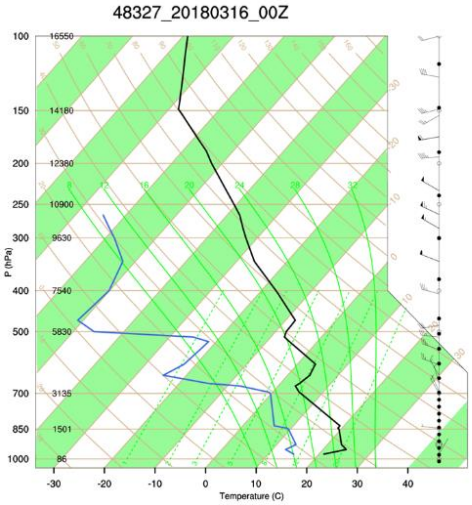
Reviewer#1

1. The fire plume rise and its simulation by WRF-Chem aren't discussed in the paper, thought there is extensive analysis of the vertical distribution of the biomass burning plumes. The fire plume rise is an important process by which aerosol and gaseous species from wildland fires are injected into the free troposphere, where they can be transported to long distances. WRF-Chem has an 1D plume rise parameterization (Freitas et al.). It isn't clear if this scheme was used, and how it performed, associated uncertainties and their impact on the findings on the results presented here.

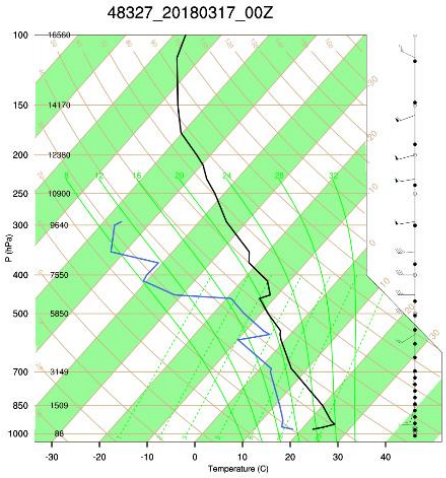
R: Thank you for your comments. Yes, the Freitas et al (2007) 1-D plume-rise model has been incorporated into WRF-Chem (Power et al. 2017; Grell et al. 2011), and this scheme was used in our simulation (Please also see Q2(a) in this response). To estimate heat flux, fires are divided into four surface categories based on WRF's land use dataset: savanna, grassland, tropical and extra-tropical forest. Simulated atmospheric sounding data for the plume rise model are computed every hour at each grid point containing an active fire. The final height reached by a plume is controlled by the thermodynamic stability of the atmospheric environment and the surface flux released from the fire (Freitas et al. 2011; Grell et al. 2011).

As mentioned in the article, our previous study (Lin et al. 2009) proposed that the mountain lee-side trough over Indochina plays a dominant role in the uplift of the BB pollution easily transport to the elevation >3000 m. Actually, this mechanism also can be applied in this case, as shown in Figure R1 (a) and (b), weak wind speed (near calm) and stable weather conditions existed in the boundary layer from the sounding (WMO 48327, Figure R1(a) and (b)) at Chiang-Mai, Thailand. According to the 850 and 700 hPa weather map (Fig. R1 (c) and (d)), a clear lee side trough formed between 16-17 March 2018 might provide an extra force for uplifting the air mass to a high elevation. Therefore, it is not easy to estimate the impact of individual factors on the plume rise height. The uncertainties of the plume injection height were not only related to the number of fire hot spots, and land use (surface categories), but also weather conditions (e.g., lee side trough existed or not).

