Response to Reviewers’ comments

We are thankful to the two reviewers for their thoughtful and constructive comments that help us improve the manuscript substantially. We have revised the manuscript accordingly. Listed below is our point-to-point response in blue to each comment that was offered by the reviewers.

Response to Reviewer #1

General comments:

The authors presented 4-year optical properties of atmospheric aerosols collected over two size fractions (PM1 and PM10). The analysis includes scattering and absorbing coefficients, along with derived values of SSA, AAE, and RFE. The manuscript is submitted as a ‘Measurement Report”. Four-year continuous datasets from a highly populated city like Beijing, China, is beneficial.

R: The authors thank the reviewer’s positive comments.

Major comments:

According to the authors, the policies implemented to improve the air quality in Beijing are the main reason for the reduction in optical properties magnitude from 2018-2021. The policies effectively reduced the absorbing aerosols, which reduced the absorption coefficients. Does the same reduction happen in the scattering coefficient also? The absorbing aerosols like black carbon, BrC, and Dust also scatter in nature. So, for a complete picture, it will be better to see the fate of scattering coefficients over the years.

R: Thank you for the reviewer’s advice. Since the two-year scattering coefficients has been published (Hu et al., 2021), therefore, this manuscript focused on optical properties other than scattering coefficients. The variation of aerosol scattering properties for submicron (PM_1) and sub-10 μm particles (PM_{10}) under dry conditions (RH <30%) in Beijing from 2018 to 2019 has been analyzed in our previous study (Hu et al., 2021). The results showed that σ_{sp} for PM_1 and PM_{10} showed the similar variation. σ_{sp} in 2019 decreased by approximately 18.4% for PM_{10}, and 16.7% for PM_1 compared with those in 2018. In addition, aerosol scattering coefficient is highly positively correlated with PM mass concentration. Decreasing $PM_{2.5}$ mass concentrations from 2018-2021 may reflect the variation of aerosol scattering coefficient to some extent. Based on our observed data, the annual mean of σ_{sp} was 160.4 Mm^{-1} in 2018, and it decreased by 19.6% (128.9 Mm^{-1}) in 2021, shown in the following Figure R1 which does not include in the revised manuscript.

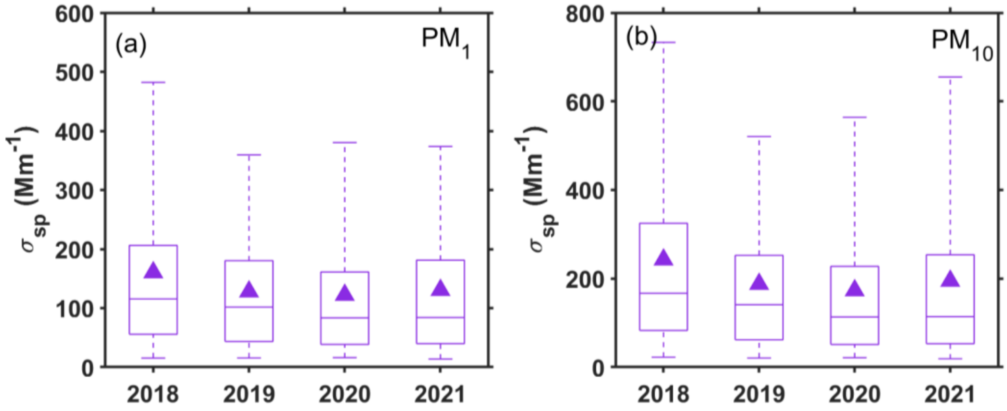

Figure R1. Annual variation of scattering coefficient (σ_{sp}) at 550 nm for (a) PM$_1$ and (b) PM$_{10}$.

We cited the results of aerosol scattering coefficient in the previous study (Hu et al., 2021), and added the sentences in the revised manuscript as follow:

"In fact, with the emission reduction and improvement of air quality, the aerosol scattering coefficient (σ_{sp}) for PM$_{10}$ and PM$_1$ also decreased in Beijing. Hu et al. (2021) revealed that σ_{sp} decreased by approximately 18.4% for PM$_{10}$, and 16.7% for PM$_1$ from 2018 to 2019 in Beijing." (Line 245-248 in revised manuscript)

Reference:

Hu, X., Sun, J., Xia, C., Shen, X., Zhang, Y., Zhang, X., and Zhang, S.: Simultaneous measurements of PM1 and PM10 aerosol scattering properties and their relationships in urban Beijing: A two-year observation, Sci. Total Environ., 770, 145215, 10.1016/j.scitotenv.2021.145215, 2021.

Figure 1 shows the overall reduction in PM$_{2.5}$. So, it is safe to assume that the PM$_1$ and PM$_{10}$ mass concentrations were also proportionally reduced. The general decrease in PM will automatically lessen the optical properties irrespective of the type of aerosol controlled.

R: We agree the reviewer's opinion. We did not express the meaning clearly in the original manuscript, and revised as the following:

"Carbonaceous aerosol, especially black carbon, is closely related to aerosol absorption (Yang et al., 2009). A continuous decrease in σ_{ab} was consistent with the continuous reduction of black carbon concentration observed in Beijing in previous studies (Ji et al., 2019; Sun et al., 2022), which was mainly related to significantly reduced primary emissions caused by effective air pollution control measures in recent years (Xia et al., 2020)." (Line 237-242 in revised manuscript)

Reference:

Yang, M., Howell, S. G., Zhuang, J., and Huebert, B. J.: Attribution of aerosol light absorption to black carbon, brown carbon, and dust in China – interpretations of atmospheric measurements during EAST-AIRE, Atmos. Chem. Phys., 9, 2035–2050, 2009.

Ji, D., Gao, W., Maenhaut, W., He, J., Wang, Z., Li, J., Du, W., Wang, L., Sun, Y., Xin, J., Hu, B., and Wang, Y.: Impact of air pollution control measures and regional transport on carbonaceous aerosols in fine particulate matter in urban Beijing, China: insights gained from long-term measurement, Atmos. Chem. Phys., 19, 8569-8590, 10.5194/acp-19-8569-2019, 2019.

Sun, J., Wang, Z., Zhou, W., Xie, C., Wu, C., Chen, C., Han, T., Wang, Q., Li, Z., Li, J., Fu, P., Wang, Z., and Sun, Y.: Measurement report: Long-term changes in black carbon and aerosol optical properties from 2012 to 2020 in Beijing, China, Atmos. Chem. Phys., 22, 561-575, 10.5194/acp-22-561-2022, 2022.

