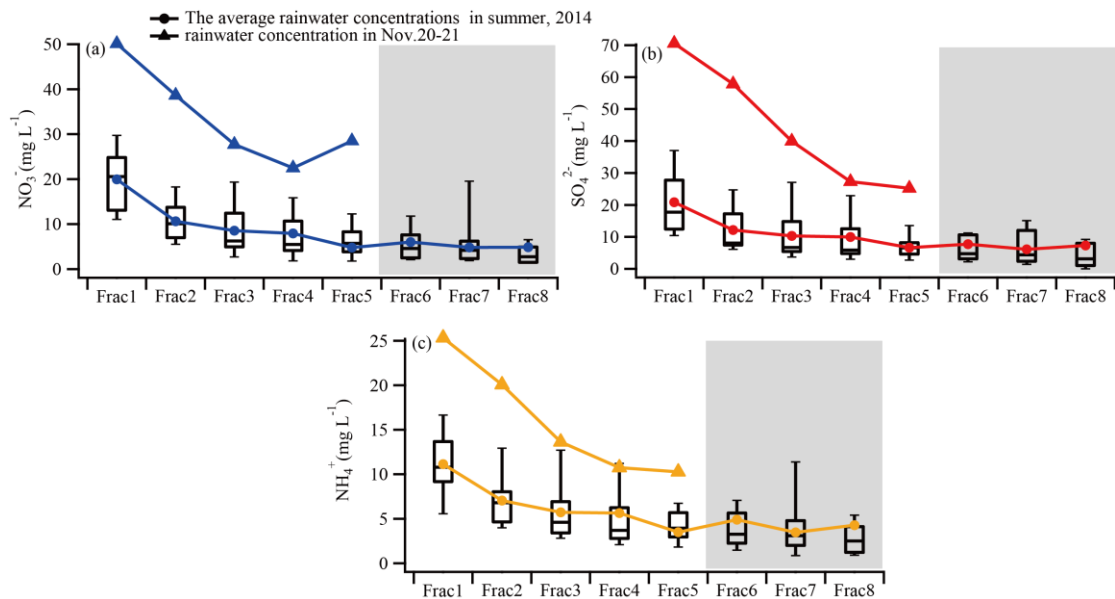


Figure s2. Evolution of the (a) NO<sub>3</sub><sup>-</sup> (blue), (b) SO<sub>4</sub><sup>2-</sup> (red) and (c) NH<sub>4</sub><sup>+</sup> (orange) of precipitation during summer 2014 and November 20<sup>th</sup> to 21<sup>st</sup> within different precipitation fractions of several sampled precipitation events (The data show the lowest, lowest 25 percentiles, median highest quartile, highest 75 percentiles, and highest value, respectively).

### Minor points:

1. P2, L39: Do developed countries really have clean atmosphere?

**[Response]:** No, sorry for our un-precise saying. And we revised this sentence as “This may be true in most clean atmosphere, e.g., some clean regions where air pollutants in the boundary layer were not sufficient.”

2. P2, L40-42: Which species are underestimated in MICS-Asia?

**[Response]:** “Recently, some regional models in MICS-Asia (Model Inter-Comparison Study for Asia) obviously underestimated SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> wet deposition in East Asia.” And we also added the species message in the manuscript.

3. P2, L42-44: How about sulfur species in TF-HTAP?

**[Response]:** The previous studies point the overestimated modeled values of volume weighted averaged (VWA) sulfur (S) in Europe and North America while underestimated S concentration values in Asia (Vet et al., 2014). And we also added the message in this manuscript as follows: “For global model assessment by

*Hemispheric Transport of Atmospheric Pollutants (TF-HTAP), wet depositions of nitrogen were also underpredicted in region of North America, Europe and Asia where measured the high level of volume weighted averaged (VWA) nitrogen (N) concentrations in rainfall as  $> 1.25 \text{ mg N L}^{-1}$ , as well as underestimated sulfur wet deposition in Asia (Vet et al., 2014).”*

4. P3, L47-48: *Are these estimations suggested the importance of below-cloud scavenging based on what?*

**[Response]:** According to the sequential sampling field measurements in polluted region, such as India and North China, below-cloud scavenging contributed to more than 53%. It indicated that below-cloud scavenging process is crucial. And in order to make the manuscript more accurate we revised as “*which contributed to more than 53% of the total wet deposition in some polluted areas such as India (Chatterjee et al., 2010) and North China (Ge et al., 2016; Xu et al., 2017) on the basis of sequential sampling field measurements.*”

5. P3, L49-51: *Also, are these estimations based on what?*

**[Response]:** These estimations are not only based on the field measurement but also on the modeling calculation. We also revised this sentence as “*can remove 50-80% of the number or mass concentration of below-cloud aerosols both by filed measurements and modeling calculation.*”

6. P3, L47-51: *If these estimations are based on model, how can we say its importance under the large uncertainty of modeling treatment?*

**[Response]:** The model calculation indeed has large uncertainty of one to two orders of magnitude for BWSC estimation (Wang et al., 2010; Wang et al., 2014b). However, not only the field measurement but also modeling calculation showed that precipitation could scavenge 50-80% of the number or mass concentration of below-cloud aerosols. Combining these two methods, it reveals the importance of the below-cloud wet scavenging.

7. P3, L57: *Does this sentence regarding gravity refer dry deposition?*

**[Response]:** Yes, but in this manuscript we mainly focus on the wet deposition, we

prefer to delete this sentence “*Especially coarse particles ( $d_p > 20 \mu\text{m}$ ) are also easily scavenged through the effect of gravity*”.

8. P4, L90-L92: *I am not sure the meaning of “rarely in autumn winter” and “typical rainfall in winter”, and these expressions will make some confusion. How rare the winter rain and what is the typical rain?*

**[Response]:** The “rare” means the precipitation is less in autumn and winter than summer. A lot of literatures mentioned that more than 80% rain events in Beijing concentrate in summer (Chen et al., 2013; Han et al., 2019). The “typical” means the moving path of the precipitation as usually used in previous studies in Beijing. Previous studies have mentioned that precipitations usually generate in the northwest mountainous area of Beijing and move along the steering flow to southeast (Xiao et al., 2015). According to the radar base reflectivity in this case, the rain event gradually moved from northwest to southeast, which is same as the “typical” rainfall moving path. To avoid the ambiguity, we revised manuscript as follows: *“In North China, precipitations were mainly concentrated in summer (more than 80%) but rare in autumn and winter (Xu et al., 2008; Gao et al., 2015; Chen et al., 2013; Han et al., 2019). However, the air pollution events were usually occurred in autumn and winter in North China Plain (NCP). Here we select a typical rainfall event moving from northwest to southeast in winter of Beijing...”*

9. P4, L111-L112: *Here, “in the winter of 2016” indicates averaged VWA concentrations during APHH-Beijing 2016 campaign? It will be helpful to show the period (on L98). In addition, VWA concentrations during previous studies (Pan et al., 2013, 2012; Xu et al., 2017) can be added here for the discussion to strength the polluted episode analyzed in this study.*

**[Response]:** No, here in line 111-112 means the winter of 2016. As we mentioned in last comment, the rain event in winter is rare and there’s only one rain event during the APHH-Beijing campaign (The case we analyze in this study). In order to strengthen the comparison, we added previous studies’ results as follows: *“much higher than the VWA concentrations in the winter of 2016 (8.3, 9.5 and 4.1 mg L<sup>-1</sup>,*

respectively) and in previous studies in Beijing (6.3, 9.1 and 4.9 mg L<sup>-1</sup> in Pan et al. (2012, 2013) and 6.2, 7.9 and 4.6 mg L<sup>-1</sup> in Xu et al. (2017) of summer).”

10. P4, L118 and L122: Where is the sampling location of SPAMS and POPC? Same as wet-only sampler or SMPS?

Reply: The sampling site of POPC is same as the SMPS. However, the SPAMS is deployed in National Environmental Monitoring Centre (CNEMC), which is located in the northeast, 8 km away from the IAP sampling site. This site is a suburban site and mainly affected by residential source. For better reading, we also supplemented the message in the manuscript as “A single-particle aerosol mass spectrometer (SPAMS) can accurately characterize aerosol particles containing various chemical compositions with diameters ranging from 0.2 to 2.5  $\mu\text{m}$ . It’s deployed during the measuring time in National Environmental Monitoring Centre (CNEMC), which is located in the northeast, 8 km away from the IAP sampling site. This site is a typical suburban site and mainly affected by residential source. More detailed fundamentals of the SPAMS and description can be found in Li et al. (2011), Lin et al. (2017) and Cheng et al. (2018). Size-resolved airborne  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  are the main focuses in this study, and the time resolution is 1 hour. In the meantime, a polarization optical particle counter (POPC) is also deployed to obtain coarse particle (0.4-10.35  $\mu\text{m}$ ) size distribution at the IAP sampling site, and time resolution is 5-min. Detailed description and settings can be found in Tian et al. (2018).”

11. P4, L111-L123: I feel the summary of these observation dataset listed in one table (full name, abbreviation, target species, and short explanation, etc.) can help the reader to understand these observations used in this study.

**[Response]:** We added the summary of the observation dataset in the supplementary file as follows:

Table s1. The observation dataset list

Observation instrument	Abbreviation	Introduction (time-resolution and mainly measured material)
Ion Chromatography	IC	anions ( $\text{SO}_4^{2-}$ , $\text{NO}_3^-$ , $\text{Cl}^-$ and $\text{F}^-$ ) and cations ( $\text{NH}_4^+$ , $\text{Na}^+$ , $\text{K}^+$ , $\text{Ca}^{2+}$ and $\text{Mg}^{2+}$ ) in the

		rainfall samples
Ambient Ion Monitor-Ion Chromatograph	AIM-IC	60 min resolution, PM <sub>2.5</sub> concentrations
Scanning Mobility Particle Sizer	SMPS	5 min resolution, 14-740 nm particle number concentration
Single-particle Aerosol Mass Spectrometer	SPAMS	60 min resolution, various chemical compositions with 0.2-2.5 μm particle number concentration, mainly focus on NO <sub>3</sub> <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> and NH <sub>4</sub> <sup>+</sup>
Polarization Optical Particle Counter	POPC	5 min resolution, 0.4-10.35 μm particle number concentration

12. P5, L125-: *The authors can put the notice that abbreviations are available in supplemental material.*

**[Response]:** The abbreviations have added in the supplementary file.

13. P7, L184-185: *More information of NAQPMS will be needed. What kind of numerical model? Are there any previous studies show the modeling performance by NAQPMS? This can also motivate the authors to improve the model performance through this study.*

