

Convective uplift of pollution from the Sichuan basin into the Asian monsoon anticyclone during the StratoClim aircraft campaign

By K. O. Lee et al.

Reply to the referees' comments

In the following, the comments made by the referees appear in black, while our replies are in red, and the proposed modified text in the typescript is in blue.

Referee #1 comments

Summary general Comments

This manuscript investigated the convective uplift of pollution from the Sichuan basin into the Asian summer monsoon anticyclone during the StratoClim aircraft campaign in 2017 by simulations with the Meso-NH cloud-chemistry model. After validation with the BT from satellite data and CO and ozone from airborne observations, the simulations are believed to study the impact of Sichuan convection on the AMA composition. Overall, the manuscript shows some interesting results, particularly the role of Sichuan basin as source region of pollution. Some major issues should be addressed before acceptance for publication in ACP.

We appreciate the time and effort you put in this review as well your helpful comments on our paper. We have worked hard to improve the manuscript. Replies to each comment are listed below.

Major Comments:

1. How to verify the simulations of aerosol in the tropopause layer? Because no measurements are used to validate the simulation. There are already some published studies, where the observations of aerosol signal are used to validate the simulations.

During the StratoClim aircraft campaign in 2017, UHSAS (ultra high sensitive aerosol spectrometer) was embarked on the Geophysica aircraft to measure the total number concentration of aerosol in size range of 0.65–1000 nm. The measured and simulated particle number concentrations are compared in Figure A. In the figure, the particle number concentrations observed by UHSAS during flights #F6 (a), #F7 (b), and #F8 (c) are displayed by red marks while the simulated domain-averaged number concentration within a box covering each flight track by Meso-NH control run is displayed by black solid line.

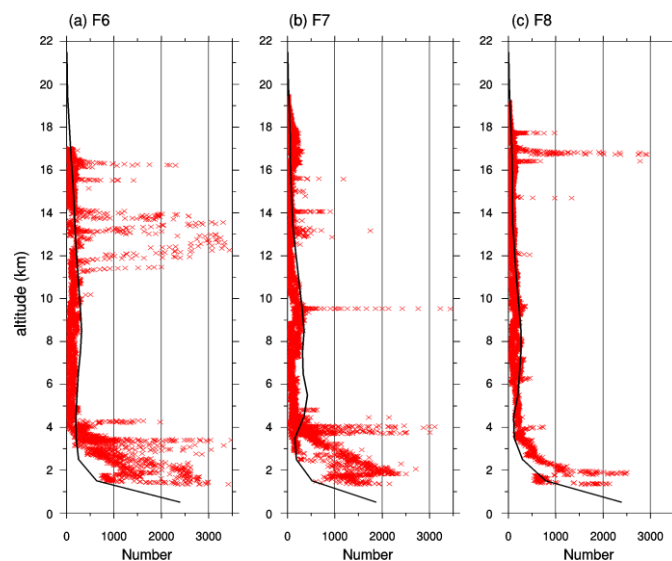


Figure A. Profiles of particle number concentration measured by UHSAS sensor (red crosses marks) aboard flights (a) #F6, (b) #F7, and (c) #F8 and simulated by the Meso-NH model in a box area covering each flight tracks (black solid line).

Figure A shows that the model captures the general profile shape of particle number concentration in the 0–400 range in the altitudes above 4 km as measured. However the model missed the large number concentration stored in lower troposphere below 4 km. Also, we also have performed the apple to apple comparison (not shown) as we did for CO and O₃ (Figures 4, 5, and 6), however model couldn't capture the detailed variation of aerosol along the flight tracks. This discrepancy may be linked to the accuracy of the initial forcing (i.e. MOZART), coarse model grid spacing (i.e. 15 km), normalized aerosol size and number distributions at initial state, and/or etc. This part will be more deeply investigated in future study to produce more realistic aerosol distribution. However the purpose here is to understand the lifecycles of primary pollutants, CO, primary organic aerosol (POA) and black carbon (BC) uplifted in the Sichuan basin by deep convection after thorough validation using airborne in situ measurements.

2. Could you validate the CO simulations with MLS data? The simulations are not so good for F6 and F8 as shown in Fig. 4, so more validations are in need. MLS CO data will surely show the enhanced CO plume after the deep convection if the results given in this paper are correct.