Xia, Y., Wu, Y., Huang, R. J., Xia, X., Tang, J., Wang, M., Li, J., Wang, C., Zhou, C., and Zhang, R.: Variation in black carbon concentration and aerosol optical properties in Beijing: Role of emission control and meteorological transport variability, Chemosphere, 254, 126849, 10.1016/j.chemosphere.2020.126849, 2020.

Minor comments:

Line 88-89 – The term σ_{ap} is not defined.
R: Corrected.

Figure 5 – Use a different color for the backscattering ratio (b) frequency line.
R: Changed as suggestion.

The author contribution list is incomplete.
R: We completed the authors' contribution list in the revised manuscript.

Response to Reviewer #2
Review of "Measurement Report: Rapid decline of aerosol absorption coefficient and aerosol optical properties effects on radiative forcing in urban areas of Beijing from 2018 to 2021" by Xinyao Hu and coauthors.

This manuscript presents 4 years of aerosol optical properties in PM_{10} and PM_1 size fractions at an urban site in Beijing. The dataset is interesting, and the manuscript is well written. I'm not sure if it is completely appropriate for ACP since the scope of the results is quite limited.

R: The authors thank reviewer's comments. Providing reliable observations of aerosol optical properties are crucial for quantifying the aerosol radiative forcing on climate. The 4-year data presented in this study provide critical optical parameters for radiative forcing assessment within two size ranges and are also helpful for evaluating the effectiveness of clean air action. We revised the manuscript carefully according to reviewers' suggestions and comments, hopefully, it is suitable to the scope of the ACP.

General comment: The authors have to make clear that talking about trends require long-term data, and apply statistical tests to verify if the decrease observed is statistically significant or not (most likely it is, but need verification). Another concern is related to atmospheric conditions. Changes in meteorological conditions over time, as well as the COVID-19 restrictions could be behind the observed “trend”. However, none of these causes are discussed in the manuscript. There have been many publications about the decrease of pollutants in 2020 during COVID-19 lockdown, but this is not even mentioned in this manuscript. Furthermore, meteorological variables such as temperature, wind, precipitation, should be proven to be similar from year to year, so the results can be more conclusive.

R: Thanks for the reviewer’s suggestion. The trends of the monthly σ_{ab} was analyzed using the Mann–Kendall (MK) method at a 95% confidence level, with the statistical parameters listed in Table S1. The Mann–Kendall trend test supported that the decrease in σ_{ab} from 2018 to 2021 was significant. We agree that at least 10 years of data are needed for climatic trend analysis. 4-year data presented in the manuscript can’t represent the aerosol optical properties climatic trend. We just discuss the variation of σ_{ab} in 4 years in the revised manuscript as the following.:

“The annual mean σ_{ab} at 550 nm of PM_{10} and PM_1 decreased by 55.0% and 53.5%, respectively, from 2018 to 2021. The Mann–Kendall trend test of monthly mean σ_{ab} for PM_1 and PM_{10} supported that the decrease in σ_{ab} for PM_1 and PM_{10} from 2018 to 2021 was significant (Table S1).” (Line 234-237 in revised manuscript)

Table. S1 Mann–Kendall trend test results ($p = 0.05$) for monthly mean value of σ_{ab} for PM_1 and PM_{10} from 2018 to 2021. Z is the standardized test statistic value.

	Trend	Z	sen's slope
PM_1 σ_{ab}	Decreasing trend	-5.8134	-0.24856
PM_{10} σ_{ab}	Decreasing trend	-5.643	-0.29905

As the reviewer’s suggestions, atmospheric conditions also have the effect on aerosol optical properties. So, we calculated the annual mean of meteorological parameters from 2018 to 2021. The results show that pressure, wind speed, temperature, RH varied slightly, while accumulated precipitation increased in 2021 compared with other 3 years. On the other hand, correlation analysis was made between aerosol optical properties and meteorological parameters. The Pearson correlation coefficients (R) between σ_{ab} and meteorological parameters are lower 0.5, indicating that weak correlation ($R<0.5$) was found between σ_{ab} and meteorological parameters. This suggests that the meteorological parameters influence on aerosol optical properties is minor, which is confirmed by previous study (Gong et al., 2022).

We added the discussion in the revised manuscript as follow:

“Gong et al. (2022) demonstrated that the emission reduction dominated the variations of $PM_{2.5}$ mass concentration in Beijing from 2013 to 2020, and meteorology and emission reduction contributed 7% and 63.2% of decreases, respectively.” (Line 231-233 in revised manuscript)

"Atmospheric conditions also have an effect on aerosol optical properties. The variations of meteorological parameters from 2018 to 2021 (Figure S4) showed that pressure, wind speed, temperature, and RH varied slightly, while accumulated precipitation increased in 2021 compared with the other 3 years. On the other hand, a correlation analysis was made between aerosol optical properties and meteorological parameters. The Pearson correlation coefficients (R) between σ_{ab} and meteorological parameters (Table S2) are lower than 0.5, indicating that a weak correlation ($R<0.5$) was found between σ_{ab} and meteorological parameters. This suggests that the meteorological parameters' influence on σ_{ab} is minor. Xia et al. (2020) revealed that the effect of emission reduction was the major reason for the decrease of BC in Beijing." (Line 248-257 in revised manuscript)

Table S2. Pearson correlation coefficient (R) between different aerosol optical properties and meteorological parameters (* Significant at $p < 0.05$).

	pressure	temperature	RH	precipitation	wind speed
PM$_1$ σ_{ab}	-0.10*	-0.05*	0.30*	-0.03*	-0.31*
PM$_{10}$ σ_{ab}	-0.10*	-0.05*	0.30*	-0.03*	-0.30*