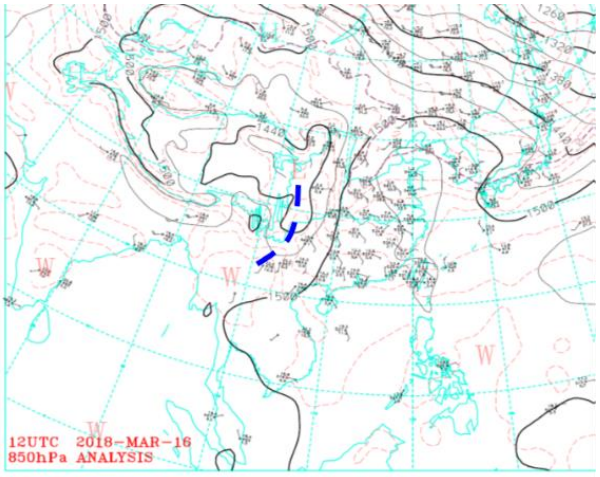
(a)



(b)



(c)



(d)

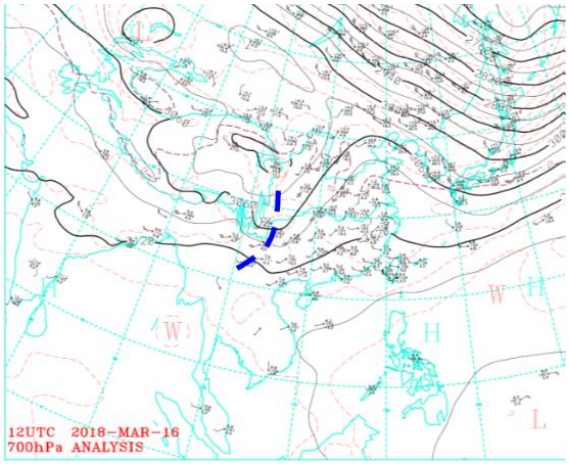


Figure R1 Sounding at station 48327 (located at ChingMei, Thailand) at (a) 00:00UTC, 16 March (b) 00:00 UTC, 17 March 2018.; The weather charts from the Central Weather Burea of Taiwan at 12 UTC on 16 March 2018 (c) 850 hPa (d) 700 hPa

References:

Freitas, S. R., Longo, K. M., Chatfield, R., Latham, D., Silva Dias, M. A. F., Andreae, M. O., Prins, E., Santos, J. C., Gielow, R., and Carvalho Jr., J. A.: Including the sub-grid scale plume rise of vegetation fires in low resolution atmospheric transport models, Atmos. Chem. Phys., 7, 3385–3398, doi:10.5194/acp-7-3385-2007, 2007.

Freitas, S. R., Longo, K. M., Alonso, M. F., Pirre, M., Marecal, V., Grell, G., Stockler, R., Mello, R. F., and S´anchez G´acita, M.: PREP-CHEM-SRC – 1.0: a preprocessor of trace gas and aerosol emission fields for regional and global atmospheric chemistry models, Geosci. Model Dev., 4, 419–433, doi:10.5194/gmd-4-419-2011, 2011.

Grell, G., Freitas S.R., Stuefer M., Fast, J., Inclusion of biomass burning in WRF-Chem: impact of wildfires on weather forecasts. Atmos. Chem. Phys., 11, 5289–5303, doi:10.5194/acp-11-5289-2011, 2011.

Lin, C.Y., H.M. Hsu, Y.H. Lee, C. H. Kuo, Y.F. Sheng, D. A. Chu: A new transport mechanism of biomass burning from Indochina as identified by modeling studies., Atmos. Chem. Phys., 9, 7901-7911. DOI: 10.5194/acp-9-7901-2009 https://doi.org/10.5194/acp-9-7901-2009, 2009

Powers G., Klemp J. B. , Skamarock W. C., Davis C. A., Dudhia J., Gill D. O., et al.The weather research and forecasting model overview, System Efforts, and Future Directions.Bulletin of the American Meteorological Society 2017 Vol. 98 Issue 8 Pages 1717-1737

2. (a) Figure 12 shows the effect of the smoke plume on cloud water. However, I can't find any description of the model configuration on how the aerosol feedback on the meteorology is simulated in this study.