**[Response]:** We have added more description of the NAQPMS: *“In this study, a three-dimensional regional model, the Nested Air Quality Prediction Modeling System (NAQPMS) was adopted to calculate the aerosol scavenging coefficient. The NAQPMS, developed by IAP, is a fully modularized chemical transport model describing regional and urban-scale air pollution (Wang et al., 2001). The meteorological condition is driven by Weather Research and Forecasting (WRF) model. The NAQPMS consists of modules used for horizontal and vertical advection (Walcek and Aleksic, 1998), diffusion (Byun and Dennis, 1995), dry and wet deposition (Zhang et al., 2003; Stockwell et al., 1990), gaseous phase, aqueous phase, and heterogeneous atmospheric chemical reactions (Zaveri and Peters, 1999; Stockwell et al., 1990; Li et al., 2012). Carbon-Bond Mechanism Z (CBM-Z) and aerosol thermodynamic equilibrium partition model (ISORROPIAII.7) have been used to calculate the gas and inorganic aerosol process. The cloud-process and aqueous chemistry module from Community Multi-scale Air Quality (CMAQ)*

*modeling system v4.7 have been coupled in model by Ge et al. (2014). More details can be found in Li et al. (2016, 2017a). The NAQPMS has been widely used in prediction of acid rain, dust and secondary pollutions and can also reproduce well the physical and chemical evolution of reactive pollutants by solving the mass balance equations in terrain-following coordinates (Chen et al., 2019; Yang et al., 2019). It has been applied in Ministry of Ecology and Environment and local Environmental Protection Bureau such as Beijing, Shanghai, Guangzhou and Nanjing, etc. The NAQPMS also made great contribution to air quality assurance during the major activities (Wang et al., 2001; Wang et al., 2014d; Wu et al., 2010).”*

*14. P8, L200-201 (Figure 1): Please include hourly (or finer time-scale) precipitation in Figure 1. We cannot catch the rainfall intensity from this sentence. Especially, we can see slight increase of aerosol concentration (except NO<sub>3</sub>) before 12:00 and what is the relation of this increase and precipitation? In my personal opinion, hourly concentration averaged during 11 November to 11 December is not needed here; remove this or add the discussion.*

**[Response]:** We have added the hourly precipitation intensity and detailed captions in Fig. 1. Clearly can be seen that the precipitation is weak in the light rain period (no more than 0.5 mm from 7:00 AM to 16:00 PM) with the slight increase of aerosol concentration. For better comparison with variation of SNA in precipitation, hourly concentration of SNA in aerosols have been displayed in Figure 1. Also compared with the monthly data, the sharply decreasing after 16:00 PM indicated that the impacts of below-cloud scavenging on aerosols concentrations. Based on this intention, the hourly averaged concentration during the APHH-Beijing was remained. As suggested, the hourly precipitation intensity are added and the manuscript is revised as “*It’s clearly visible in Fig. 1 that all aerosol concentrations on the rainy day were much lower than the hourly averaged aerosol concentrations during the APHH-Beijing campaign, especially during the precipitation time indicating the below-cloud scavenging impacts.*”

*15. P8, L208: From what to decreased by nearly 6  $\mu\text{g m}^{-3}$  in the air? Need to add the variation about N and A.*



**[Response]:** “ $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  decreased from 50.1, 70.6 and 25.3  $\text{mg L}^{-1}$  to 28.5, 25.2 and 10.3  $\text{mg L}^{-1}$  (or a reduction of 43.2, 64.3 and 59.5%) in the rainwater. Accordingly, aerosol nitrate, sulfate and ammonium decreased from 13.8, 8.3 and 8.4  $\mu\text{g m}^{-3}$  to 1.2, 2.2 and 0.1  $\mu\text{g m}^{-3}$  in the air (decreased by more than 6  $\mu\text{g m}^{-3}$ ).” And we have added this description in the manuscript.

16. P9, L244: Typo of “Fig. 4”?

**[Response]:** Yes, we have revised it.

17. P10, L259: Figure 5 shows  $R^2$ , hence “coefficient of determination” is appropriate (or, Figure 5 include typo of  $R^2$ ?).

**[Response]:** In L259, P10, the description changed to “coefficient of determination”.

18. P10, L277-P11, L295 and caption of Table 2: To include the information of ambient concentration before the rainfall events (stated in P11, L285) is useful if explicitly stated in the caption.

**[Response]:** We have added the original aerosol concentrations before the rainfall into Table 2 as shown in the manuscripts.

19. P11, L294: I guess that NMB is usually ‘normalized mean bias’. Is this typo or some kind of another metric?

**[Response]:** Yes, it’s a typo and we have revised it as “Normalized Mean Bias”.

### Technical comments:

1. P8, L203: Typo of “NAAQS”?

**[Response]:** Yes, we have revised it.

2. P13, L331-332: Check this reference style. There is no information related to volume, page, and year.

**[Response]:** We have revised it in the manuscript. “Andronache, C., Grönholm, T., Laakso, L., Phillips, V., and Venäläinen, A.: Scavenging of ultrafine particles by rainfall at a boreal site: observations and model estimations, *Atmos. Chem. Phys.*, 6, 4739-4754, 2006.”

3. P14, L354-356: Check this reference style regarding the location of year.

**[Response]:** We have revised the location of the year. “Gao, X. D., Chen, X Y., Ding,

Z. W., Yang, W. Q. Investigation of the variation of atmospheric pollutants from chemical composition of precipitation along an urban-to-rural transect in Beijing. *Acta Scientiae Circumstantiae (in Chinese)*, 35(12): 4033-404, 2015.”

4. P15, L390-391: This is not the latest edition.

**[Response]:** We have revised the reference to latest edition as: “Seinfeld, J. H., and S.N. Pandis.: *Atmospheric chemistry and physics: from air pollution to climate change*. John Wiley and Sons, Inc., NY, *Atmospheric Chemistry and Physics*, 2016”, and also check this reference throughout the manuscript.

5. P16, L445-447: Check this reference style regarding the location of year.

**[Response]:** We have revised the location of the year. “Xu, J., Zhang, X. L., Xu, X. B., Ding, G. A., Yan, P., Yu, X. L., Chen, H. B., Zhou, H. G. Variations and source identification of chemical compositions in wet deposition at Shangdianzi background station. *Acta Scientiae Circumstantiae (in Chinese)*, 28(5):1001 -1006, 2008.”

6. Throughout the manuscript, please unify “APHH” or “APHH-Beijing”.

**[Response]:** We have unified the description of “APHH-Beijing” through the manuscript.

7. Throughout the manuscript, please remove period “mg.m<sup>-3</sup>” and “μg.m<sup>-3</sup>”.

**[Response]:** We have removed the period in “mg.m<sup>-3</sup>”, “μg.m<sup>-3</sup>” and “mg.L<sup>-1</sup>” throughout the manuscript.

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# Multimethod determination of the below-cloud wet scavenging coefficients of aerosols in Beijing, China

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**Abstract.** Wet scavenging is one of the most efficient processes that remove aerosols from the atmosphere. This process is not well constrained in chemical transport models (CTMs) due to a paucity of localized parameterization regarding below-cloud wet scavenging coefficient (BWSC). Here we conducted field measurements of the BWSC during the Atmospheric Pollution and Human Health-Beijing (APHH-Beijing) campaign of 2016. Notably, the observed BWSC values based on the updated aerosol mass balance agree well with another estimation technique, and they fall in a range of  $10^{-5} \text{ s}^{-1}$ . The measurement in this winter campaign, combined with that in summer of 2014, supported an exponential power distribution of BWSCs with rainfall intensity. The observed parameters were also compared with both the theoretical calculations and modeling results. We found that the theoretical estimations can effectively characterize the observed BWSCs of aerosols with size smaller than  $0.2 \mu\text{m}$  and larger than  $2.5 \mu\text{m}$ . However, the theoretical estimations were one magnitude lower than observed BWSCs within  $0.2\text{-}2.5 \mu\text{m}$ , a domain size range of urban aerosols. Such an underestimation of BWSC through theoretical method has been confirmed not only in APHH-Beijing campaign but also in all the rainfall events in summer of 2014. Since the model calculations usually originated from the theoretical estimations with simplified scheme, the significant lower BWSC could well explain the underprediction of wet depositions in polluted regions as reported by the Model Inter-Comparison Study for Asia (MICS-Asia) and the global assessment of the Task Force on Hemispheric Transport of Atmospheric Pollutants (TF-HTAP). The findings highlighted that the wet deposition module in the CTMs requires improvement based on field measurement estimation to construct a more reasonable simulation scheme for BWSC, especially in polluted regions.

## 1 Introduction

Wet deposition is one of the dominant aerosol sinks on both global and regional scales (Min et al., 2005;Textor et al., 2006), and can be divided into in-cloud (particles are activated as cloud condensation nuclei and absorbed by cloud water) and below-cloud scavenging (aerosols and gas are captured by raindrops or snow particles after the hydrometeors leave the clouds) (Zhao et al., 2015). Previously, below-cloud scavenging is thought to be less important than in-cloud process and always simplified or even ignored in most global and regional chemical transport models (CTMs)(Tang et al., 2006;Bae et al., 2010;Barth et al., 2000;ENVIRON.Inc, 2005;Stier et al., 2005). This may be true in most clean atmosphere, e.g., some clean regions where air pollutants in the boundary layer were not sufficient. This may be not the case in polluted regions. Recently, some regional models in MICS-Asia (Model Inter-Comparison Study for Asia) obviously underestimated  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  wet deposition in East Asia (Wang et al., 2008). For global model assessment by Hemispheric Transport of Atmospheric Pollutants (TF-HTAP), wet depositions of nitrogen were also underpredicted in region of North America, Europe and Asia where measured the high level of volume weighted averaged (VWA) nitrogen (N) concentrations in rainfall as  $> 1.25 \text{ mg N L}^{-1}$ , as well as underestimated sulfur wet deposition in Asia (Vet et al., 2014). Besides the uncertainties in emission inventory and chemical mechanism, the below-cloud scavenging process may also contribute to certain effects on the wet deposition simulation (Wang et al., 2008). Actually, below-cloud scavenging cannot be negligible in CTMs, which

contributed to more than 53% of the total wet deposition in some polluted areas such as India (Chatterjee et al., 2010) and North China (Ge et al., 2016; Xu et al., 2017) [on the basis of sequential sampling field measurements](#).

