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:

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

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 µm, 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 µm. NAQPMS used in this study assumed SNA resides in fine mode size range (0.1-2.5 µm) and the geometric mean diameter of 0.5 µm 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):

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

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 µm) and the geometric mean diameter of 0.5 µm 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 NAOPMS, 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 (ISORROPIAI1.7) have been used to calculated 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 75th and 25th 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 μ m), are much lower than the other two measurements. The observed BWSCs by original O2 are larger than the updated O2' method. However, O2' (5.7×10⁻⁵, 8.9×10⁻⁵ and 5.4×10⁻⁵ s⁻¹ for NO₃⁻, SO₄²⁻ and NH₄⁺) is much closer to the results of O1 (~10⁻⁵ s⁻¹ for particle size in the range of 0.014-10.35 μ 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.

Reference:

- Byun, D. W., Dennis, R.: Design artifacts in Eulerian air-quality models evaluation of the effects of layer thickness and vertical profile correction on surface ozone concentrations. Atmos. Environ. 29 (1), 105–126, 1995.
- Chatterjee, A., Jayaraman, A., Rao, T. N., Raha, S.: In-cloud and below-cloud scavenging of aerosol ionic species over a tropical rural atmosphere in India, J Atmos Chem, 66, 27-40, 10.1007/s10874-011-9190-5, 2010.
- Chen. X. S., Yang, W. Y., Wang, Z. F., Li, J., Hu, M., An, J. L., Wu, Q. Z., Wang, Z., Chen, H. S., Wei, Y., Du, H. Y., Wang, D. W.: Improving new particle formation simulation by coupling a volatility-basis set (VBS) organic aerosol module in NAQPMS+APM. Atmos. Environ. 204, 1–11, 2019.
- Chen, Y. Y., Tian, H. Z., Yang, D. Y., Zou, B. D., Lu, H. F., Lin, A. G.: Correlation Between Acidic Materials and Acid Deposition in Beijing During 1997-2011. Environmental Science (in Chinese), 34(5), 1958-1963, 2013.
- ENVIRON.INC.: User's Guide Comperhensive Air Quality Model with Extension (CAMx) Version 4.42. 101 Rowland Way, Suite 220; Novato, California, 2005.
- Ge, B. Z., Wang, Z. F., Xu, X. B., Wu, J. B., Yu, X. L., Li, J.: Wet deposition of acidifying substances in different regions of China and the rest of East Asia: Modeling with updated NAQPMS, Environ Pollut, 187, 10-21, 2014.
- Ge, B. Z., Wang, Z F., Gbaguidi, A. E., and Zhang, Q.: Source Identification of Acid Rain Arising over Northeast China: Observed Evidence and Model Simulation, Aerosol Air Qual Res, 16, 1366-1377, 10.4209/aaqr.2015.05.0294, 2016.
- Ge, B. Z., Xu, X. B., Ma, Z. Q., Pan, X. L., Wang, Z., Lin, W. L., Ouyang, B., Xu, D. H., Lee, J., Zheng, M., Ji, D. S., Sun, Y. L., Dong, H. B., Squires, F. A., Fu, P. Q., Wang, Z. F.: Role of ammonia on the feedback between AWC and inorganic aerosol formation during heavy pollution in NCP. Earth and Space Science, 6, 1675–1693, 2019.
- Han, L. H., Wang, H. M., Xiang, X., Zhang, H. L., Yan, H. T., Cheng, S.Y., Wand, H.Y., Zheng, A. H., Guo, J. H.: The characteristics of precipitation and its impact on fine particles at a representative region in Beijing. China Environmental

Science (in Chinese), 39(9): 3635-3646, 2019.