R: Thank you for the suggestions. Table R1 (also see Table 1 in the revision article) summarizes the model configuration in this revision. Regarding the aerosol indirect effect and the discussion on aerosol feedback, please see the responses in the next question (Q2 (b)) and Q4.

Resolution	10km
Microphysics	Lin
Cumulus parameterization	Grell 3D ensemble scheme
Planetary Boundary Layer	Mellor-Yamada-Janjic TKE scheme
Longwave radiation	RRTMG
Shortwave radiation	RRTMG
Fire emissions	FINN V1.5
Anthropogenic emissions	MICS-Asia III(2010) + Taiwan Emission Data System ver 9.0 (2013)
Biogenic emissions	MEGAN V2.04
Chemistry option	RACM Chemistry with MADE/VBS aerosols using KPP library along with the volatility basis set (VBS) used for Secondary Organic Aerosols
Photolysis option	Madronich
wet scavenging	On , (Neu and Prather, 2012)
Cloud chemistry	On,
feedback from the aerosols to the radiation schemes	On
the time interval for calling the biomass-burning plume rise subroutine	180 min
feedback from the parameterized convection to the atmospheric radiation and the photolysis schemes	On
Subgrid-scale wet scavenging	on
Subgrid aqueous chemistry	on

Table R1 WRF-Chem model configuration and physics and chemistry options in this study. (RRTMG=Rapid Radiative Transfer Model for General Circulation Models;FINN=Fire Inventory from National Center for Atmospheric Research)

(b) There are several feedback mechanisms of the aerosols on meteorology. Although WRF-Chem contains a few parameterizations to simulate these processes, large uncertainties remain with respect accurate representation of the aerosol-radiation-microphysics interactions. Authors present the results for such complex phenomena in a single graph without thorough discussion and sensitivity analysis (e.g. direct vs. indirect feedback). Moreover, given the relatively low aerosol concentrations in the smoke plumes analyzed here the sensitivity of the simulated cloud water concentrations to smoke plumes seem to be overly large.

R: Thank you for the suggestions. We agree that large uncertainties remain exited and still a challenge to accurately represent the aerosol-radiation-microphysics interactions, even in the state-of-the-art numerical model. Therefore, our intended purpose is trying to evaluate the potential impacts of long-range transport BB pollution from Indochina on East Asia.

Although the concentrations of individual aerosol components are low, the sum of measurement major aerosol components (OA+BC+SO_4^{2-} +NO_3^- +NH_4^+) on 19 March at BPTL (BB plume transport layer, 2000-4000 m) could be more than 13 μg/m^3 (Fig. 6b). Thus, the related feedback could be significant. As suggested by the reviewer, we further did the sensitivity analysis for the direct and indirect effects and related short-wave reduction at the ground surface in this revision.

Fig R2a shows the results along the flight for the simulated OA of the control simulation (CTRL, i.e. running with aerosol direct and indirect effect) and the case ROCD (Running Only Considered Direct effect). As mentioned in the article, a frontal system was just located from the East China Sea (ECS) to Southern China (SC) (Fig. 2e) on the event day, 19 March 2018. Most of the time the difference was not significant between the CTRL and the case ROCD, except for during 03:30-04:20 UTC where the flight was located north of 28 °N (Fig. R2b) and a frontal cloud band existed (Fig. 2g). In other words, the effect of wet scavenging reduced the aerosol concentration bias in the ECS and SC, as for the frontal system providing the moist air mass in the event flight F0319. **(L436-441)** (also see Figure 9 in the revised article)