The simulation for F6 (Fig. 4b) couldn't capture the short 2 observed-CO peaks during 32000–36000 s, but rather smooth CO enhancements. However, except for this 4000 s period, the model captures very well most of the variations. For F8 (Fig. 4d), the model underestimates the CO concentration during the first leg (32000–34000 s) and last leg (42000–44300 s), and it misses short CO variations occurring at 35000 s, 35400 s, and 37500 s. Nevertheless, the model reproduces the general CO variations during F8.

Even if the model failed to capture some detailed CO features, it successfully captured the general CO variations during F6 and F8. It is also important to mention that the agreement is even better for F5 and F7. And, concerning F7 the model reproduces CO enhancements clearly identified as Sichuan contributions by the sensitivity tests performed with the model.

Such model-observation agreement is therefore largely enough to validate the ability of the model to document the impact of overshooting convection on the AMA composition. The missed short CO peaks for F6 and are probably linked to the coarse model horizontal resolution not adapted to capture the finest plumes while the overestimated CO concentrations during take-off and landing may be related to the emission inventory. The above discussion has been further strengthened in the manuscript.

♣ Page 8, line 231–237

"[...] As for flight #6 the model is not able to reproduce the short CO peaks but instead produces longer and smoother increases. For flight #8, the model missed the very short CO peaks at ~17 km. This is probably linked to a too coarse model grid spacing not adapted to capture fine CO plumes. For F8, At the beginning and end of the flight, the aircraft ascends to 19.4 km and the simulation overestimates the concentrations by up to 30 ppb. About 40 ppb overestimation which is also clear for the tropospheric profiles during take-off and landing. This is probably linked to the emission inventory."

As mentioned by referee, MLS has been widely used for studies in the upper troposphere and lower stratosphere (UTLS). In particular, it has been used to document large scale variations of CO in the AMA and to validate UTLS CO distributions from global scale models (Park et al., 2007; Park et al., 2009; Barret et al., 2008). Nevertheless, MLS has a limited spatio-temporal coverage (3500 profiles a day) and can hardly resolve fine scale (< 100km; < 2–5 days) CO structures such as those reproduced by our simulations. Furthermore, in August and September 2017 (d213–d273) MLS data are not available on the site official MLS site (<https://disc.gsfc.nasa.gov/datasets/>). Thus authors kindly propose to keep Figure 4 as it is.

Minor Comments:

1. L43–45: A recent paper by Bian et al. (2020) gives a comprehensive review of the deep convection on the UTLS composition during the ASM, which is recommended to be cited here. Bian, J. et al. 2020: Transport of Asian surface pollutants to the global stratosphere from the Tibetan Plateau region during the Asian summer monsoon, *National Science Review*, 7, 516–533, doi:10.1093/nsr/nwaa005.

Thank you for suggesting the recent article. This has been cited in the manuscript.

♣ Page 2, line 44–45

“[...] have a significant chemical and radiative impact (Mason and Anderson, 1963; Dickerson et al., 1987; Randel and Park, 2006; Su et al., 2011; Fadnavis et al., 2013; Gu et al., 2016; [Bian et al., 2020](#)). [...]”

♣ Page 17, line 508–510

[Bian, J., Li, D., Bai, Z., Li, Q., Lyu, D., and Zhou, X.: Transport of Asian surface pollutants to the global stratosphere from the Tibetan Plateau region during the Asian summer monsoon. *National Science Review*, 7, 516–533, doi:10.1093/nsr/nwaa005, 2020.](#)

2. L52: Higher tropopause over the ASM region is shown by Bian et al. (2012), which also shows the structure of AMA and therefore is recommended to be cited here. Bian, J., L. L. Pan, L. Paulik, H. Vomel, H. Chen, and D. Lu, 2012: In situ water vapor and ozone measurements in Lhasa and Kunming during the Asian summer monsoon. *Geophys. Res. Lett.*, 39, L19808, doi:10.1029/2012GL052996.

Thank you again. This has been cited in the manuscript.

♣ Page 2, line 52–54

“[...] above the ASM is relatively high (16–17.5 km) and the AMA extends into the lower stratosphere spanning from around 200 hPa to 70 hPa (12–18.5 km above sea level), i.e. approximately the whole UTLS (Highwood and Hoskins, 1998; Randel and Park, 2006; [Bian et al., 2012](#)).”