50 Extensive efforts have been focused on the study of wet scavenging, and many researchers have noted that precipitation, even light rain, can remove 50-80% of the number or mass concentration of below-cloud aerosols [both by field measurements and modeling calculation](#) (Andronache, 2004b; Zhang et al., 2004). The below-cloud wet scavenging coefficient (hereafter, BWSC), denoted  $K$  or  $K(d_p)$  for size-resolved values, is a parameter that describes scavenging ability characteristics fairly well. The main factors affecting the BWSC include raindrop number size distribution, collection efficiency and raindrop terminal velocity, remain un-known and hence make the large uncertainties of BWSC (Wang et al., 55 2010). Seinfeld and Pandis (2016) proposed that collection efficiency (Brownian diffusion, directional interception, inertial impaction, thermophoresis and diffusion electrophoresis) is critical in the below-cloud scavenging process. Coarse particles (aerosol particle sizes  $d_p$  ranging from 2-20  $\mu\text{m}$ ) are easily scavenged by inertial impaction. Fine particles ( $d_p < 0.2 \mu\text{m}$ ) can be removed by Brownian diffusion. However, accumulation mode aerosols ( $0.2 \mu\text{m} < d_p < 2 \mu\text{m}$ ) are neither efficiently scavenged by Brownian diffusion nor by directional interception or inertial impaction, and this particle size range is called the “Greenfield gap” (Slinn, 1984). Recently, Bae et al. (2010) added phoretic and electric charging effects to the collection efficiency assessment and found that the BWSCs increase by up to 20 times in the 0.2-3  $\mu\text{m}$  particle size range. Wang et al. (2014c) also improved the understanding of the electrical effects of the collision efficiency, which is also assumed to be a major source of uncertainty but is always ignored in theoretical estimations. It also improved the BWSC estimation by an order of magnitude. The raindrop number size distribution and raindrop terminal velocity are both represented by empirical 60 mathematical functions, and these factors are non-negligible. In order to minimize the computational burden, the calculation of BWSCs in most global and regional-scale models are expressed as the product of rain intensity multiplied by the collection efficiency, where the later is simplified as a constant or calculated based on the work of Slinn (Bae et al., 2010; Slinn, 1984). This simplification may undoubtedly bring into large uncertainties and make the simulated wet deposition within a factor of two ranges of the observations, which is significant larger compared with the 30% bias of the prediction of air pollutants evaluation (Vet et al., 2014; Zhu et al., 2018). 70

Over the past few decades, a lot of wet scavenging coefficient (WSC) field measurements have been the focus of a large number of studies (Andronache, 2004b; Jylhä, 1991; Laakso et al., 2003; Okita et al., 1996; Wang et al., 2014c; Xu et al., 2017). In their field measurements, Okita et al. (1996) used the precipitation intensity, cloud-base height and the ratio between the sulfate concentration in aerosols of air mass and in rainwater to estimate the WSC, and this method is widely applied in most 75 field measurements at present (Andronache, 2004b; Yamagata et al., 2009). However, this method cannot distinguish the below cloud part from the whole wet scavenging process, which is important to the parameterization scheme in CTMs. Xu et al. (2017) adopted sequential sampling and estimated the BWSCs of various soluble inorganic ions using the washout fraction concentration. In addition, the BWSCs display a strong dependency on the aerosol particle size distribution. Laakso et al. (2003) indicated that the BWSCs could be calculated by the aerosol particle number concentrations for various size



80 ranges both before and after rain. This size-resolved method was also applied in Lanzhou (Zhao et al., 2015), Huang  
Mountain (Wang et al., 2014c), China, Southern Finland (Andronache et al., 2006) and India (Chate et al., 2003). In general,  
both methods are widespread for the estimation of WSCs/BWSCs but few were focused on the differences among these  
methods.

85 In this study, we compare the WSCs/BWSCs estimated from original and updated observational methods with the  
theoretical and model calculations under the same conditions to perform a multimethod evaluation to describe its  
characteristics. First, we introduce the basic circumstances of the data collected with multiple observation instruments. Then,  
we present the various methods, compare the results and discuss the discrepancies among the different results. Finally, we  
evaluate the effect of below-cloud scavenging on aerosol concentrations and wet depositions based on multimethod  
techniques.

## 90 **2 Methods and Data**

### **2.1 Sampling site and measurement data**

In North China, precipitations were mainly concentrated in summer (more than 80%) but rare in autumn and winter (Xu et al., 2008; Gao et al., 2015; Chen et al., 2013; Han et al., 2019). However, the air pollution events were usually occurred in autumn and winter in North China Plain (NCP). Here we select a typical rainfall event moving from northwest to southeast in winter of Beijing (a typical air polluted city in NCP) to study the characteristic of BWSC and its implications to aerosol scavenging. The sampling site is situated on top of the two-floor building of the Institute of Atmospheric Physics (IAP, 39°58'28"N, 116°22'1"E), located between the north 3<sup>rd</sup> and 4<sup>th</sup> Ring Road of Beijing. The site is a typical urban site and is 1 km away from the main road to the north and east, near residential buildings to the south and a park to the west, and the pollution at this site is mainly from traffic and domestic sources (Sun et al., 2015). The selected rainfall case lasts from 6:56 AM on November 20<sup>th</sup> to 1:18 AM on November 21<sup>st</sup>, 2016, which is during the wintertime Atmospheric Pollution and Human Health-Beijing (APHH-Beijing) campaign of 2016 (Shi et al., 2018). Thus, comprehensive measurements of air pollutants and simulations of pollution mechanisms are available for our use to investigate the wet scavenging process. Fig. s1 shows the radar base reflectivity with echo coverage over the urban area of Beijing from 11:54 AM on November 20<sup>th</sup>, and gradually moving from northwest to southeast. The total recorded amount of rainfall is 5.2 mm, and the rainfall is more concentrated on the evening of November 20<sup>th</sup> (beginning at 16:29 PM on November 20<sup>th</sup>) during this event. Notably, the rainfall is nonuniform across Beijing (Song et al., 2015). For example, the recorded rainfall in the southern suburban area of Beijing is approximately 7 mm according to the Beijing Meteorological Administration, and this rainfall end at approximately 9:00 AM on November 21<sup>st</sup>. In this study, the precipitation chemistry and aerosol components sampling processes occur at the exact same time at the APHH-Beijing measurement site.

110 An automatic wet-only sequential rainfall sampler is deployed to obtain rainfall samples with 1 mm increments in one precipitation event. Four anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$  and  $\text{F}^-$ ) and five cations ( $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) in these samples

are measured by ion chromatography (IC, Dionex 600, USA). The VWA concentrations of the major soluble inorganic ions, i.e.  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  (hereafter, SNA) in this rainfall are 35.8, 48.7 and 17.5  $\text{mg L}^{-1}$ , respectively, and much higher than the VWA concentrations in the winter of 2016 (8.3, 9.5 and 4.1  $\text{mg L}^{-1}$ , respectively) and in previous studies in Beijing (6.3, 9.1 and 4.9  $\text{mg L}^{-1}$  in Pan et al. (2012, 2013) and 6.2, 7.9 and 4.6  $\text{mg L}^{-1}$  in Xu et al.(2017) of summer). An ambient ion monitor-ion chromatograph (AIM-IC) developed by URG Corp., Chapel Hill, NC and Dionex Inc., Sunnyvale, CA, is used to measure the  $\text{PM}_{2.5}$  composition. The time resolution is 60 min. A detailed description of the measured concentration in the rainfall and aerosols can be found in Xu et al. (2017).

Thirty meters away from the sampling site, a scanning mobility particle sizer (SMPS) is deployed to observe the particle number size distribution with a 5-min time resolution. The SMPS is used to measure particle number concentration from 14 to 740 nm. A detailed description of the SMPS and methods can be found in Du et al. (2017).

A single-particle aerosol mass spectrometer (SPAMS) can accurately characterize aerosol particles containing various chemical compositions with diameters ranging from 0.2 to 2.5  $\mu\text{m}$ . It's deployed during the measuring time in China National Environmental Monitoring Centre (CNEMC), which is located in the northeast, 8 km away from the IAP sampling site. This site is a typical suburban site and mainly affected by residential source. More detailed fundamentals of the SPAMS and description can be found in Li et al. (2011), Lin et al. (2017) and Cheng et al. (2018). Size-resolved airborne  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  are the main focuses in this study, and the time resolution is 1 hour. In the meantime, a polarization optical particle counter (POPC) is also deployed to obtain coarse particle (0.4-10.35  $\mu\text{m}$ ) size distribution at the IAP sampling site, and time resolution is 5-min. Detailed description and settings can be found in Pan et al.(2016), Pan et al.(2017), Tian et al. (2018) and Pan et al.(2019).

## 2.2 Methods

### 2.2.1 Theoretical basis

Seinfeld and Pandis (2016) proposed the following basic equation of variation of the particle number concentration  $N(d_p)$ :

$$\frac{dN(d_p)}{dt} = -K(d_p)N(d_p) \quad (1)$$

This equation considers that there is no chemical reaction or emission, and wet scavenging is an exponential process.  $d_p$  is the diameter of the aerosol particle, and  $K(d_p)$  is the size-resolved BWSC obtained by the following equation:

$$K(d_p) = \int_0^\infty \frac{\pi}{4} D_p^2 U_t(D_p) E(D_p, d_p) N(D_p) dD_p \quad (2)$$

where  $D_p$  is the raindrop diameter.  $U_t(D_p)$  and  $N(D_p)$  are the falling terminal velocity and concentration of raindrops, respectively. There are two approaches for describing  $U_t(D_p)$ : an empirical formula and a physically based formula. Many expressions have been employed for various raindrop diameter ranges. In addition, there are still no available mathematical

functions that can accurately characterize the natural raindrop size spectra, and exponential, gamma and lognormal distributions are still used to represent  $N(D_p)$  (Wang et al., 2010). Marshall and Palmer (1948) proposed the M-P distribution of raindrop size distribution, which is mostly applied to calculations of BWSCs.  $E(D_p, d_p)$  is the collision efficiency of raindrops and aerosol particles, which, in most studies, mainly involves Brownian diffusion, interception and inertial impaction due to dimensional analysis without accounting for thermophoresis, diffusiophoresis and electric charges (Slinn, 1984; Wang et al., 2010). An extensive number of studies have realized that using only the three main mechanisms results in underestimation of the collision efficiency, and the contributions from the other mechanisms were added in these studies (Andronache, 2004c; Andronache et al., 2006; Bae et al., 2010). Assuming that a certain size aerosol particle can be captured by raindrops of any size,  $K(d_p)$  can be calculated theoretically when the falling terminal velocity, raindrop size distribution and collision efficiency are given. In Wang et al. (2014c)'s study, they added thermophoresis, diffusiophoresis and electric charges to the quantitative calculation, and we considered this updated to be the theory's result.

### 2.2.2 Observational method

In addition to the theoretical calculation, field observations are also critical for estimating BWSCs. One approach is based on the change in the number concentration of aerosols (called **OI** in this study). When rainfall occurs from  $t_0$  to  $t_1$ , Eq (1) can be integrated as follows:

$$K(d_p) = \frac{1}{t_1 - t_0} \ln \left[ \frac{N_0(d_p)}{N_1(d_p)} \right] \quad (3)$$

where  $N_0(d_p)$  and  $N_1(d_p)$  are the measured aerosol particle number concentrations before the rain occurs ( $t_0$ ) and after the rain ends ( $t_1$ ), respectively (Laakso et al., 2003).

In addition, Andronache (2004b) proposed that the WSC can be estimated by the bulk model based on the aerosol mass balance within a certain bulk, which assumes that there is a box with a horizontal area  $A$  and vertical height  $h$  above the observation site. The aerosol flux  $F$  on the surface per unit time and area is defined as the following equation:

$$F = K \times M \quad (4)$$

where  $K$  is the WSC and  $M$  is the mass of the aerosols in the given box.  $M$  can be described as follows:

$$M = C_a \times A \times h \quad (5)$$

where  $C_a$  is the average aerosol concentration in the box.