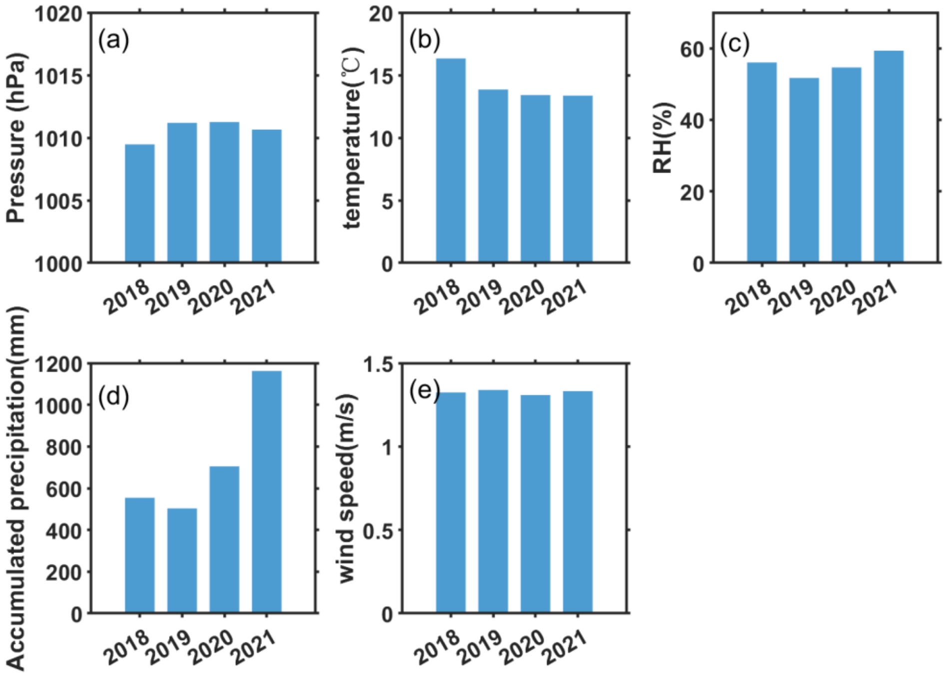

Figure S4. The annual variation of meteorological parameters from 2018 to 2021.

As the mentioned by the reviewer, our study period covered the period of the outbreak of COVID-19, which produced serious impacts in China. We added the discussion of the influence of COVID-19 in the revised manuscript.

“Compared with 2018, σ_{ab} in the winter of 2019, 2020 and 2021 decreased by 3.0%, 24.9% and 53.2%, respectively. In the winter of 2019, the lockdown of COVID-19 caused emission reduction from human activities in China (Tian et al., 2020; Le et al., 2020), however, the unexpected smallest reduction of σ_{ab} was observed in the winter of 2019 compared with the winter of 2020 and 2021. This is related to the fact that severe haze pollution still occurred in the North China Plain and BC concentrations rose unexpectedly during the lockdown period (Liu et al., 2021; Jia et al., 2021).” (Line 287-294 in revised manuscript)

Reference:

Gong, S., Zhang, L., Liu, C., Lu, S., Pan, W., and Zhang, Y.: Multi-scale analysis of the impacts of meteorology and emissions on PM(2.5) and O(3) trends at various regions in China from 2013 to 2020 2. Key weather elements and emissions, Sci Total Environ, 824, 153847, 10.1016/j.scitotenv.2022.153847, 2022.

Tian, H., Liu, Y., Li, Y., Wu, C. H., Chen, B., Kraemer, M. U. G., Li, B., Cai, J., Xu, B., Yang, Q., Wang, B., Yang, P., Cui, Y., Song, Y., Zheng, P., Wang, Q., Bjornstad, O. N., Yang, R., Grenfell, B. T., Pybus, O. G., and Dye, C.: An investigation of transmission control measures during the first 50 days of the COVID-19 epidemic in China, Science, 368, 638-642, 10.1126/science.abb6105, 2020.

Le, T., Wang, Y., Liu, L., Yang, J., Yung, Y. L., Li, G., and Seinfeld, J. H.: Unexpected air pollution with marked emission reductions during the COVID-19 outbreak in China, Science, 369, 702-706, 10.1126/science.abb7431, 2020.

Liu, Y., Wang, Y., Cao, Y., Yang, X., Zhang, T., Luan, M., Lyu, D., Hansen, A. D. A., Liu, B., and Zheng, M.: Impacts of COVID-19 on Black Carbon in Two Representative Regions in China: Insights Based on Online Measurement in Beijing and Tibet, Geophysical Research Letters, 48, 10.1029/2021gl092770, 2021.

Jia, M., Evangeliou, N., Eckhardt, S., Huang, X., Gao, J., Ding, A., and Stohl, A.: Black Carbon Emission Reduction Due to COVID-19 Lockdown in China, Geophys Res Lett, 48, e2021GL093243, 10.1029/2021GL093243, 2021.

L50: variation-> variable

R: Corrected.

L59: for reference in-situ SSA values refer better to in-situ techniques. Consider the following references which are more appropriate:

Laj et al. 2020, Atmos. Meas. Tech., 13, 4353–4392, 2020

Pandolfi et al. 2018, Atmos. Chem. Phys., 18, 7877–7911, 2018

R: Thanks for your advice. We revised the sentence and replaced the references as suggestions.

“Previous studies found that SSA values range from slightly less than 0.8 to almost purely scattering particles with SSA close to 1 at worldwide locations (Laj et al., 2020; Pandolfi et al., 2018), and higher SSA values indicate a tendency towards a cooling effect (Li et al., 2022a).” (Line 60-63 in revised manuscript)

Reference:

Laj, P., Bigi, A., Rose, C., Andrews, E., Lund Myhre, C., Collaud Coen, M., Lin, Y., Wiedensohler, A., Schulz, M., Ogren, J. A., Fiebig, M., Gliß, J., Mortier, A., Pandolfi, M., Petäja, T., Kim, S.-W., Aas, W., Putaud, J.-P., Mayol-Bracero, O., Keywood, M., Labrador, L., Aalto, P., Ahlberg, E., Alados Arboledas, L., Alastuey, A., Andrade, M., Artíñano, B., Ausmeel, S., Arsov, T., Asmi, E., Backman, J., Baltensperger, U., Bastian, S., Bath, O., Beukes, J. P., Brem, B. T., Bukowiecki, N., Conil, S., Couret, C., Day, D., Dayantolis, W., Degorska, A., Eleftheriadis, K., Fetfatzis, P., Favez, O., Flentje, H., Gini, M. I., Gregorič, A., Gysel-Beer, M., Hallar, A. G., Hand, J., Hoffer, A., Hueglin, C., Hooda, R. K., Hyvärinen, A., Kalapov, I., Kalivitis, N., Kasper-Giebl, A., Kim, J. E., Kouvarakis, G., Kranjc, I., Krejci, R., Kulmala, M., Labuschagne, C., Lee, H.-J., Lihavainen, H., Lin, N.-H., Löschau, G., Luoma, K., Marinoni, A., Martins Dos Santos, S., Meinhardt, F., Merkel, M., Metzger, J.-M., Mihalopoulos, N., Nguyen, N. A., Ondracek, J., Pérez, N., Perrone, M. R., Petit, J.-E., Picard, D., Pichon, J.-M., Pont, V., Prats, N., Prenni, A., Reisen, F., Romano, S., Sellegri, K., Sharma, S., Schauer, G., Sheridan, P., Sherman, J. P., Schütze, M., Schwerin, A., Sohmer, R., Sorribas, M., Steinbacher, M., Sun, J., Titos, G., Toczko, B., Tuch, T., Tulet, P., Tunved, P., Vakkari, V., Velarde, F., Velasquez, P., Villani, P., Vratolis, S., Wang, S.-H., Weinhold, K., Weller, R., Yela, M., Yus-Diez, J., Zdimal, V., Zieger, P., and Zikova, N.: A global analysis of climate-relevant aerosol properties retrieved from the network of Global Atmosphere Watch (GAW) near-surface observatories, Atmospheric Measurement Techniques, 13, 4353-4392, 10.5194/amt-13-4353-2020, 2020.

Pandolfi, M., Alados-Arboledas, L., Alastuey, A., Andrade, M., Angelov, C., Artiñano, B., Backman, J., Baltensperger, U., Bonasoni, P., Bukowiecki, N., Collaud Coen, M., Conil, S., Coz, E., Crenn, V., Dudoitis, V., Ealo, M., Eleftheriadis, K., Favez, O., Fetfatzis, P., Fiebig, M., Flentje, H., Ginot, P., Gysel, M., Henzing, B., Hoffer, A., Holubova Smejkalova, A., Kalapov, I., Kalivitis, N., Kouvarakis, G., Kristensson, A., Kulmala, M., Lihavainen, H., Lunder, C., Luoma, K., Lyamani, H., Marinoni, A., Mihalopoulos, N., Moerman, M., Nicolas, J., amp, apos, Dowd, C., Petäjä, T., Petit, J.-E., Pichon, J. M., Prokopciuk, N., Putaud, J.-P., Rodríguez, S., Sciare, J., Sellegri, K., Swietlicki, E., Titos, G., Tuch, T., Tunved, P., Ulevicius, V., Vaishya, A., Vana, M., Virkkula, A., Vratolis, S., Weingartner, E., Wiedensohler, A., and Laj, P.: A European aerosol phenomenology – 6: scattering properties of atmospheric aerosol particles from 28 ACTRIS sites, Atmospheric Chemistry and Physics, 18, 7877-7911, 10.5194/acp-18-7877-2018, 2018.

Li, J., Carlson, B. E., Yung, Y. L., Lv, D., Hansen, J., Penner, J. E., Liao, H., Ramaswamy, V., Kahn, R. A., Zhang, P., Dubovik, O., Ding, A., Lacis, A. A., Zhang, L., and Dong, Y.: Scattering and absorbing aerosols in the climate system, Nature Reviews Earth & Environment, 10.1038/s43017-022-00296-7, 2022a.

L54: What about the impact of backscatter fraction in the radiative forcing efficiency? That’s another important aerosol property to take into account.

R: Thanks for your advice. Backscatter fraction is a key variable to calculate the aerosol radiative forcing efficiency (RFE). We modified the sentence as:

"The backscatter fraction (b) describes how much aerosol particles scatter radiation in the backward hemisphere compared with the total scattering, which is a crucial variable for aerosol radiative forcing efficiency (RFE) calculations (Andrews et al., 2011; Sheridan and Ogren, 1999; Luoma et al., 2019). Previous studies found that the magnitude of RFE increases with increasing b (Shen et al., 2018). Typical values of b for the atmospheric aerosol at 550 nm were from approximately 0.05 to 0.20 (Titos et al., 2021)." (Line 63-69 in revised manuscript)

Reference:

Andrews, E., Ogren, J. A., Bonasoni, P., Marinoni, A., Cuevas, E., Rodríguez, S., Sun, J. Y., Jaffe, D. A., Fischer, E. V., Baltensperger, U., Weingartner, E., Coen, M. C., Sharma, S., Macdonald, A. M., Leaitch, W. R., Lin, N. H., Laj, P., Arsov, T., Kalapov, I., Jefferson, A., and Sheridan, P.: Climatology of aerosol radiative properties in the free troposphere, Atmos. Res., 102, 365-393, 10.1016/j.atmosres.2011.08.017, 2011.

Sheridan, P. J., and Ogren, J. A.: Observations of the vertical and regional variability of aerosol optical properties over central and eastern North America, J. Geophys. Res., 104, 16793-16805, 10.1029/1999jd900241, 1999.

Luoma, K., Virkkula, A., Aalto, P., Petäjä, T., and Kulmala, M.: Over a 10-year record of aerosol optical properties at SMEAR II, Atmos. Chem. Phys., 19, 11363-11382, 10.5194/acp-19-11363-2019, 2019.

Shen, Y., Virkkula, A., Ding, A., Wang, J., Chi, X., Nie, W., Qi, X., Huang, X., Liu, Q., Zheng, L., Xu, Z., Petäjä, T., Aalto, P. P., Fu, C., and Kulmala, M.: Aerosol optical properties at SORPES in Nanjing, east China, Atmos. Chem. Phys., 18, 5265-5292, 10.5194/acp-18-5265-2018, 2018.

Titos, G., Burgos, M. A., Zieger, P., Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Weingartner, E., Henzing, B., Luoma, K., O'Dowd, C., Wiedensohler, A., and Andrews, E.: A global study of hygroscopicity-driven light-scattering enhancement in the context of other in situ aerosol optical properties, Atmospheric Chemistry and Physics, 21, 13031-13050, 10.5194/acp-21-13031-2021, 2021.

L110: Measurements were performed at a single measurement site. Change the title to "… an urban area…", otherwise it is misleading.

R: Changed as suggested.

L112: together with the coordinates, indicate the altitude of the site.

R: We added the altitude of the site in the revised manuscript.

L114: This is not clear, measurements are taken at 53m agl?