We also carefully checked the hourly variations of the impact of aerosol indirect effect in the simulation. Figures R3 (a)-(c) indicate the simulated spatial distribution of $PM_{2.5}$ and cloud water for the CTRL simulation at an altitude of 2000 m at 02:00, 04:00, and 06:00 UTC, respectively. The simulated cloud water area could be represented by the location of the frontal system, from Korea and Japan to southern China. First of all, we examine the role of the indirect effect (chemistry-microphysics interactions) in the

simulation. Figure R3 (d)-(f) showed the difference between the control simulation (case CTRL) and the case ROCD. It was noted that wet scavenging mainly occurred along the frontal system and north of it, from Japan to southern China, i.e. major impacts were over ECS and SC. These results are also consistent with the finding in Figure 12 and the results in Figure 9. The impact of the BB (i.e. difference between with (ctrl run) and without BB emission) on these regions was shown in Figure R3 (g)-(i). The results indicated that the BB plume was transported mainly south of 30 N and the enhancement area of cloud water was along the frontal system. Figure R4 shows the cloud water difference when the aerosol indirect effect turned off in the simulation over different regions on 19 March 2018. The significant cloud water shortage over ECSA, and SCA could be as high as 2.4 mg/kg and 1.5 mg/kg, respectively. In other words, the role of the chemistry-microphysics interactions (indirect effect) plays an important role in the cloud water enhancement in the SCA and ECSA in this study **(L549-554)** (also see Figure 12b in the revised article).

Figure R5 shows the simulated downward shortwave flux at the noontime at ground surface due to BB was 2-4% and 5-7% reduction over the regions ECSA and SCA, respectively, during 18-19 March 2018. However, a significant shortwave flux reduction at noontime at the ground surface could be 15-20% due to aerosol indirect effect in the region SCA during 18-19 March 2018. The combination of BB aerosols enhancement and increased cloud water results in shortwave radiation reduction, implying the possibility of regional climate change in East Asia driven by BB aerosols. **(L557-560)** (also see supplementary Fig. 4a-b in the revised article)**.**

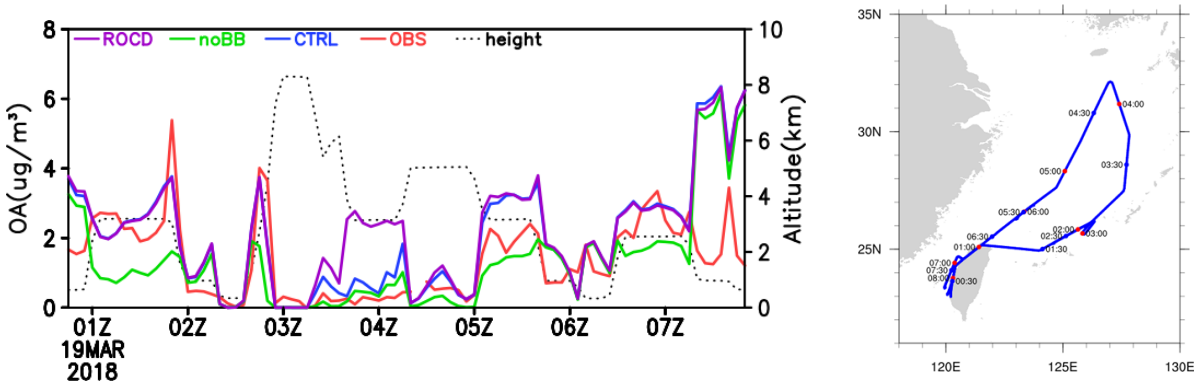


Figure R2 (a) Observed (OBS, red) and simulated concentration with (CTRL, blue) and without indirect effect (ROCD, purple) and without BB emission (noBB, green) along with the flight altitude for OA on 19 March (b) The HALO flight and detailed locations on 19 March.