In addition,  $F$  can also be characterized by the following expression:

$$F = C_p \times P \times A \quad (6)$$

where  $C_p$  is the aerosol concentration in the precipitation collected at the measurement site,  $P$  is the precipitation intensity, and  $A$  is the horizontal area for the assumed box. And the wet deposition  $D_{ep}$  in a certain time  $\Delta t$  can be expressed as:

$$D_{ep} = C_p \times P \times \Delta t = K \times C_a \times h \times \Delta t \quad (7)$$

And  $K$  becomes the following expression:

$$K = \frac{C_p}{C_a} \times \frac{P}{h} \quad (8)$$

where  $C_p$  and  $C_a$  are the paired aerosol concentrations in the precipitation and aerosol, respectively, during rainfall (Okita et al., 1996). In addition, Andronache (2004b) pointed out that the aerosol concentration in the vertical profile should be considered and updated Eq (8) as follows:

$$K = \frac{C_p}{C_a(0) \times f} \times \frac{P}{h} \quad (9)$$

where  $C_a(0)$  is the aerosol concentration at the surface,  $h$  is the cloud-base height during rainfall, and

$f = \frac{\sum_{z=0}^{z=h} C_a(z)}{C_a(0)} \times h'(z) / \sum_{z=0}^{z=h} h'(z)$  is the vertical distribution factor of aerosols. Among these variables,  $C_a(z)$  are the aerosol

concentrations at the  $z$ -level height, respectively, and  $h'(z)$  is the depth of the layers in the vertical direction. This approach is called O2.

Moreover, most studies have mentioned that the prevailing wind in Beijing can efficiently reduce the aerosol concentrations (Chan and Yao, 2008; Gonzalez and Aristizabal, 2012). In previous studies by Xu et al. (2017), the north and northwest winds have been recognized as the clear streams to scavenge aerosols in situ, and the effects of clean wind is also considered in this study. In addition, with the help of the 1 mm increments sequential rainfall sampling, Xu et al. (2017) has found that the later increments maintained at a stable, low level, which can be separated into rainout process only. Similar with Eq (9), an updated below-cloud estimated method using  $C_{p, \text{below}}$  has been developed as Eq (10) and called as O2':

$$K = \frac{C_{p, \text{below}}}{C_a'(0) \times f} \times \frac{P}{h} \quad (10)$$

where  $C_{p, \text{below}}$  is the washout concentration that have been eliminated the rainwater concentrations in each increment and

$C_a'(0)$  is the aerosol concentration at the surface that considered the eliminated effects of north and north-west wind.

### 2.2.3 Modeling calculation

In this study, a three-dimensional regional model, the Nested Air Quality Prediction Modeling System (NAQPMS) was adopted to calculate the aerosol scavenging coefficient. The NAQPMS, developed by IAP, is a fully modularized chemical transport model describing regional and urban-scale air pollution (Wang et al., 2001). The meteorological condition is driven by Weather Research and Forecasting (WRF) model. The NAQPMS consists of modules used for horizontal and vertical advection (Walcek and Aleksic, 1998), diffusion (Byun and Dennis, 1995), dry and wet deposition (Zhang et al., 2003; Stockwell et al., 1990), gaseous phase, aqueous phase, and heterogeneous atmospheric chemical reactions (Zaveri and Peters, 1999; Stockwell et al., 1990; Li et al., 2012). Carbon-Bond Mechanism Z (CBM-Z) and aerosol thermodynamic equilibrium partition model (ISORROPIA1.7) have been used to calculate the gas and inorganic aerosol process. The cloud-process and aqueous chemistry module from Community Multi-scale Air Quality (CMAQ) modeling system v4.7 have been coupled in model by Ge et al. (2014). More details can be found in Li et al. (2016, 2017a). The NAQPMS has been widely used in prediction of acid rain, dust and secondary pollutions and can also reproduce well the physical and chemical evolution of reactive pollutants by solving the mass balance equations in terrain-following coordinates (Chen et al., 2019; Yang et al., 2019). It has been applied in Ministry of Ecology and Environment and local Environmental Protection Bureau such as Beijing, Shanghai, Guangzhou and Nanjing, etc. The NAQPMS also made great contribution to air quality assurance during the major activities (Wang et al., 2001; Wang et al., 2014d; Wu et al., 2010).

The below-cloud scavenging module from Comprehensive Air Quality Model with Extensions (CAMx) v4.42 was used to calculate the below-cloud wet scavenging process and the wet scavenging coefficient was briefly described as follows (Environ, 2005):

$$K = \frac{4.2 \times 10^{-7} \times E \times P}{d_p} \quad (11)$$

where  $d_p$  is the mean rain drop size and related to precipitation intensity. The collision efficiency  $E$  is a function of aerosol particle size and mainly considers Brownian diffusion, interception and inertial impaction. NAQPMS used in this study assumed SNA resides in fine mode size range (0.1-2.5  $\mu\text{m}$ ) and the geometric mean diameter of 0.5  $\mu\text{m}$  was used in the calculation of  $E$ .

To briefly describe these methods, Table 1 lists the formulas. The theoretical estimated scavenging coefficients are labeled T. The field observations estimated by Eq (3) and (9) are labeled O1 and O2, respectively. The updated estimated method by Eq (10) is labeled as O2'. The modeling results are labeled M, and these results are compared with different methods in section 3.

### 3 Results and Discussion

#### 3.1 Impacts of below-cloud wet scavenging on aerosols

In this case, the total precipitation amount was relatively low, but the precipitation duration was long. SNA represented the majority of the ions in the rainwater, accounting for 73% of the total and their temporal variations are shown in Fig. 1. The precipitation duration is marked with the blue frame. In the early stage, marked with light blue stripes, the precipitation duration was long and the precipitation intensity was weak. In the later period, from 16:29 PM on November 20<sup>th</sup> to 1:18 AM on November 21<sup>st</sup>, the precipitation began to strengthen and is marked with the blue shading. Before this event, a severe haze occurred which exceeded the National Ambient Air Quality Standard (NAAQS, 75  $\mu\text{g m}^{-3}$ ) (Shi et al., 2018). When rain occurred, both the aerosols in the air and the SNA concentration in the rainwater gradually decreased, especially during the later stage. It's clearly visible in Fig. 1 that all aerosol concentrations on the rainy day were much lower than the hourly averaged aerosol concentrations during the APHH-Beijing campaign, especially during the precipitation time indicating the below-cloud scavenging impacts. Following the rain, SNA reached relatively stable and low values.  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  decreased from 50.1, 70.6 and 25.3  $\text{mg L}^{-1}$  to 28.5, 25.2 and 10.3  $\text{mg L}^{-1}$  (or a reduction of 43.2, 64.3 and 59.5%) in the rainwater. Accordingly, aerosol nitrate, sulfate and ammonium decreased from 13.8, 8.3 and 8.4  $\mu\text{g m}^{-3}$  to 1.2, 2.2 and 0.1  $\mu\text{g m}^{-3}$  in the air (decreased by more than 6  $\mu\text{g m}^{-3}$ ).

The time series and averaged spectrum distribution of particle number size distributions measured by POPC, SPAMS and SMPS are shown in Fig. 2. With the help of three instruments, the size distributions cover a rather wide range, from 0.014 to 10.35  $\mu\text{m}$ . The spectrum distribution exhibited unimodal distributions peaked in the size range of 20-90 nm. The spectrum distribution for SPAMS of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  both showed particularly high consistency in terms of variation patterns, magnitude and particle size distribution (Lang et al., 2016). And for POPC, the trend was also in consistent well with the coarse size of SPAMS. As shown from Fig. 2(a), for POPC and SMPS, the number concentration did not immediately decrease due to relatively weak precipitation intensity before 16:29 PM on November 20<sup>th</sup>. And in the later period, the number concentration decreased sharply and remained at a low level. It agreed well with the radar echo and precipitation intensity during this rain event. In order to investigate the BWSC, 16:29 PM on November 20<sup>th</sup> is taken as the before the rain occurs time in calculating the O1 and it will not repeat in following sections.

### 3.2 Multimethod comparison of BWSCs

For further analysis, the estimated BWSCs based on multiple methods were compared and shown in Fig. 3. As for the observational methods, e.g., O1, O2 and O2', there is no significant difference in the range of magnitude between them. The observed O1 by SMPS, which cover the range of Aitken and accumulation mode aerosols (0.014-0.74  $\mu\text{m}$ ), are much lower than the other two measurements (0.2-2.5  $\mu\text{m}$  for SPAMS and 0.4-10.35  $\mu\text{m}$  for POPC, respectively). The observed BWSCs by original O2 are larger than the updated O2' method. However, O2' ( $5.7 \times 10^{-5}$ ,  $8.9 \times 10^{-5}$  and  $5.4 \times 10^{-5} \text{ s}^{-1}$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ ) is much closer to the results of O1 ( $\sim 10^{-5} \text{ s}^{-1}$  for particle size in the range of 0.014-10.35  $\mu\text{m}$ ). Since O1 is based on the variations of the aerosol numbers below the cloud, it may be more suitable for the estimation of the BWSCs. It also indicates that the updated O2' is much more reasonable than the original O2 for estimation of BWSCs of various chemical species. In contrast, the T's BWSC as  $1.9 \times 10^{-6} \text{ s}^{-1}$  has an order of magnitude lower than the observational results. Considering the

255 effects of thermophoresis, diffusiophoresis and electric charges, there is a wider range of three orders of magnitude ( $10^{-6}$ - $10^{-4}$   $s^{-1}$ ) (Wang et al., 2010; Wang et al., 2014c). In addition, the BWSC for **M** ( $3.2 \times 10^{-6} s^{-1}$ ) is also one order of magnitude lower than the field measurements. The low BWSC in CTMs can explain the underestimation of simulated wet deposition, which is mainly thought caused by chemical process, modeled precipitation and emission in previous studies (Wang et al., 2008; Ge et al., 2011). Thus, the observed **O1** and **O2'** may revise the **T** and **M** results in the future.