♣ Page 16, line 511–512

[Bian, J., Pan, L.L., Paulik, L., Vomel, H., Chen, H., and Lu, D.: In situ water vapor and ozone measurements in Lhasa and Kunming during the Asian summer monsoon. *Geophys. Res. Lett.*, 39, L19808, doi:10.1029/2012GL052996, 2012.](#)

3. L64–68: The different contribution from Indian and China sources to the UTLS is investigated by Yan et al. (2015), which conducts the simulation for one month with WRF-chem model and is recommended to be cited here. Yan, R. and J. Bian, 2015: Tracing the boundary layer sources of carbon monoxide in the Asian summer monsoon anticyclone using WRF-Chem. *Adv. Atmos. Sci.*, 32(7), 943–951, doi:10.1007/s00376-014-4130-3.

This has been cited in the manuscript.

♣ Page 2, line 64–65

“[...] demonstrated that the BL pollution uplifted to the AMA was mostly from Indian or South Asian sources (Park et al., 2009; [Yan and Bian, 2015](#); [Barret et al., 2016](#)). [...]”

♣ Page 22, line 685–686

[Yan, R. and J. Bian: Tracing the boundary layer sources of carbon monoxide in the Asian summer monsoon anticyclone using WRF-Chem. *Adv. Atmos. Sci.*, 32\(7\), 943–951, doi:10.1007/s00376-014-4130-3, 2015.](#)

4. L180–190: How are the CCN activation and second activation by entrainment in the convective cloud considered which is critical to the simulation of aerosol profile as shown by Yu et al. (2018)? Yu, P., K.D. Froyd, R.W. Portmann, O.B. Toon, S. R. Freitas, C. G. Bardeen, C. Brock, T. Fan, R. S. Gao, J. M. Katich, A. Kupc, S. Liu, C. Maloney, D. M. Murphy, K. H. Rosenlof, G. Schill, J. P. Schwarz and C. Williamson (2019), Efficient In-cloud Removal of Aerosols by Deep Convection, *Geophys. Res. Lett.*, 45, 1061-1069, <https://doi.org/10.1029/2018GL080544>

We do not use CCN activation and second activation in this simulation. There is no great physical sense in using aerosol activation parameterization at the horizontal resolution of the model (i.e. 15 km) without being able to compute supersaturation in the air parcel. The model is not in cloud-resolving-model (CRM) configuration. We therefore use a microphysical scheme at a time well adapted to this scale (ICE3, Pinty and Jabouille, 1998). This scheme follows the approach of Lin et al. (1983) in that a three-class ice parameterization is coupled to a Kessler's scheme for warm processes. The convection scheme is Kain-Fritsch-Bechtold (Bechtold et al., 2001) also widely used by the international community and well adapted to this resolution. Moreover the employed scheme is able to show the convective uplift of pollutant from boundary layer to the UTLS via deep convective cloud. To the sake of clarity about cloud and aerosol interaction, more information has been included in the manuscript.

♣ Page 6, lines 170–175

"[...] Tulet et al., 2005, 2006, 2010). The impaction scavenging by raindrops depends mainly on Brownian motion, interception, and inertial impaction following a formula originally described by Slinn (1983). Two lognormal modes of particles are considered, mode #1 (i.e. Aitken mode) of smaller particles with initial mean radius of 0.036 μm and standard deviation (σ) of 1.86, and mode #2 (i.e. accumulation mode) of larger particle with initial mean radius of 0.385 μm and σ of 1.29. The coarse mode of the particles is strongly leached by impaction, while the Aitken and nucleation modes are collected by Brownian motion. The gas to particle [...]"

♣ Page 6, lines 187–192

"[...] each condensed water species has a nonzero fall speed. In this study, Meso-NH simulation have a horizontal grid spacing of 15 km with parameterized convection resulting from a trade-off between a high resolution for detailed dynamics of the mesoscale convective systems an efficient run over a large domain covering the entire AMA. There is certainly an effect of not explicitly considering aerosol activation on clouds that is difficult to quantify without performing a higher resolution simulation. However, in deep convection, high vertical velocities create significant supersaturation and tend to activate much of the available aerosol spectrum. The turbulence parameterisation is based on a 1.5-order closure [...]"

5. Fig. 6 and Fig. 7: CO data from MLS is suggested to compare with the model simulation.

Please see our reply to major comment #2.