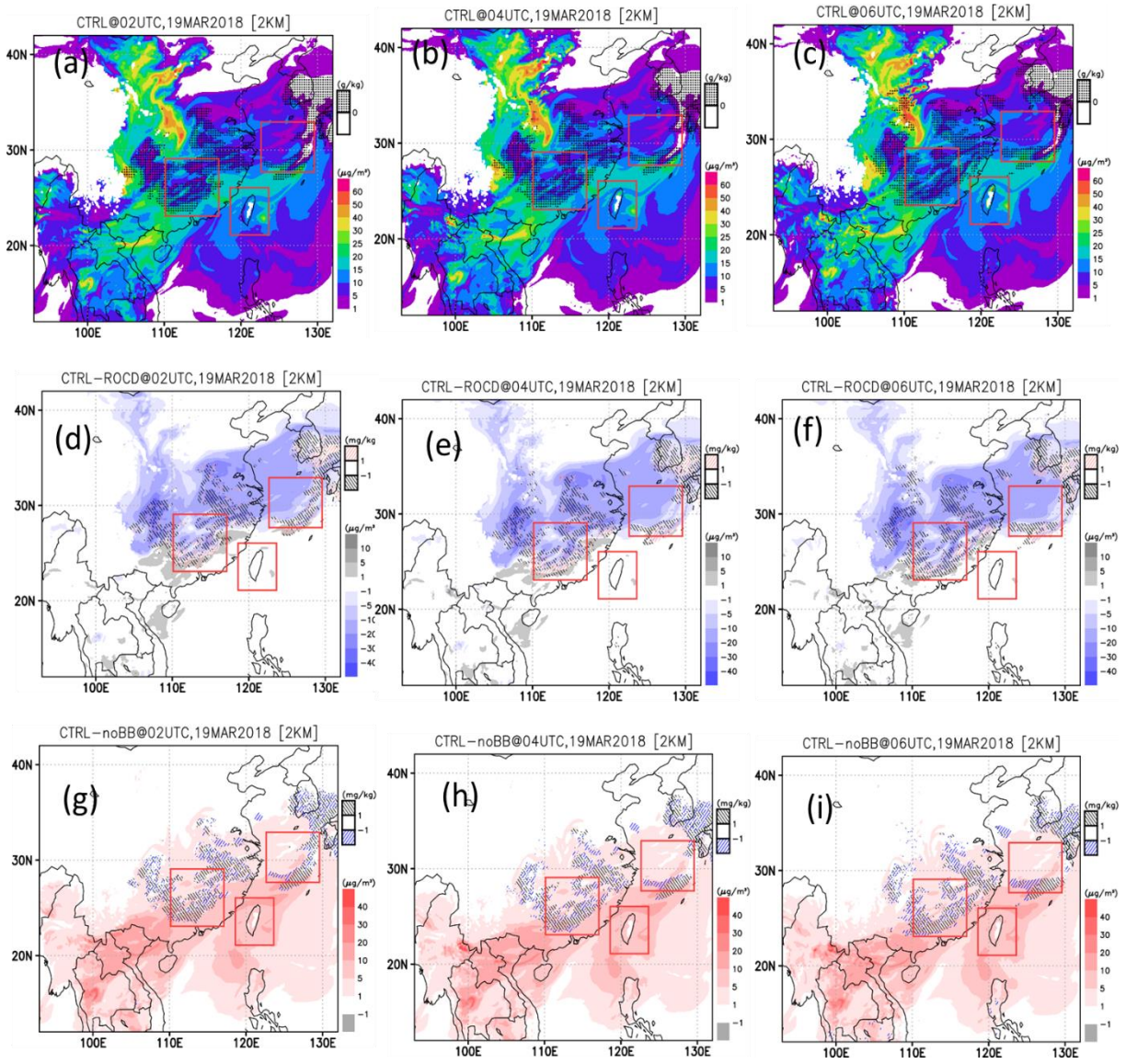


Figure R3 (a)-(c) Spatial distribution of control simulation for PM2.5 (color) and cloud water(dot-shaded) at altitude 2000 m at 02:00, 04:00, and 06:00 UTC, respectively, on 19 March 2018. (d)-(f): the effect of indirect effect, i.e. the simulation difference between the case CTRL (control) simulation and ROD (running only considered direct effect), respectively. (g)-(i): the effect of BB plume transport, i.e. the simulation difference between with and without BB emission, respectively.