260 To further compare the BWSCs based on the particle size, the results in this study are compared with those of previous studies in Fig. 4. The size-resolved BWSCs of 0.014-0.74, 0.2-2.5 and 0.7-10.35  $\mu m$  are the total number concentration by SMPS, SPAMS and POPC, respectively, which are within a certain range ( $1.81 \times 10^{-5}$ - $8.53 \times 10^{-5} s^{-1}$ ). At approximately 0.2  $\mu m$  (the lower limit detection of the multicomponent analysis), the **O1** of 0.014-0.74 and 0.2-2.5  $\mu m$  results have a gap that mainly originates from the use of different experimental instruments and their detection limits. However, the estimated results for larger sizes ( $d_p > 3 \mu m$ ) by POPC have great fluctuation, mainly due to less number concentrations ( $< 2 cm^{-3}$ ) and were considered as unreliable in this work. The BWSC from **O1** showed a slowly decreasing trend in 0.014-0.2  $\mu m$  and a significant increasing trend as  $d_p > 0.2 \mu m$  in this study, which is similar with the results of Huang Mountain (Wang et al., 2014c) and southern Finland (Laakso et al., 2003). Besides, the BWSC from **O2'** for SNA are similar to the results of **O1** in 2.5  $\mu m$ . Although there is a different trend with that reported in Lanzhou (Zhao et al., 2015) before 0.6  $\mu m$ , both studies exhibited an increasing trend after 0.6  $\mu m$ . The difference of BWSCs from **O1** in each sites may due to the measuring conditions (Wang et al., 2010). However, compared to the **T**, this difference is very small as shown in Fig. 4. Different from 270 the observational results, the theoretical results show a strong dependence on the particle size with obvious decreasing trend ( $d_p < 1 \mu m$ ) and quickly increasing trend ( $d_p > 1 \mu m$ ). As Seinfeld and Pandis (2016) mentioned, Brownian diffusion and inertial impaction are the principal mechanisms affecting collection efficiency with  $d_p$  smaller than 0.2  $\mu m$  and larger than 2.5  $\mu m$ , respectively. Theoretical estimation can effectively characterize the observed BWSC of aerosols in these two ranges. 275 For the “Greenfield gap”, there is large difference between the BWSC from **O1** and **T** with the later is one order of magnitude lower. One reason is that all the influencing mechanisms still have not been fully considered and understood (Seinfeld, 2016), another reason is the existing ideal assumptions in derivation, such as no chemical reactions or emissions in the scavenging process; Ignored irregular surface of the aerosols and hygroscopic growth will increase the concentration of particles and then influence the scavenging efficiency (Wang et al., 2014c). Other extensive explanation is that the 280 turbulent flow fluctuation, evaporation and breakup of raindrops are also important but neglected processes (Wang et al., 2010).

### 3.3 The parameterization of BWSCs

To discuss the uncertainties of the BWSC underestimation by theoretical calculations in different rainfall events, nine rain events at the same sampling site in summer of 2014 (by **O2'**) have also been included. As shown in Fig. 5, a strong

285 relationship between the BWSCs and precipitation intensity obeys exponential power distribution both in summer of 2014  
 and the rainfall event in winter of APHH-Beijing campaign in Beijing with the coefficients of determination for SNA are  
 over than 0.68. Since the estimated BWSCs for SNA based on O1 and O2' in this event are in line with previous studies in  
 summer, it indicated that the wet scavenging rule and regression fitting formulas are also universal in Beijing not only in  
 summer but also in winter. In fact, this exponential power relationship has been confirmed in previous studies (Jylhä,  
 290 1991;Okita et al., 1996;Andronache, 2004a;Wang et al., 2014a;Wang et al., 2014b;Xu et al., 2017):

$$K = a \times P^b \quad (12)$$

where parameter  $b$  represents the change rate of BWSCs along with  $P$ , while  $a$  is equal to the WSCs when the  $P = 1$   
 mm/h. Both  $a$  and  $b$  relate to chemical species and aerosols particle size.

For the further comparison, Fig.6 displays the parameterization of  $a$  and  $b$  in the exponential power relationship for  
 295 BWSCs with the precipitation intensity by multimethod, i.e., theoretical method and field measurement methods. For the  
 theory calculation, the parameter  $a$  varies from a relatively wide range of  $2.8 \times 10^{-8}$ - $6.7 \times 10^{-5} \text{ s}^{-1}$  and BWSCs also have a  
 wide range of 3-4 orders of magnitude for given precipitation intensities (Andronache, 2003;Wang et al., 2014b). Similar  
 with the multimethod comparison of estimated BWSCs in this rainfall event of APHH-Beijing campaign, parameterization  
 for BWSCs obtained by O2/O2' shows higher magnitude of variations with the precipitation intensity with all of the straight  
 300 line lie above the upper range of the T. It indicates recent theory calculated BWSCs has an obvious underestimation not only  
 in a rainfall event but also in the parameterization of large number of rainfall events with different precipitation intensities  
 and need revised or updated by the field measurement estimation.

### 3.4 Impacts and implications

To investigate the impacts of the wet scavenging on aerosol concentrations in the air and the wet depositions in rainfall,  
 305 multimethod-estimated BWSCs included in Table 2 were used to rebuild the aerosol concentrations and wet depositions after  
 one hour of rainfall event. Assuming the aerosol concentrations in the air are only influenced by the wet scavenging during  
 rainfall event, its variation should be followed by Eq (13) according to Seinfeld and Pandis (2016):

$$\frac{dC_a}{dt} = -KC_a \quad (13)$$

$$C_a = C_{a0} e^{-Kt} \quad (14)$$

310 Variations of aerosol concentration can be resolved as Eq (14), in which the  $K$  is a constant BWSC. Where,  $K$  and  $t$   
 are the BWSCs and the scavenging time, and  $C_{a0}$  is the original aerosol concentration before the rainfall. In this study, the  
 $C_{a0}$  have been observed as 17.6, 9.8, 9.7 and  $74.7 \mu\text{g m}^{-3}$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{PM}_{2.5}$ , respectively. After one hour of  
 the wet scavenging by rainfall, the concentration of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{PM}_{2.5}$  decreased to 14.3, 7.6, 7.9 and  $65.2 \mu\text{g m}^{-3}$   
 respectively. Previous studies have confirmed that the exponential power distribution between the WSCs and precipitation



315 intensity as Eq (12). And the size-resolved BWSC are accumulated for calculating the total BWSC for PM<sub>2.5</sub>. As it is shown  
in Table 2, the calculated aerosol concentrations using **T** and **M** BWSCs performed the obvious overestimation of PM<sub>2.5</sub>  
concentrations with the bias from 2.3 to 9  $\mu\text{g m}^{-3}$ , while showed similar with the observation for **O1** and **O2'** BWSCs (bias <  
1  $\mu\text{g m}^{-3}$ ). It should be noted that the magnitude of BWSCs in the range of  $10^{-5}$ - $10^{-4}$  perform the better-calculated aerosol  
320 concentrations than that the lower range. Wet deposition has also been reconstructed according to Eq (7), with the  
precipitation intensity setting as 0.17 mm/h, and the column height considering as 3 km. The **Normalized Mean Bias (NMB)**  
for the below-cloud wet depositions of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> are -28% and -33%, while for SO<sub>4</sub><sup>2-</sup> is -49% according to the BWSCs  
in this study shown in Table 2.

Overall, the **O1** and updated **O2'** field observation results can effectively characterize the below-cloud scavenging ability  
whereas **T** and **M** have obvious deviation. Therefore, the field measurements are needed to compensate for the defects in the  
325 theoretical and modeling calculations that provides room to make further progress in wet deposition numerical simulation.

#### 4 Conclusions

An evaluation of below-cloud wet scavenging ability is first conducted based on field measurements, and accompanied with  
the theoretical estimation and modeling calculation. The averaged BWSCs obtained by field measurements are similar to  
each other of  $10^{-5} \text{ s}^{-1}$  and there exists strong exponential power relationship between BWSCs and precipitation intensity.  
330 Theoretical estimations coincide well with the observed BWSCs of aerosols with the  $d_p$  in ranges of smaller than 0.2  $\mu\text{m}$   
and larger than 2.5  $\mu\text{m}$ , but are one magnitude lower than observed BWSCs within 0.2-2.5  $\mu\text{m}$ . In the form of exponential  
power distribution of BWSCs with precipitation intensity, the upper range of theoretical results is also lower than the  
measurement estimation. Thus, the underestimation of BWSC through theoretical method has been confirmed not only in  
APHH-Beijing campaign but also in all rainfall events in summer of 2014. These theoretical values are usually applied in  
335 CTMs with simplified scheme and accordingly the model calculations show lower BWSCs. It may explain the  
underprediction of the wet deposition both in global and in regional models of polluted regions. Field measurements are  
currently required to compensate for the theoretical and modeling calculations and to construct a more reasonable and  
suitable simulation scheme to improve the wet deposition simulation, especially in polluted regions.

#### Competing interests

340 The authors declare that they have no conflict of interest.

#### Author contribution

DX, BG and ZW designed the whole structure of this work, XC performed the modeling calculation, YS, NC, ML, XP, ZM and YP prepared the SMPS data, the POPC data and the SPAMS data, respectively. DX performed the sequential sampling of rainwater, analyzed the data. DX and BG prepared the manuscript with contributions from all-authors.

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**Table 1. List of multimethod calculations for the BWSCs**

	Formula	Reference	Symbol
Theory	$K(d_p) = \int_0^\infty \frac{\pi}{4} D_p^2 U_i(D_p) E(D_p, d_p) N(D_p) dD_p$	Seinfeld and Pandis (2016), Wang et al. (2014c)	T
Field Observation	$K(d_p) = \frac{1}{t_1 - t_0} \ln \left[ \frac{N_0(d_p)}{N_1(d_p)} \right]$	Laakso et al. (2003)	O1
	$K = \frac{C_p}{C_a(0) \times f} \times \frac{P}{h}$	Andronache (2004b)	O2
	$K = \frac{C_{p,below}}{C_a(0) \times f} \times \frac{P}{h}$	Xu et al. (2017)	O2'
Modeling calculation	$K = \frac{4.2 \times 10^{-7} \times E \times P}{d_p}$	Wang et al. (2001)	M

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**Table 2. The observed aerosol concentrations before/after one-hour rainfall, wet depositions after one-hour rainfall, and parameters of the exponential power fittings, WSCs, rebuilt aerosol concentrations and wet depositions after one hour compare with multimethod.**