6. Fig. 10: CALIPSO data for aerosol is suggested to compare with the model simulation if the signal is so strong.

Indeed vertical profiles of backscatter retrieved from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) on board CALIPSO (Winker et al. 2009) with a wavelength at 532 nm has been widely used to understand the particle contribution. CALIOP data has been thus employed in many studies regarding of not only aerosol (Mielonen et al., 2009; Yorks et al., 2009; Devasthale and Thomas, 2011), but also hydrometeors, i.e. ice crystals in high clouds (Yoshida et al., 2010; Baum et al., 2011; Lee et al., 2019).

StratoClim campaign took place during a break phase of the monsoon with an intense convective activity over south China. Many deep convective clouds transported a large amount of both solid hydrometeor and boundary aerosol to ATAL, thus the ATAL composition during StratoClim campaign is a mixture of both. Actually using CALIOP data, the stratospheric hydration by deep convections during flight #7 had been studied by Lee et al. (2019). They reported that ice content up to 1.9 eq. ppmv distributes in altitudes of 17–18 km in the upper troposphere. Figure A (part of Figure 3 in Lee et al., 2019) demonstrates the V-shaped high backscatter signal region in altitudes of 16–18 km (pointed by a white arrow in Fig. Aa) are successfully reproduced by Meso-NH simulation (Fig. Ab) as observed by CALIOP. Furthermore the V-shaped region is mostly composed with ice contents (Fig. Ac). Their study shows that during active convection phase of summer monsoon, strong backscatter signals in high altitudes detected by CALIOP might not point out only aerosol. Thus we prefer not to include CALIOP data in Figure 10.

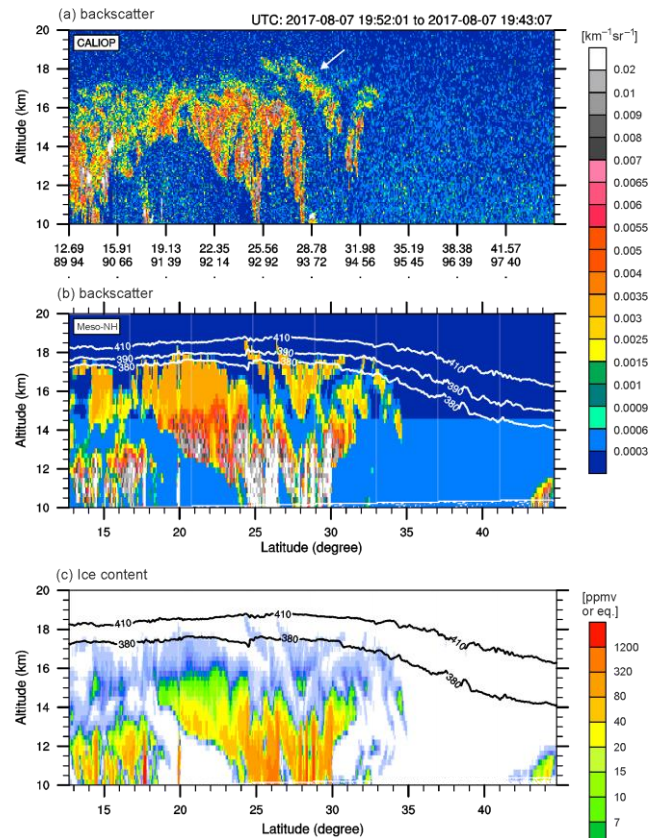


Figure A. Backscatters at 532 nm (a) measured by CALIOP around 20:00 UTC and (b) retrieved by the Meso-NH simulation, and ice content (eq. ppmv) produced by the Meso-NH simulation along the CALIOP track at 20:00 UTC on 7 August 2017. (Lee et al., 2019)

7. L340: Aerosols with radius of 0.385 μm are easily removed from the convection by activation.

You are right. For the sake of clarity, additional information has been included in the manuscript.

♣ Page 6, lines 170–175

“[...] Tulet et al., 2005, 2006, 2010). The impaction scavenging by raindrops depends mainly on Brownian motion, interception, and inertial impaction following a formula originally described by Slinn (1983). Two lognormal modes of particles are considered, mode #1 (i.e. Aitken mode) of smaller particles with initial mean radius of 0.036 μm and standard deviation (σ) of 1.86, and mode #2 (i.e. accumulation mode) of larger particle with initial mean radius of 0.385 μm and σ of 1.29. The coarse mode of the particles is strongly leached by impaction, while the Aitken and nucleation modes are collected by Brownian motion. The gas to particle [...]”