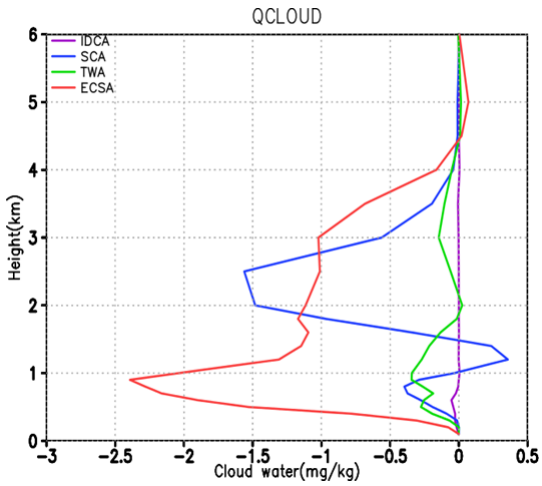


Figure R4 Simulated vertical distribution of cloud water difference between with and without indirect effect in the model on 19 March 2018.

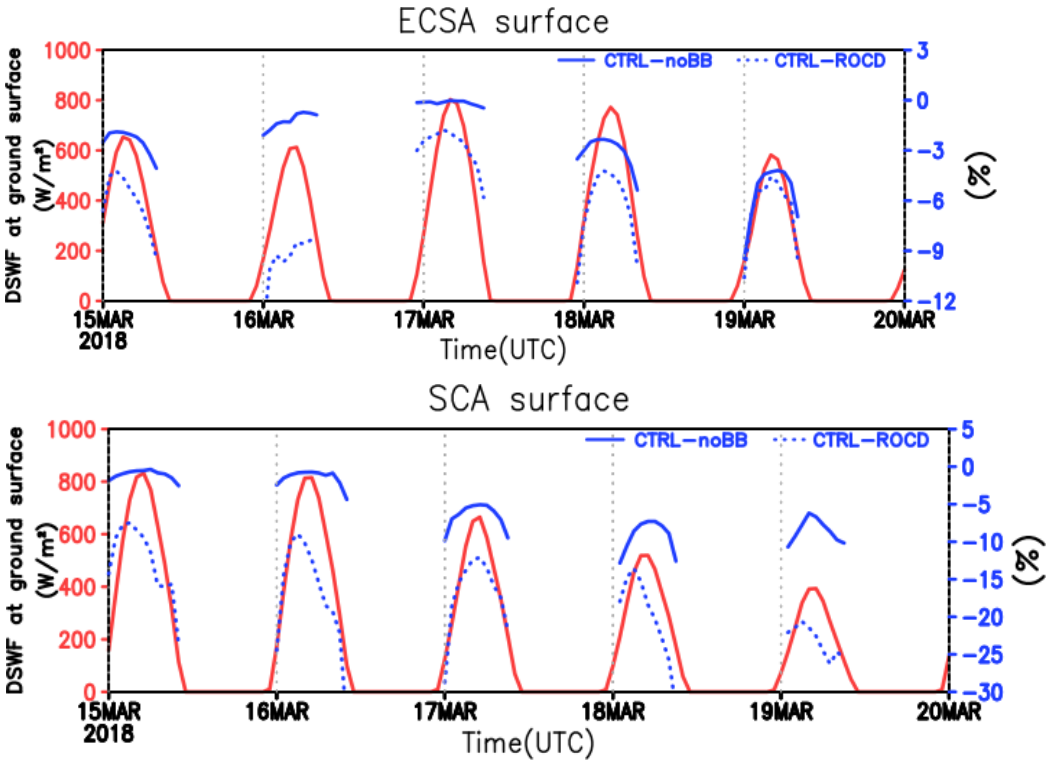


Figure R5 Simulated mean downward short wave flux (DSWF) (red) reduction at the ground surface over the regions in Fig.1a and contributed by BB (%, blue), aerosol indirect effect (%, dashed) during 15-19 March 2018. (a) ECSA (b) SCA

3. The concluding statements (L575-579) aren't necessarily based on the findings from this study.

R: Thank you for the suggestions. This paragraph has been amended in this revision.