Source	Species	$K = aP^b$		BWSC (s <sup>-1</sup> )	Aerosol concentration ( $\mu\text{g m}^{-3}$ )	Wet deposition ( $\text{mg m}^{-2}$ )	Supplementary information
		$a$	$b$				
	NO <sub>3</sub> <sup>-</sup>				17.6/14.3	3.2	
	SO <sub>4</sub> <sup>2-</sup>				9.8/7.6	5.9	Observed aerosol concentrations before/after one-hour rainfall, and wet depositions after one-hour rainfall
	NH <sub>4</sub> <sup>+</sup>				9.7/7.9	1.8	
	PM <sub>2.5</sub>				74.7/65.2	-	
This study <sup>a</sup>	NO <sub>3</sub> <sup>-</sup>	2.5×10 <sup>-4</sup>	0.61	5.7×10 <sup>-5</sup>	14.4	2.3	
	SO <sub>4</sub> <sup>2-</sup>	7.6×10 <sup>-5</sup>	0.80	8.9×10 <sup>-5</sup>	7.1	3.0	Winter in Beijing, O2'
	NH <sub>4</sub> <sup>+</sup>	1.1×10 <sup>-4</sup>	0.52	5.4×10 <sup>-5</sup>	8.0	1.2	
This study <sup>a</sup>	PM	-	-	4.1×10 <sup>-5</sup>	64.6	-	0.014-10.35 $\mu\text{m}$ , winter in Beijing, O1
Okita et al. (1996) <sup>a*</sup>	SO <sub>4</sub> <sup>2-</sup>	1.38×10 <sup>-4</sup>	0.74	3.8×10 <sup>-5</sup>	8.6	0.6	Sado, Japan, winter of 1992, O2
Andronache (2004b) <sup>a*</sup>	SO <sub>4</sub> <sup>2-</sup>	4.0×10 <sup>-4</sup>	0.81	9.5×10 <sup>-4</sup>	7.0	3.3	AIRMoN in the USA, O2'
Yamagata et al. (2009) <sup>a*</sup>	SO <sub>4</sub> <sup>2-</sup>	-	-	9.8×10 <sup>-6</sup> - 2.5×10 <sup>-4</sup>	4.0-9.5	0.04-15.8	The Arctic, late spring of 2004, O2
Laakso et al. (2003) <sup>a</sup>	PM	-	-	9.1×10 <sup>-6</sup>	72.3	-	0.01-0.51 $\mu\text{m}$ , southern Finland, 6 years of observations, O1
Wang et al. (2014c) <sup>a</sup>	PM	4.2×10 <sup>-5</sup>	0.16	3.1×10 <sup>-5</sup>	66.7	-	0.01-1 $\mu\text{m}$ , Huang Mountain, 2001 summer, O1
Zhao et al. (2015) <sup>a</sup>	PM	-	-	3.2×10 <sup>-5</sup>	66.6	-	0.01-10 $\mu\text{m}$ , Lanzhou, 2012.9-2013.8, O1
This study <sup>b</sup>	PM <sub>2.5</sub>			1.9×10 <sup>-6</sup>	74.2	-	T

Wang et al. (2014b) <sup>b</sup>	PM <sub>2.5</sub>	$3.83 \times 10^{-7}$ - $6.89 \times 10^{-4}$	0.64-0.91	$10^{-8}$ - $10^{-2}$	-	-	
This study <sup>c</sup>	PM <sub>2.5</sub>	-	-	$3.2 \times 10^{-6}$	73.8	-	NAQPMS, <a href="#">M</a>
Luo et al. (2019) <sup>c</sup>	PM <sub>2.5</sub>	-	-	$2.8 \times 10^{-5}$	67.5	-	GEOS-Chem

<sup>a</sup> field observation, <sup>b</sup> theory and <sup>c</sup> modeling calculation

\* WSC and \*\* the wet scavenging effects on PM<sub>2.5</sub>

540 **Figure captions**

**Figure 1. Hourly average aerosol concentration during November 11<sup>th</sup> to December 11<sup>th</sup> (box, the data show the lowest, lowest 25 percentiles, median highest quartile, highest 75 percentiles, and highest value, respectively) and the rainy period on November 20<sup>th</sup> to 21<sup>st</sup> (red line and hollow circles) for (a) NO<sub>3</sub><sup>-</sup>, (b) SO<sub>4</sub><sup>2-</sup> and (c) NH<sub>4</sub><sup>+</sup>. The rainwater concentrations are shown as follows: (a) NO<sub>3</sub><sup>-</sup>, blue; (b) SO<sub>4</sub><sup>2-</sup>, red; and (c) NH<sub>4</sub><sup>+</sup>, orange (line and triangles). Time series of the NO<sub>3</sub><sup>-</sup> (blue), SO<sub>4</sub><sup>2-</sup> (red) and NH<sub>4</sub><sup>+</sup> (orange) concentrations in the rainfall (lines and triangles) and in the air (lines), and rainfall (d).**

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**Figure 2. Time series of particle number size distributions (a) are measured by POPC, SPAMS (take SO<sub>4</sub><sup>2-</sup> for example) and SMPS, respectively. The averaged spectrum distribution of number concentration during the APHH-Beijing campaign (a) for SMPS (purple line), POPC (green line) and NO<sub>3</sub><sup>-</sup> (blue line), SO<sub>4</sub><sup>2-</sup> (red line) and NH<sub>4</sub><sup>+</sup> (orange line) by SPAMS.**

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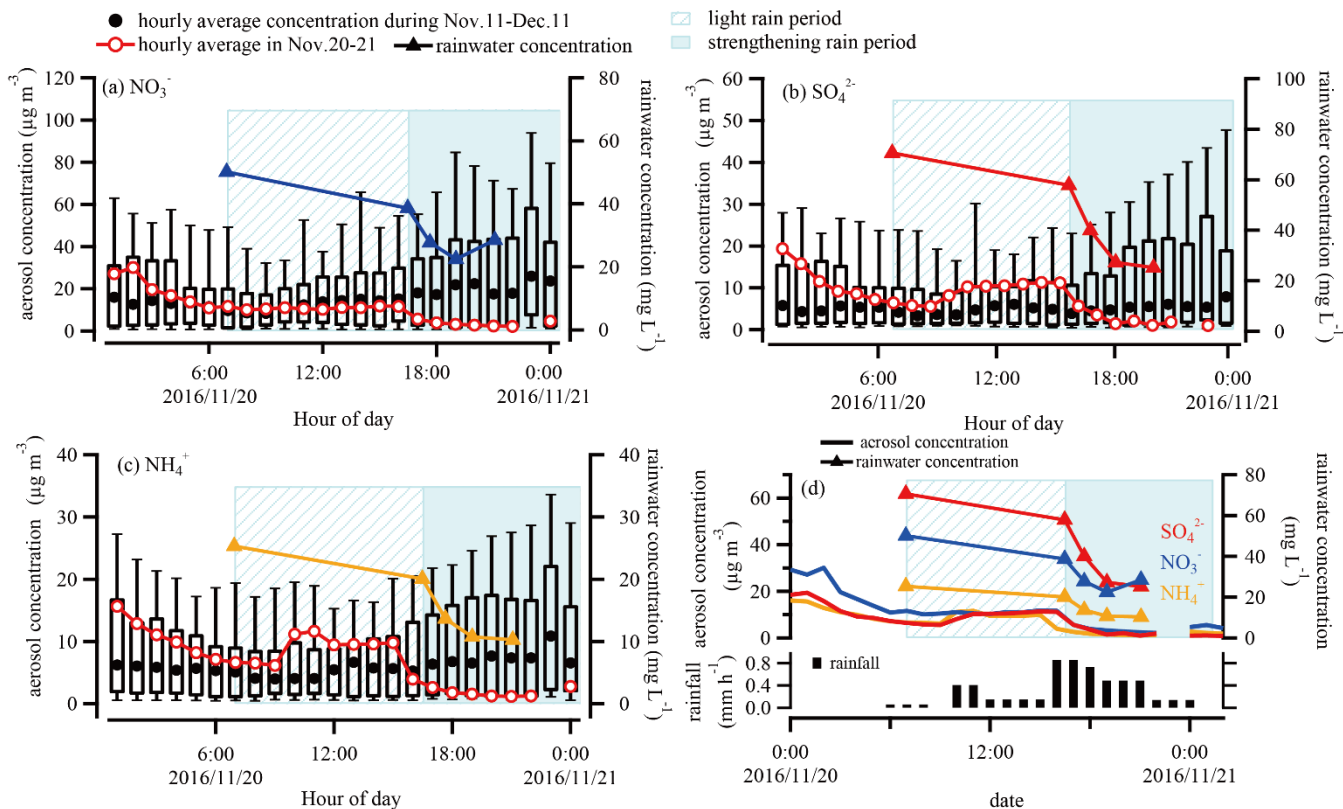
**Figure 3. Box and whisker plots of the multimethod estimation of the BWSCs. The top and bottom of the boxes represent the 75<sup>th</sup> and 25<sup>th</sup> percentiles, and central lines mean the median BWSCs. The whiskers represent maximum and minimum BWSCs, respectively.**

**Figure 4. Multimethod estimation of the BWSCs and comparisons with previous studies.**

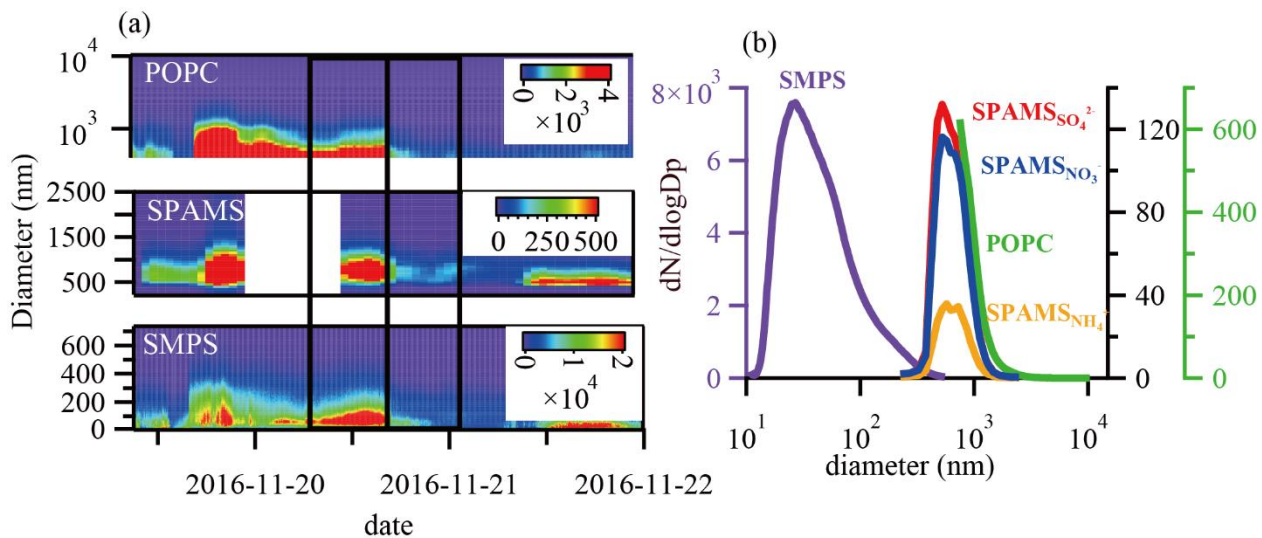
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**Figure 5. Scatter plots of the BWSCs and precipitation intensity for NO<sub>3</sub><sup>-</sup> (a), SO<sub>4</sub><sup>2-</sup> (b) and NH<sub>4</sub><sup>+</sup> (c) (black dots: O2' in summer by Xu et al. (2017), light blue triangle: O2' in this case, deep blue triangle: O1).**

**Figure 6. The parameterization of BWSCs with the rainfall intensities.**



560 **Figure 1. Hourly average aerosol concentration during November 11<sup>th</sup> to December 11<sup>th</sup> (box, the data show the lowest, lowest 25 percentiles, median highest quartile, highest 75 percentiles, and highest value, respectively) and the rainy period on November 20<sup>th</sup> to 21<sup>st</sup> (red line and hollow circles) for (a)  $\text{NO}_3^-$ , (b)  $\text{SO}_4^{2-}$  and (c)  $\text{NH}_4^+$ . The rainwater concentrations are shown as follows: (a)  $\text{NO}_3^-$ , blue; (b)  $\text{SO}_4^{2-}$ , red; and (c)  $\text{NH}_4^+$ , orange (line and triangles). Time series of the  $\text{NO}_3^-$  (blue),  $\text{SO}_4^{2-}$  (red) and  $\text{NH}_4^+$  (orange) concentrations in the rainfall (lines and triangles) and in the air, and rainfall (d).**



565 **Figure 2.** Time series of particle number size distributions (a) are measured by POPC, SPAMS (take SO<sub>4</sub><sup>2-</sup> for example) and SMPS, respectively. The averaged spectrum distribution of number concentration during the APHH-Beijing campaign (a) for SMPS (purple line), POPC (green line) and NO<sub>3</sub><sup>-</sup> (blue line), SO<sub>4</sub><sup>2-</sup> (red line) and NH<sub>4</sub><sup>+</sup> (orange line) by SPAMS.