R: We are sorry that we did not express this clearly. We revised the sentence as follow "The laboratory is on the roof of CAMS building and the measurements are taken at 53m above ground level" (Line 123-125 in the revised manuscript)

L125: A whole paragraph is dedicated to the TAP instrument, but nothing is said about the nephelometer. How often zero checks were done? How often was the instrument calibrated?

R: Thanks for your advice. We modified the title of section 2.2 and added some information about nephelometer as:

"The integrating nephelometer measured the scattering coefficient (σ_{sp}) (angular range of 7–170◦) and backscattering coefficient (σ_{bsp}) (angular range of 90–170◦) at 450, 550, and 700 nm. The scattering and backscattering coefficient were corrected for truncation and instrument non-idealities using the method described by Anderson and Ogren (1998). Details are given in Hu et al. (2021). To ensure the data's accuracy and reliability, the nephelometer was calibrated regularly using filtered ambient air using a HEPA filter and CO_2 with a purity of 99.999%. A zero-check was automatically performed once per hour to obtain a nephelometer background." (Line 136-143 in the revised manuscript)

Reference:

Anderson, T. L., and Ogren, J. A.: Determining Aerosol Radiative Properties Using the TSI 3563 Integrating Nephelometer, Aerosol Sci. Technol. , 29, 57-69, 10.1080/02786829808965551, 1998.

Hu, X., Sun, J., Xia, C., Shen, X., Zhang, Y., Zhang, X., and Zhang, S.: Simultaneous measurements of PM1 and PM10 aerosol scattering properties and their relationships in urban Beijing: A two-year observation, Sci. Total Environ., 770, 145215, 10.1016/j.scitotenv.2021.145215, 2021.

L178: Here it is introduced the backscatter coefficient. State that before when talking about the neph, and also in the introduction as an important variable for RFE calculations.

R: Thanks for the reviewer's suggestions. We added some information of scattering and backscattering coefficient in instruments and measurements parts, and the statement about the importance of backscatter fraction for RFE calculations in the introduction, as the following

"The integrating nephelometer measured scattering coefficient (σ_{sp}) (angular range of 7–170◦) and backscattering coefficient (σ_{bsp}) (angular range of 90–170◦) at 450, 550, 700 nm. The scattering and backscattering coefficient were corrected for truncation and instrument non-idealities using the method described by Anderson and Ogren (1998)." (Line 136-139 in the revised manuscript)

“The backscatter fraction (b) describes how much aerosol particles scatter radiation in the backward hemisphere compared with the total scattering, which is a crucial variable for aerosol radiative forcing efficiency (RFE) calculations (Andrews et al., 2011; Sheridan and Ogren, 1999; Luoma et al., 2019). Previous studies found that the magnitude of RFE increases with increasing b (Shen et al., 2018). Typical values of b for the atmospheric aerosol at 550 nm were from approximately 0.05 to 0.20 (Titos et al., 2021).” (Line 63-69 in revised manuscript)

Reference:

Anderson, T. L., and Ogren, J. A.: Determining Aerosol Radiative Properties Using the TSI 3563 Integrating Nephelometer, Aerosol Sci. Technol. , 29, 57-69, 10.1080/02786829808965551, 1998.

Andrews, E., Ogren, J. A., Bonasoni, P., Marinoni, A., Cuevas, E., Rodríguez, S., Sun, J. Y., Jaffe, D. A., Fischer, E. V., Baltensperger, U., Weingartner, E., Coen, M. C., Sharma, S., Macdonald, A. M., Leaitch, W. R., Lin, N. H., Laj, P., Arsov, T., Kalapov, I., Jefferson, A., and Sheridan, P.: Climatology of aerosol radiative properties in the free troposphere, Atmos. Res., 102, 365-393, 10.1016/j.atmosres.2011.08.017, 2011.

Sheridan, P. J., and Ogren, J. A.: Observations of the vertical and regional variability of aerosol optical properties over central and eastern North America, J. Geophys. Res., 104, 16793-16805, 10.1029/1999jd900241, 1999.

Luoma, K., Virkkula, A., Aalto, P., Petäjä, T., and Kulmala, M.: Over a 10-year record of aerosol optical properties at SMEAR II, Atmos. Chem. Phys., 19, 11363-11382, 10.5194/acp-19-11363-2019, 2019.

Shen, Y., Virkkula, A., Ding, A., Wang, J., Chi, X., Nie, W., Qi, X., Huang, X., Liu, Q., Zheng, L., Xu, Z., Petäjä, T., Aalto, P. P., Fu, C., and Kulmala, M.: Aerosol optical properties at SORPES in Nanjing, east China, Atmos. Chem. Phys., 18, 5265-5292, 10.5194/acp-18-5265-2018, 2018.

Titos, G., Burgos, M. A., Zieger, P., Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Weingartner, E., Henzing, B., Luoma, K., O'Dowd, C., Wiedensohler, A., and Andrews, E.: A global study of hygroscopicity-driven light-scattering enhancement in the context of other in situ aerosol optical properties, Atmospheric Chemistry and Physics, 21, 13031-13050, 10.5194/acp-21-13031-2021, 2021.

L180: Make it clear that the RFE calculations are referred to dry conditions (as the optical properties are measured) and do not refer to ambient conditions. Previous studies have demonstrated the influence of RH on the RFE (see for example Titos et al., 2021, Atmos. Chem. Phys., 21, 13031–13050, 2021, their figure 5).

R: We are sorry that we didn’t express clearly. We revised the manuscript following the reviewer’s suggestions:

“Note that RFE in this study was in a dry condition. As the backscatter fraction (b) and single scattering albedo are all RH-dependent, the RFE is also sensitive to RH (Fierz-Schmidhauser et al., 2010). Previous studies revealed that RFE increased as the elevating RH (Titos et al., 2021; Xia et al., 2023).” (Line 200-203 in the revised manuscript)

Reference:

Fierz-Schmidhauser, R., Zieger, P., Gysel, M., Kammermann, L., DeCarlo, P. F., Baltensperger, U., and Weingartner, E.: Measured and predicted aerosol light scattering enhancement factors at the high alpine site Jungfraujoch, Atmos. Chem. Phys., 10, 2319–2333, 2010.