Minor comments:

4. It'd be helpful to add a Table to list the WRF-Chem model configuration. Some of the settings are listed in the text. Information on the lateral boundary conditions for the chemical species, their cycling between the subsequent simulations and fire plumerise are missing. How the wet removal of the gas and aerosol species are parameterized in the model?

   R: Thank you for the suggestion. A table listing the model configuration has been added in this revision (Table1, please see in Q2(a)). To avoid the global information disturbing the simulation result, we do not include global chemical information at the lateral boundary. We used the WRF-Chem (V4.1.1) default grid-scale wet-scavenging scheme, which is based on Neu and Prather (2012) and updated by Bela et al. (2016) to include ice retention factors in the grid-scale wet-scavenging. When wet scavenging occurs, the amount of trace gas that dissolves in cloud water is governed by Henry's law. The chemistry option, RACM chemistry with MADE/VBS aerosol scheme was used in this study (Table R1 in Q2a).

   References:

   Bela, M. M., Barth, M. C., Toon, O. B., Fried, A., Homeyer, C. R., Morrison, H., et al. (2016). Wet scavenging of soluble gases in DC3 deep convective storms using WRF-Chem simulations and aircraft observations. Journal of Geophysical Research: Atmospheres, 121,4233–4257. https://doi.org/10.1002/2015JD024623

   Neu, J. L., & Prather, M. J. (2012). Toward a more physical representation of precipitation scavenging in global chemistry models: Cloud overlap and ice physics and their impact on tropospheric ozone. Atmospheric Chemistry and Physics, 12(7), 3289–3310. https://doi.org/10.5194/acp-12-3289-2012

5. The paper doesn't provide any information about the measurement uncertainties for the chemical species. For instance, the AMS data (OA, sulfate concentrations reported here) usually have significant uncertainty due to the collection efficiency and cutoff size (< 1 micron).

R: The uncertainty of the AMS regarding ionization efficiency (IE) and collection efficiency (CE) was determined to be 34% for ammonium and nitrate, 36% for sulfate, and 38% for organics. The related information can be seen in Bahreini et al.,(2009; including the auxiliary material S1: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2008JD011493).

Middlebrook et al (2012) (http://dx.doi.org/10.1080/02786826.2011.620041) recommend a collection efficiency for low nitrate concentrations of 0.5, which is the value we used. Then the overall uncertainty is somewhat reduced and estimated to be around 30%.

The size cut of the inlet is not an uncertainty, but an instrument feature, therefore one should always refer to "submicron aerosol" when describing AMS data (at least those with a classical inlet setup).

References:

Bahreini R., Ervens, B., Middlebrook, A. M., et al., Organic aerosol formation in urban and industrial plumes near Houston and Dallas, Texas,J. Geophys. Res., 114, D00F16, doi:10.1029/2008JD011493, 2009.

Middlebrook A. M., Bahreini R., Jimenez Jose L. and Canagaratna M. R.) Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, Aerosol Science and Technology, 46:3, 258-271,DOI: 10.1080/02786826.2011.620041, 2012.

6. L156: For WRF-Chem the more recent paper (Powers et al.) can be also cited.

R: Thank you for the suggestions. The recent paper (Power et al. 2017), has been cited in this revision. **(L160)**

7. L272: What do you mean by "stable"?

R: The text has been dropped in this revision.

8. Chapter 3.3: this chapter needs to be shortened.

R: This section has been modified in this revision.