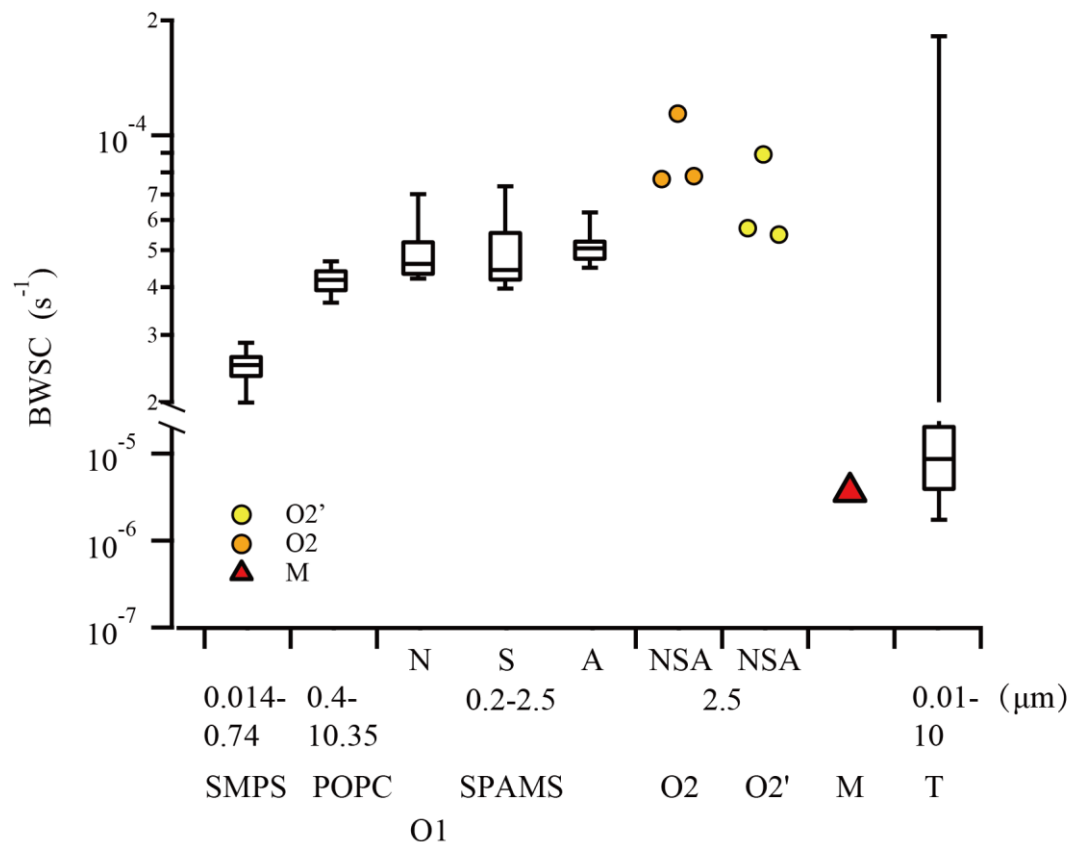


Figure 3. Box and whisker plots of the multimethod estimation of the BWSCs. The top and bottom of the boxes represent the 75<sup>th</sup> and 25<sup>th</sup> percentiles, and central lines mean the median BWSCs. The whiskers represent maximum and minimum BWSCs, respectively.

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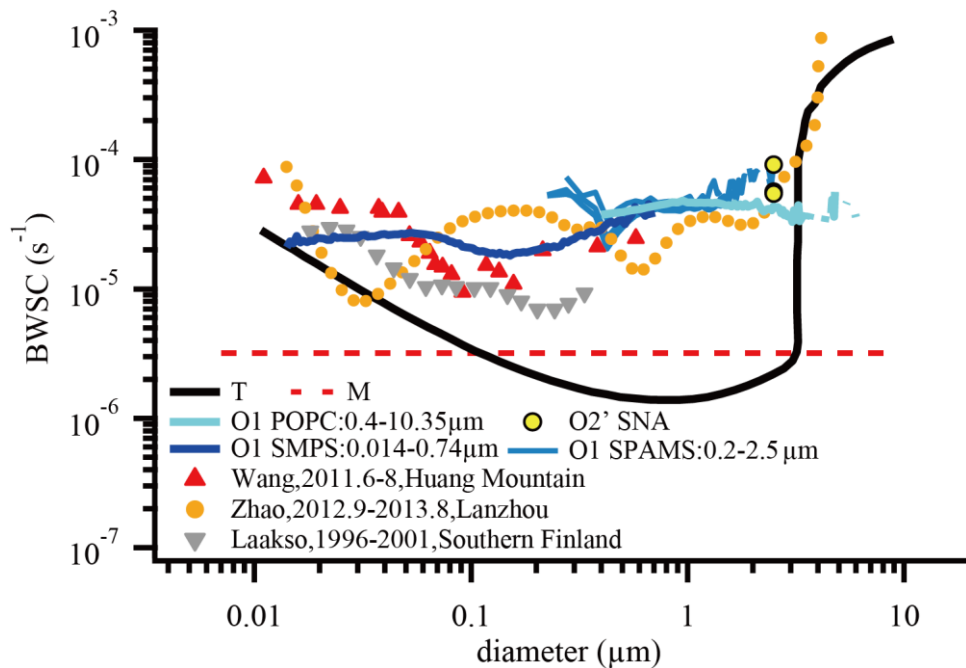
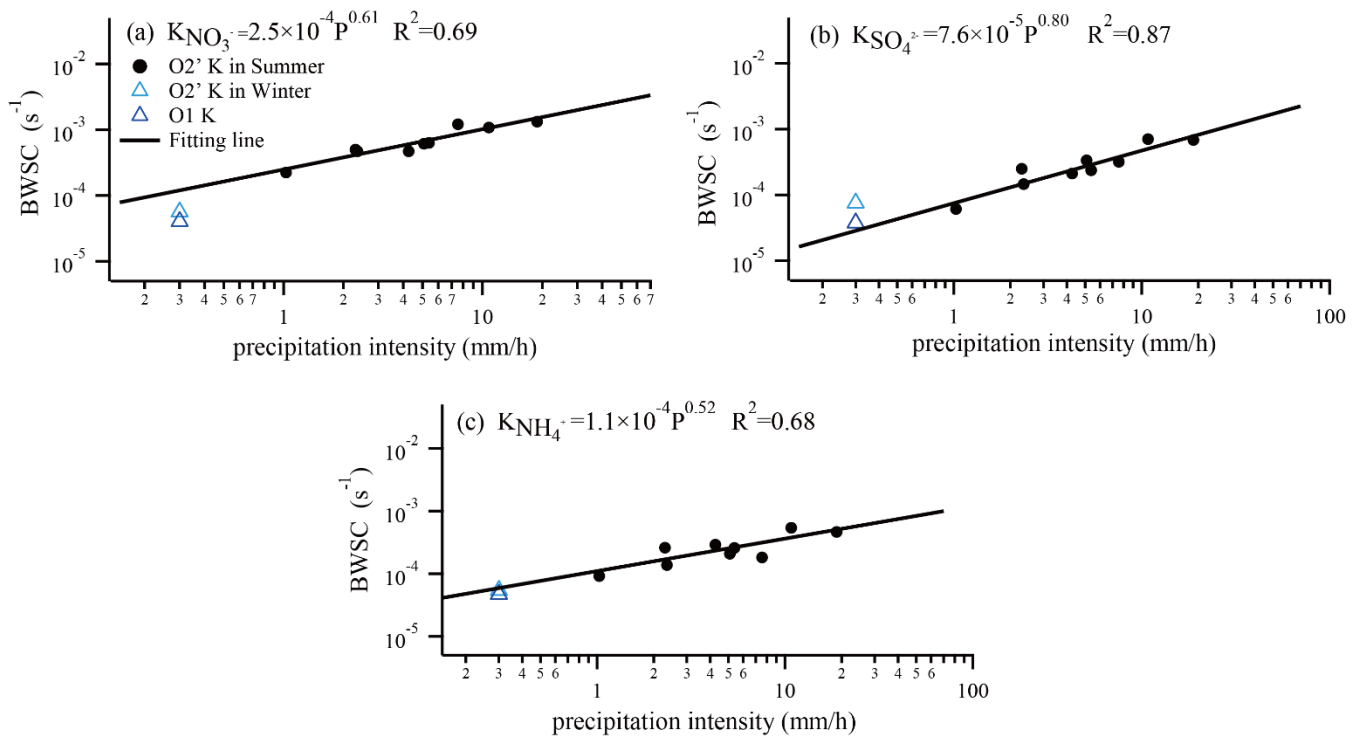


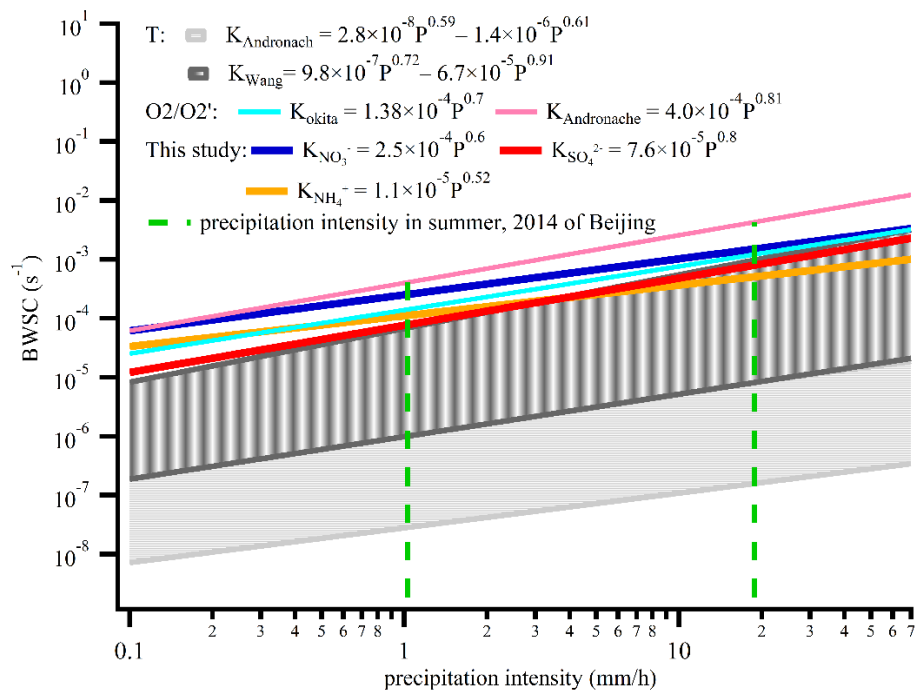
Figure 4. Multimethod estimation of the BWSCs and comparisons with previous studies.