Titos, G., Burgos, M. A., Zieger, P., Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Weingartner, E., Henzing, B., Luoma, K., O'Dowd, C., Wiedensohler, A., and Andrews, E.: A global study of hygroscopicity-driven light-scattering enhancement in the context of other in situ aerosol optical properties, Atmospheric Chemistry and Physics, 21, 13031-13050, 10.5194/acp-21-13031-2021, 2021.

Xia, C., Sun, J., Hu, X., Shen, X., Zhang, Y., Zhang, S., Wang, J., Liu, Q., Lu, J., Liu, S., and Zhang, X.: Effects of hygroscopicity on aerosol optical properties and direct radiative forcing in Beijing: Based on two-year observations, Sci Total Environ, 857, 159233, 10.1016/j.scitotenv.2022.159233, 2023.

L214: It doesn’t make sense to compare Beijing with Hyytiälä, of coarse values are higher at Beijing… Consider other studies performed in urban areas:

Lyamani et al. 2010, Atmos. Chem. Phys., 10, 239–254, 2010

Titos et al. 2012, JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 117, D04206, doi:10.1029/2011JD016671, 2012

Pandolfi et al. 2018, Atmos. Chem. Phys., 18, 7877–7911, 2018

R: Thanks for reviewer’s suggestion. We compared our results with the measurement at an urban site in Spain from March 2006 to February 2007 (Titos et al., 2012) and revised the sentence as:

“The annual mean σab for PM$_{10}$ and PM$_1$ in 2021 was 9.8 Mm^{-1} and 8.7 Mm^{-1}, which were both lower than the result observed in Nainital, in the GH region, India (Dumka et al., 2015), and the measurement at an urban site in Spain from March 2006 to February 2007 (Titos et al., 2012).” (Line 242-245 in the revised manuscript)

Reference:

Dumka, U. C., Kaskaoutis, D. G., Srivastava, M. K., and Devara, P. C. S.: Scattering and absorption properties of near-surface aerosol over Gangetic–Himalayan region: the role of boundary-layer dynamics and long-range transport, Atmospheric Chemistry and Physics, 15, 1555-1572, 10.5194/acp-15-1555-2015, 2015.

Titos, G., Foyo-Moreno, I., Lyamani, H., Querol, X., Alastuey, A., and Alados-Arboledas, L.: Optical properties and chemical composition of aerosol particles at an urban location: An estimation of the aerosol mass scattering and absorption efficiencies, Journal of Geophysical Research, 117, D04206, 10.1029/2011jd016671, 2012.

L225: You could use the reference Collaud-Coen et al. 2020, Atmos. Chem. Phys., 20, 8867–8908, 2020 for comparison of differences in the scattering and absorption trends. The study of Collaud-Coen et al., is a long-term trend analysis and the results are different but I think it could be mostly due to the difference in the time period considered.

R: Thanks for reviewer's suggestion. We compared our observed results of σ_{ab} and SSA with those from the recommended references. The following sentences were added to the section:

"Actually, σ_{ab} that was observed at a background station in China and the European stations, which was with time series longer than 10 years, also observed the reduction. σ_{ab} showed a statistically significant decreasing trend in Mt.Waliguan, a background station in China, from 2008–2018 (Collaud Coen et al., 2020), which was similar to a decreasing trend of black carbon (BC) in Mt.Waliguan from 2008-2017, mainly related to emission reduction (Dai et al., 2021). A statistically significant decrease of 10-year σ_{ap} was found in 12 stations in Europe, which was similar to a decreasing trend in BC concentration in Europe related primarily to traffic emission decreases (Collaud Coen et al., 2020)." (Line 257-265 in revised manuscript)

"Collaud Coen et al. (2020) found that SSA observed in Mt. Waliguan, a background station in Asia, presented an increasing trend based on 10-year datasets, which were related to more recent abatement policies." (Line 273-275 in the revised manuscript)

Reference:

Collaud Coen, M., Andrews, E., Alastuey, A., Arsov, T. P., Backman, J., Brem, B. T., Bukowiecki, N., Couret, C., Eleftheriadis, K., Flentje, H., Fiebig, M., Gysel-Beer, M., Hand, J. L., Hoffer, A., Hooda, R., Hueglin, C., Joubert, W., Keywood, M., Kim, J. E., Kim, S.-W., Labuschagne, C., Lin, N.-H., Lin, Y., Lund Myhre, C., Luoma, K., Lyamani, H., Marinoni, A., Mayol-Bracero, O. L., Mihalopoulos, N., Pandolfi, M., Prats, N., Prenni, A. J., Putaud, J.-P., Ries, L., Reisen, F., Sellegri, K., Sharma, S., Sheridan, P., Sherman, J. P., Sun, J., Titos, G., Torres, E., Tuch, T., Weller, R., Wiedensohler, A., Zieger, P., and Laj, P.: Multidecadal trend analysis of in situ aerosol radiative properties around the world, Atmospheric Chemistry and Physics, 20, 8867-8908, 10.5194/acp-20-8867-2020, 2020.

Dai, M., Zhu, B., Fang, C., Zhou, S., Lu, W., Zhao, D., Ding, D., Pan, C., and Liao, H.: Long-Term Variation and Source Apportionment of Black Carbon at Mt. Waliguan, China, Journal of Geophysical Research: Atmospheres, 126, 10.1029/2021jd035273, 2021.

General comment: make clear that the time period considered is not long enough to establish statistically significant trends. For trend analysis, at least 10 years of data are needed. On the other hand, the authors could apply statistical tests to check if the decrease between 2018 and 2021 was statistically significant.

R: Thanks for the reviewer’s suggestion, which is the same as above question. The trend of the monthly σ_{ab} was analyzed using the Mann–Kendall (MK) method at a 95% confidence level, with the statistical parameters listed in Table S1. The Mann–Kendall trend test supported that the decrease in σ_{ab} from 2018 to 2021 was significant. We agree that at least 10 years of data are needed for climatic trend analysis. 4-year data presented in the manuscript can’t represent the aerosol optical properties climatic trend. We just discuss the variation of σ_{ab} in 4 years in the revised manuscript as the following.:

“The annual mean σ_{ab} at 550 nm of PM_{10} and PM_1 decreased by 55.0% and 53.5%, respectively, from 2018 to 2021. The Mann–Kendall trend test of monthly mean σ_{ab} for PM_1 and PM_{10} supported that the decrease in σ_{ab} for PM_1 and PM_{10} from 2018 to 2021 was significant (Table S1).” (Line 234-237 in revised manuscript)

Table. S1 Mann–Kendall trend test results ($p = 0.05$) for monthly mean value of σ_{ab} for PM_1 and PM_{10} from 2018 to 2021. Z is the standardized test statistic value.