- Li, J., Wang, Z., Zhuang, G., Luo, G., Sun, Y. L., Wang, Q.: Mixing of Asian mineral dust with anthropogenic pollutants over East Asia: a model cast study of a superduststorm in March 2010. Atmos. Chem. Phys. 12, 7591–7607, 2012.
- Li, J., Du, H.Y., Wang, Z.F., Sun, Y.L., Yang, W.Y., Li, J.J., Tang, X., Fu, P.Q.: Rapid formation of a severe regional winter haze episode over a megacity cluster on the North China Plain. Environ. Pollut. 223, 605–615, 2017.
- Stockwell, W.R., Middleton, P., Chang, J.S., Tang, X.Y.: The second generation regional acid deposition model chemical mechanism for regional air quality modeling. J. Geophys. Res. Atmos. 95 (D10), 16343–16367, 1990.
- Vet, R., Artz, R. S., Carou, S., Shaw, M., Ro, C. U., Aas, W., Baker, A., Bowersox, V. C., Dentener, F., Galy-Lacaux, C., Hou, A., Pienaar, J. J., Gillett, R., Forti, M. C., Gromov, S., Hara, H., Khodzher, T., Mahowald, N. M., Nickovic, S., Rao, P. S. P., and Reid, N. W.: A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmos Environ, 93, 3-100, 2014.
- Walcek, C. J., Aleksic, N. M.: A simple but accurate mass conservative, peak-preserving, mixing ratio bounded advection algorithm with Fortran code. Atmos. Environ. 32 (22), 3863–3880, 1998.
- Wang, X. H.; Zhang, L. M.; Moran, M. D.,: Uncertainty assessment of current size-resolved parameterizations for below-cloud particle scavenging by rain, Atmos. Chem. Phys., 10, 5685-5705, 10.5194/acp-10-5685-2010, 2010.
- Wang, X. H.; Zhang, L. M.; Moran, M. D.,: Development of a new semi-empirical parameterization for below-cloud scavenging of size-resolved aerosol particles by both rain and snow, Geosci Model Dev, 7, 799-819, 10.5194/gmd-7-799-2014, 2014b.
- Wang, Z. F., Maeda, T., Hayashi, M., Hsiao, L. F., and Liu, K. Y.: A nested air quality prediction modeling system for urban and regional scales: Application for high-ozone episode in Taiwan, Water Air Soil Poll, 130, 391-396, 2001.

Wang, Z. F., Li, J., Wang, Z., Yang, W. Y., Tang, X., Ge, B. Z., Yan, P. Z., Zhu, L. L.,

Chen, X. S., Chen, H. S., Wand, W., Li, J. J., Liu, B., Wang, X. Y., Wand, W., Zhao, Y, L., Lu, N., Su, D. B.: Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control, Science China: Earth Sciences, 57, 3-13, 2014d.

- Wu, Q. Z., Wang, Z. F., Xu, W. S., Huang, J. P., Gbaguidi, A. E.: Multi-model simulation of PM10 during the 2008 Beijing Olympic Games: Effectiveness of emission restrictions. J Environ Sci (in Chinese), 30, 1739–1748, 2010.
- Xiao, X., Chen, M. X., Gao, F., Wang, Y. C.: A thermodynamic mechanism analysis on enhancement or dissipation of convective systems from the mountains under weak synoptic forcing. Chinese Journal of Atmospheric Sciences (in Chinese), 39(1), 100-124, 2015.
- Xu, D. H., Ge, B. Z., Wang, Z. F., Sun, Y. L., Chen, Y., Ji, D. S., Yang, T., Ma, Z. Q., Cheng, N. L., Hao, J. Q., and Yao, X. F.: Below-cloud wet scavenging of soluble inorganic ions by rain in Beijing during the summer of 2014, Environ Pollut, 230, 963-973, https://doi.org/10.1016/j.envpol.2017.07.033, 2017.
- Yang, W. Y., Li, J., Wang, W. G., Li, J. L., Ge, M. F., Sun, Y. L., Chen. X. S., Ge, B. Z., Tong S, R., Wang, Q. Q., Wang, Z. F.: Investigating secondary organic aerosol formation pathways in China during 2014. Atmos. Environ. 213, 133–147, 2019.
- Zaveri, R. A., Peters, L.K.: A new lumped structure photochemical mechanism for large-scale applications. J. Geophys. Res. Atmos. 104 (D23), 30387–30415, 1998.
- Zhang, L., Brook, J.R., Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models. Atmos. Chem. Phys. 3 (6), 2067–2082, 2003.