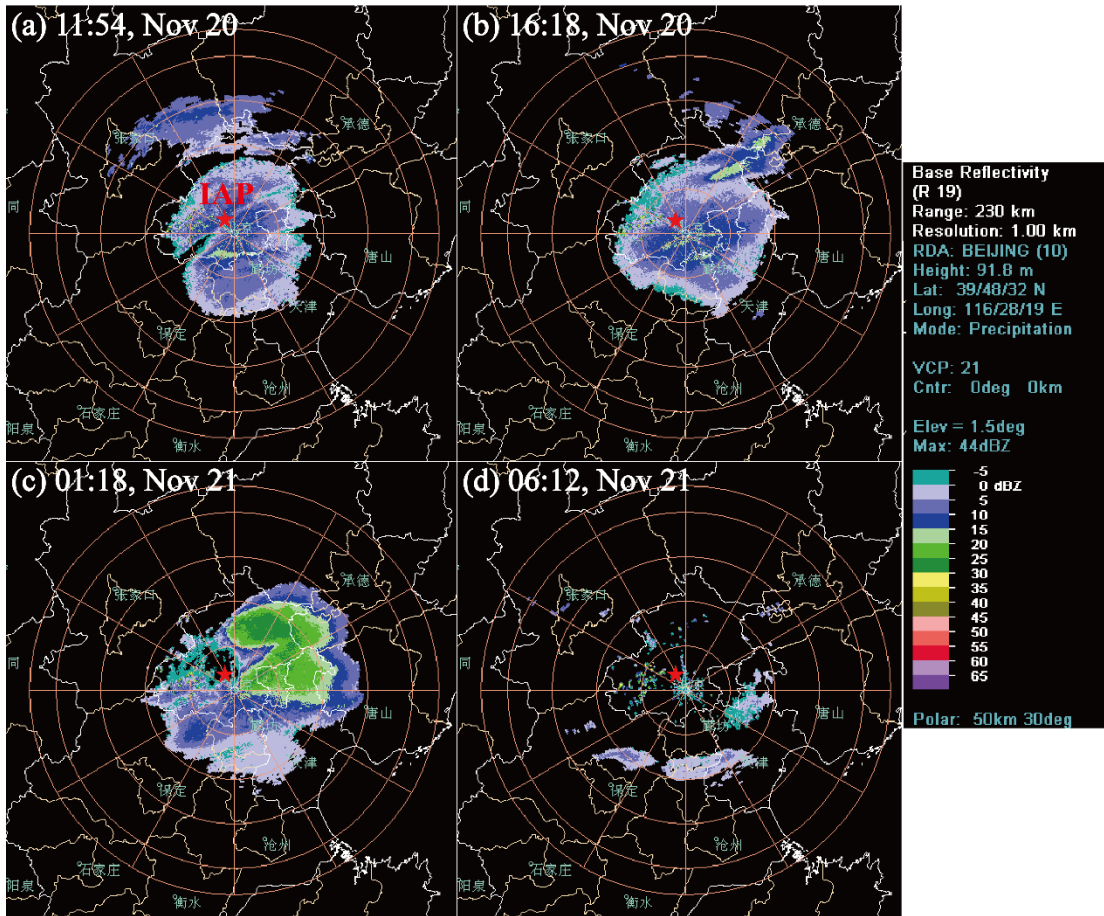


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Figure 5. Scatter plots of the BWSCs and precipitation intensity for  $NO_3^-$  (a),  $SO_4^{2-}$  (b) and  $NH_4^+$  (c) (black dots: O2' in summer by Xu et al. (2017), light blue triangle: O2' in this case, deep blue triangle: O1).



**Figure 6.** The parameterization of BWSCs with the rainfall intensities.



1

2 **Figure s1. The radar base reflectivity during the rainfall event (the sampling site at the IAP is**  
 3 **indicated by the red star).**

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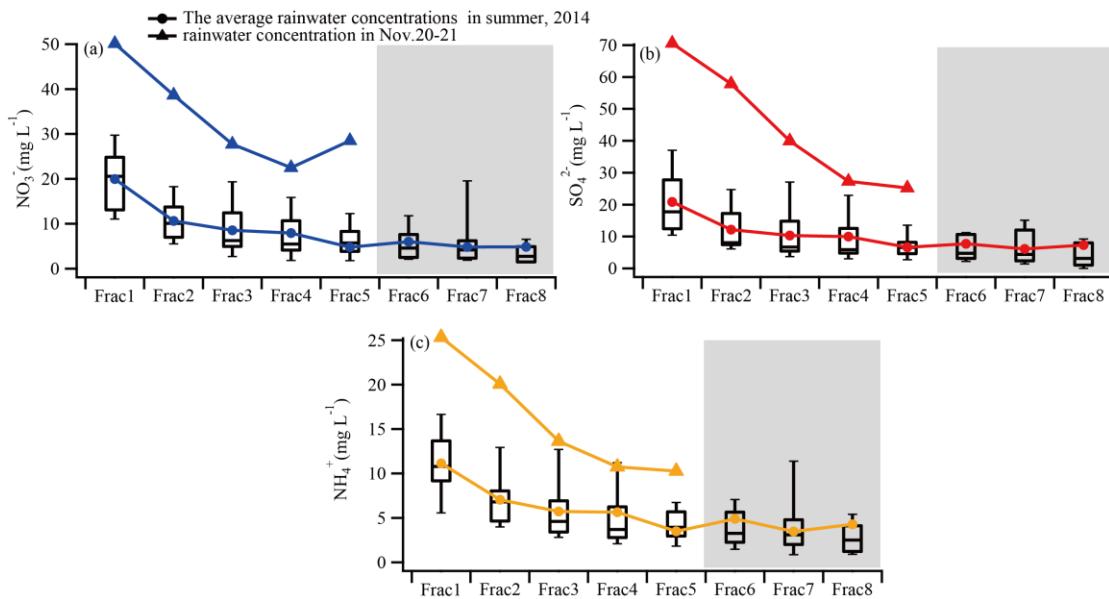
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11 The Figure. s2 shows the average concentrations of SNA in summer of 2014 (Box and whisker plot) and this  
 12 rain event in winter of APHH-Beijing campaign. The VWA concentrations are no more than 30, 40 and 15  
 13  $\text{mg L}^{-1}$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ , respectively, and decreased sharply during the beginning of rainfall and  
 14 remained at low levels during the event. The in-cloud scavenging process is considered as the median value  
 15 of the concentrations after accumulated precipitation exceeds 5 mm. These values were 2.75, 3.33, and 2.51  
 16  $\text{mg L}^{-1}$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ , respectively, in summer of 2014 (as shown in Figure. s2 marked in grey  
 17 shadow) (Xu et al., 2017).



18  
 19 **Figure s2. Evolution of the (a)  $\text{NO}_3^-$  (blue), (b)  $\text{SO}_4^{2-}$  (red) and (c)  $\text{NH}_4^+$  (orange) of precipitation during**  
 20 **summer 2014 and November 20<sup>th</sup> to 21<sup>st</sup> within different precipitation fractions of several sampled**  
 21 **precipitation events (The data show the lowest, lowest 25 percentiles, median highest quartile, highest**  
 22 **75 percentiles, and highest value, respectively).**

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26 **Table s1. The observation dataset list**

Observation instrument	Abbreviation	Introduction
		(time resolution and mainly measured material)
Ion Chromatography	IC	anions (SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> and F <sup>-</sup> ) and cations (NH <sub>4</sub> <sup>+</sup> , Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>2+</sup> and Mg <sup>2+</sup> ) in the rainfall samples
Ambient Ion Monitor-Ion Chromatograph	AIM-IC	60 min resolution, PM <sub>2.5</sub> concentrations
Scanning Mobility Particle Sizer	SMPS	5 min resolution, 14-740 nm particle number concentration
Single-particle Aerosol Mass Spectrometer	SPAMS	60 min resolution, 0.2-2.5 μm particle number concentration, mainly focus on NO <sub>3</sub> <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> and NH <sub>4</sub> <sup>+</sup>
Polarization Optical Particle Counter	POPC	5 min resolution, 0.4-10.35 μm particle number concentration

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Nomenclature			
$d_p$	aerosol particle diameter	$F$	aerosol flux
$D_p$	raindrop diameter	$D_{ep}$	wet deposition
$N(d_p)$	aerosol particle number concentration	$C_p$	concentration in the precipitation
$N(D_p)$	concentration of raindrops	$C_{a0}$	original aerosol concentration before the rainfall
$K(d_p)$	size-resolved below-cloud scavenging coefficient	$C_a(0)$	aerosol concentration at the surface
$K$	below-cloud scavenging coefficient	$C_a(z)$	aerosol concentration at height $z$
$U_i(D_p)$	falling velocity of raindrops	$C_{p, \text{below}}$	washout concentration
$E(D_p, d_p)$	collision efficiency	$P$	precipitation intensity
$A$	horizontal area of the given box	$h'(z)$	depth of the layers in the vertical direction
$h$	vertical height of the given box or cloud-base height	$f$	vertical distribution of aerosols
$M$	aerosol mass of the given box	$C'_a(0)$	aerosol concentration at the surface and eliminates effects of north and north-west wind
$C_a$	average aerosol concentration	BWSC	below-cloud wet scavenging coefficient
APHH-Beijing	Atmospheric Pollution and Human Health-Beijing	CTMs	Chemical Transport Models

MICS-Asia	Model Inter-Comparison Study for Asia	TF-HTAP	Hemispheric Transport of Atmospheric Pollutants
VWA	volume weighted averaged	NCP	North China Plain
IAP	Institute of Atmospheric Physics	IC	Ion Chromatography
AIM-IC	Ambient Ion Monitor-Ion Chromatograph	SMPS	Scanning Mobility Particle Sizer
SPAMS	Single-particle Aerosol Mass Spectrometer	POPC	Polarization Optical Particle Counter
SNA	$\text{NO}_3^-$ , $\text{SO}_4^{2-}$ and $\text{NH}_4^+$	NAQPMS	the Nested Air Quality Prediction Modeling System
NAAQS	the National Ambient Air Quality Standard	CAMx	Comprehensive Air Quality Model with Extensions
CMAQ	Community Multi-scale Air Quality modeling system	CNEMC	China National Environmental Monitoring Centre