	Trend	Z	sen's slope
PM_1 σ_{ab}	Decreasing trend	-5.8134	-0.24856
PM_{10} σ_{ab}	Decreasing trend	-5.643	-0.29905

L293: There are many assumptions behind the calculation of RFE and the impact of this assumptions should be discussed in more detail. The RH impact in RFE should be included here, so the results are relative to ambient conditions and not dry conditions.

R: Thanks for reviewer’s suggestion. We added the discussion about the impact of assumptions behind the calculation of RFE including RH impact. The modified sentence as follow:

“In eq. (6) The fractional day length (D), solar constant (So), atmospheric transmission (T_{at}), fractional cloud amount (Ac), and surface reflectance (Rs) were constants, which were widely used in previous studies (Delene and Ogren, 2002; Andrews et al., 2011; Sherman et al., 2015; Shen et al., 2018). These values are the globally averaged values and don’t always represent the conditions in Beijing, but using the same constants makes it possible to compare the intrinsic forcing efficiency of the aerosols measured at different stations around the world and to study how the RFE changes with varying SSA and b (Sherman et al., 2015; Luoma et al., 2019). On the other hand, RFE is sensitive to RH as the aerosol optical properties are different due to hygroscopic growth (Fierz-Schmidhauser et al., 2010; Luoma et al., 2019). Previous studies demonstrate that SSA increases with RH, while b decreases with increasing RH (Carrico et al., 2003;

Cheng et al., 2008). The change of SSA to increase with RH and of b to decrease with RH will have opposite effects on the RFE, and thus to some extent, the RH dependencies of these two parameters will counterbalance each other (Luoma et al., 2019). Titos et al. (2021) found that the range of forcing enhancement in different types of sites varies from almost no enhancement up to a factor of 3–4 at RH=90 %. The results observed in urban Beijing showed that the aerosol radiative forcing at RH = 80 % was 1.48 times that under dry conditions (Xia et al., 2023). RFE was calculated at dry state in this study, while atmosphere is not generally dry in the ambient air. Thus, the RFE in this study does not represent ambient conditions. The simplified RFE in this study does not represent the actual value for the aerosol forcing; however, it can still indicate how the changes in aerosol optical properties affect the climate (Delene and Ogren, 2002; Andrews et al., 2011; Sherman et al., 2015).” (Line 360-382 in the revised manuscript)

Reference:

Carrico, C. M., Rood, P. K. a. M. J., and Bates, P. K. Q. a. T. S.: Mixtures of pollution, dust, sea salt, and volcanic aerosol during ACE-Asia: Radiative properties as a function of relative humidity, J. Geophys. Res., 108, 8650, 10.1029/2003jd003405, 2003.

Cheng, Y. F., Wiedensohler, A., Eichler, H., Su, H., Gnauk, T., Brüggemann, E., Herrmann, H., Heintzenberg, J., Slanina, J., and Tuch, T.: Aerosol optical properties and related chemical apportionment at Xinken in Pearl River Delta of China, Atmospheric Environment, 42, 6351-6372, 10.1016/j.atmosenv.2008.02.034, 2008.

Delene, D. J., and Ogren, J. A.: Variability of Aerosol Optical Properties at Four North American Surface Monitoring Sites, J. Aerosol Sci., 59, 1135-1150, 10.1175/1520-0469(2002)059<1135:VOAOPA>2.0.CO;2, 2002.

Andrews, E., Ogren, J. A., Bonasoni, P., Marinoni, A., Cuevas, E., Rodríguez, S., Sun, J. Y., Jaffe, D. A., Fischer, E. V., Baltensperger, U., Weingartner, E., Coen, M. C., Sharma, S., Macdonald, A. M., Leaitch, W. R., Lin, N. H., Laj, P., Arsov, T., Kalapov, I., Jefferson, A., and Sheridan, P.: Climatology of aerosol radiative properties in the free troposphere, Atmos. Res., 102, 365-393, 10.1016/j.atmosres.2011.08.017, 2011.

Sherman, J. P., Sheridan, P. J., Ogren, J. A., Andrews, E., Hageman, D., Schmeisser, L., Jefferson, A., and Sharma, S.: A multi-year study of lower tropospheric aerosol variability and systematic relationships from four North American regions, Atmospheric Chemistry and Physics, 15, 12487-12517, 10.5194/acp-15-12487-2015, 2015.

Sheridan, P. J., and Ogren, J. A.: Observations of the vertical and regional variability of aerosol optical properties over central and eastern North America, J. Geophys. Res., 104, 16793-16805, 10.1029/1999jd900241, 1999.

Shen, Y., Virkkula, A., Ding, A., Wang, J., Chi, X., Nie, W., Qi, X., Huang, X., Liu, Q., Zheng, L., Xu, Z., Petäjä, T., Aalto, P. P., Fu, C., and Kulmala, M.: Aerosol optical properties at SORPES in Nanjing, east China, Atmos. Chem. Phys., 18, 5265-5292, 10.5194/acp-18-5265-2018, 2018.

Luoma, K., Virkkula, A., Aalto, P., Petäjä, T., and Kulmala, M.: Over a 10-year record of aerosol optical properties at SMEAR II, Atmos. Chem. Phys., 19, 11363-11382, 10.5194/acp-19-11363-2019, 2019.

Titos, G., Burgos, M. A., Zieger, P., Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Weingartner, E., Henzing, B., Luoma, K., O'Dowd, C., Wiedensohler, A., and Andrews, E.: A global study of hygroscopicity-driven light-scattering enhancement in the context of other in situ aerosol optical properties, Atmospheric Chemistry and Physics, 21, 13031-13050, 10.5194/acp-21-13031-2021, 2021.

Fierz-Schmidhauser, R., Zieger, P., Gysel, M., Kammermann, L., DeCarlo, P. F., Baltensperger, U., and Weingartner, E.: Measured and predicted aerosol light scattering enhancement factors at the high alpine site Jungfraujoch, Atmos. Chem. Phys., 10, 2319–2333, 2010.

Titos, G., Burgos, M. A., Zieger, P., Alados-Arboledas, L., Baltensperger, U., Jefferson, A., Sherman, J., Weingartner, E., Henzing, B., Luoma, K., O'Dowd, C., Wiedensohler, A., and Andrews, E.: A global study of hygroscopicity-driven light-scattering enhancement in the context of other in situ aerosol optical properties, Atmospheric Chemistry and Physics, 21, 13031-13050, 10.5194/acp-21-13031-2021, 2021.

Xia, C., Sun, J., Hu, X., Shen, X., Zhang, Y., Zhang, S., Wang, J., Liu, Q., Lu, J., Liu, S., and Zhang, X.: Effects of hygroscopicity on aerosol optical properties and direct radiative forcing in Beijing: Based on two-year observations, Sci Total Environ, 857, 159233, 10.1016/j.scitotenv.2022.159233, 2023.

Section of cluster analysis: not sure if this section is very informative relative to the main objective of the paper. Further discussion on the changes observed over the years is difficult to draw.

R: Thanks for this comment. Based on cluster analysis, we try to give some information on how the aerosol optical properties vary with the air mass pathways. We revised this section carefully and found we miss-matched the data when we plotted Figure 8. So, we corrected the Figure 8 and revised the paragraph as follow:

“In addition to local emissions, regional transport is also an important source of particulate matter in Beijing (Chang et al., 2019). Based on previous studies, aerosol source regions and air mass pathways could also affect aerosol optical properties, and the different origins of air masses showed different aerosol optical properties (Zhuang et al., 2015; Pu et al., 2015). The air mass back-trajectories analysis in the North China Plain revealed that the absorption coefficients and SSA were high when the air masses came from densely populated and highly industrial areas (Yan et al., 2008). Therefore, air mass back-trajectories were analyzed in this study to explore the regional transports’ influence on aerosol optical properties. First, the air mass back trajectories during 2018–2021 were calculated and clustered (Fig. 7); then, we statistic the aerosol optical properties of each cluster from 2018-2021 (Fig. 8). Based on the Euclidean distance, the back trajectories were classified into five clusters, in which clusters 1, 2 and 3, which originated from the clean areas in Mongolia and eastern Inner Mongolia, and transported to Beijing along the pathway with low emissions, were corresponded to low σ_{ab} and low $PM_{2.5}$ (Fig. 8a, d). Cluster 4 from the south of Beijing and cluster 5 from the west of Beijing were referred to as the polluted air masses, and the average $PM_{2.5}$ concentrations and σ_{ab} of clusters 4 and 5 were higher than those of clusters 1, 2, and 3 in each year (Fig. 8a, d). Cluster 4 passed through Shandong and Hebei Province, which was heavily polluted before arriving in Beijing. Cluster 5 passed through polluted Shanxi and Hebei during transport. Higher σ_{ab} and $PM_{2.5}$ mass concentrations were mainly distributed in clusters 4 and 5 each year. Lower AAE in cluster 4 indicates that the southern air mass carries more freshly emitted BC particles. SSA of cluster 4 from the south was higher (Fig. 8b), which may relate to low BC/PM$_{2.5}$ ratios in south air masses (Xia et al., 2020). Zhang et al. (2013) found that high levels of secondary inorganic aerosols related to high humidity were transported by southern air masses, which enhanced heterogeneous reaction and leaded to relatively low BC/PM$_{2.5}$ ratios. Fig. 7b showed percentage of each cluster accounting for the total back trajectories in each year. The results indicated that variation in each cluster fraction from 2018 to 2021 was slight. In general, cluster 1-5 accounted for 19%-21%, 13%-17%, 16%-20%, 29%-36%, 12%-20% of total back trajectories, respectively. Notably, the percentage of polluted-relevant air masses (cluster 4 and cluster 5) was ~50% each year, indicating that the transport from the south and the west of has a considerable impact on the aerosol optical properties. σ_{ab} corresponding to clusters 4 and 5 decreased by 47.3% and 58.4%, and a decrease of PM$_{2.5}$ mass concentration from clusters 4 and 5 was 38.9% and 37.4% during 2018 - 2021 (Fig. 8a, d), which may result from the air quality has improved caused by control of source emissions in surrounding regions of Beijing. Therefore, the comprehensive control of atmospheric pollution in Beijing and surrounding regions would be highly effective in reducing air pollution in Beijing." (Line 406-442 in the revised manuscript)

Reference:

Chang, X., Wang, S., Zhao, B., Xing, J., Liu, X., Wei, L., Song, Y., Wu, W., Cai, S., Zheng, H., Ding, D., and Zheng, M.: Contributions of inter-city and regional transport to PM2.5 concentrations in the Beijing-Tianjin-Hebei region and its implications on regional joint air pollution control, Sci Total Environ, 660, 1191-1200, 10.1016/j.scitotenv.2018.12.474, 2019.

Pu, W., Zhao, X., Shi, X., Ma, Z., Zhang, X., and Yu, B.: Impact of long-range transport on aerosol properties at a regional background station in Northern China, Atmospheric Research, 153, 489-499, 10.1016/j.atmosres.2014.10.010, 2015.

Xia, Y., Wu, Y., Huang, R. J., Xia, X., Tang, J., Wang, M., Li, J., Wang, C., Zhou, C., and Zhang, R.: Variation in black carbon concentration and aerosol optical properties in Beijing: Role of emission control and meteorological transport variability, Chemosphere, 254, 126849, 10.1016/j.chemosphere.2020.126849, 2020.

Yan, P., Tang, J., Huang, J., Mao, J. T., Zhou, X. J., Liu, Q., Wang, Z. F., and Zhou, H. G.: The measurement of aerosol optical properties at a rural site in Northern China, Atmospheric Chemistry and Physics, 8, 2229–2242, 10.5194/acp-8-2229-2008, 2008.

Zhang, R., Jing, J., Tao, J., Hsu, S. C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of PM2.5 in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, 10.5194/acp-13-7053-2013, 2013.

Zhuang, B. L., Wang, T. J., Liu, J., Ma, Y., Yin, C. Q., Li, S., Xie, M., Han, Y., Zhu, J. L., Yang, X. Q., and Fu, C. B.: Absorption coefficient of urban aerosol in Nanjing, west Yangtze River Delta, China, Atmos. Chem. Phys., 15, 13633-13646, 10.5194/acp-15-13633-2015, 2015.