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

Comments

The revised version has answered most of my concerns. My only concern is the simulation of the aerosol in the upper troposphere and lower stratosphere, which has no support from measurement and is not the critical concern of this manuscript. My suggestion is to remove this part.

We appreciate the time and effort you put in this review as well your helpful comments on our paper. In this study, we only discuss and display primary particles POA and BC in order to compare their distributions with a primary gas, CO. This allows to show that these particles have a different fate and different distributions due to their interactions with clouds which is not the case of CO. It is true that the part regarding the simulation of aerosol, i.e. primary particles POA and BC, in the upper troposphere and lower stratosphere (UTLS) has not been supported by measurement. However authors believe that the model results shows interesting characteristics of primary particles with respect to the development of deep convective system, and the need of future field campaigns with innovative instrument which can detect various aerosol particles. With these reason, we kindly propose to keep the part about the simulation of aerosol as it is, while further discussion has been included as below.

♣ Page 12, lines 378−381

"[…] stored at the mountain foothills into the AMA. The simulation results shows us interesting aspects of primary particles of POA and BC with respect to the development of deep convective clouds however it is not supported by measurement and show the need of future field campaigns deploying instrument which can detect various aerosol particles."

Referee #2 comments

Summary general Comments

This paper discussed the chemical properties of the South Asian Upper Troposphere Lower Stratosphere during the Asian Summer Monsoon. They used Meso-NH cloud-chemistry model to simulate the monsoon deep convection on the composition of Asian Monsoon Anticyclone at the 15 km spatial resolution during the StratoClim Campaign in 2017. The simulated CO, O3, POA, and BC were compared with StratoClim and IAGOS observations. Overall, this paper is good, except for some defects in the analysis and model description.

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.

General Comments:

1. The simulation was conducted at 15 km resolution. The Kain-Fritsch-Bechtold scheme was used to simulate the parameterized (subgrid) convection. Do you use any scheme to simulate the subgrid convective transport and wet scavenging of the chemical properties? Or maybe the Kain-Fritsch-Bechtold scheme included the subgrid convective transport and wet scavenging of the chemical properties? Could you show more details about the subgrid convective transport and wet scavenging of the chemical properties in the model description part?

The subgrid convective transport and wet scavenging of the chemical properties are very important at 15 km spatial resolution especially in the developing stage of the convection. You can refer to the following two papers for more details:

Grell, G.A., and Freitas, S.R. (2014). A scale and aerosol aware stochastic convective parameterization for weather and air quality modeling. Atmospheric Chemistry and Physics, 14(10), 5233−5250. https://doi.org/10.5194/acp-14-5233-2014.

Li, Y., Pickering, K.E., Barth, M.C., Bela, M.M., Cummings, K.A., and Allen, D.J. (2018). Evaluation of parameterized convective transport of trace gases in simulation of storms observed during the DC3 field campaign. Journal of Geophysical Research: Atmospheres, 123, 11238−11261. https://doi.org/10.1029/2018JD028779.

As mentioned in previous studies, e.g. Grell and Freitas (2014) and Li et al. (2018), to obtain reasonable simulations of the impact of convection on tropospheric composition, subgrid convective transport needs to be computed in a manner consistent with the subgrid convection in the driving meteorological model. In this study, the Kain-Fritsch-Bechtold scheme (Bechtold et al., 2001) was used with a horizontal grid spacing of 15 km. Scavenging by subgrid wet convective updrafts is applied within the convective mass transport algorithm in order to prevent soluble tracers from being transported to the top of the convective updraft and then dispersed on the grid scale. The transport model provides wet convective air mass fluxes through each grid level in the updraft. This piece of information has been included in the model description part (section 2.3).

♣ Page 6, lines 194−198

"[…] et al., 2001). In order to obtain reasonable simulations of the impact of convection on tropospheric composition, subgrid convective transport needs to be computed (Grell and Freitas, 2014; Li et al., 2018). Scavenging by subgrid wet convective updrafts is applied within the convective mass transport algorithm in order to prevent soluble tracers from being transported to the top of the convective updraft and then dispersed on the grid scale. The transport model provides wet convective air mass fluxes through each grid level in the updraft. [...]"

♣ References

Grell, G.A. and Freitas, S.R.: A scale and aerosol aware stochastic convective parameterization for weather and air quality modelling. Atmospheric Chemistry and Physics, 14(10), 5233−5250. doi:105194/acp-14-5233-2014, 2014.

Li, Y., Pickering, K.E., Barth, M.C., Bela, M.M., Cummings, K.A., and Allen, D.J.: Evaluation of parameterized convective transport of trace gases in simulation of storms observed during the DC3 field campaign. Journal of Geophysical Research: Atmosphere, 123, 11238−11261. doi:10.1029.2018JD028779, 2018.

2. The concentrations of the chemical properties in the upper troposphere are very sensitive to the relative location of the storm. Could you plot the flight the flight tracks over the BT plots (or over radar reflectivity, or anything that can show the storm location) for each flight?

Agreed. Figure 1 has been extended by including four sub-figures. The StratoClim flight track of #5, #6, #7, and #8 are displayed in (b), (c), (d), and (e), respectively, on the BT distribution at the time around flight departure. Manuscript and caption of Figure 1 have been accordingly revised as below:

♣ Page 4, lines 102−108

"We will use data from M55-Geophysica flights #5, #6, #7, and #8, which took place from Kathmandu in Nepal (Table 1; for the tracks see ~~red~~ bluish lines marked by 'STCLM' in Fig. 1a and green lines in Fig. 1b−1e). During those flights, the AMICA (Airborne Mid Infrared Cavity enhanced Absorption spectrometer) and FOZAN-II (Fast OZone ANalyzer) instruments measured the CO and O_3 concentrations respectively. During 03:00−07:25 UTC on 4 August for flight #5 (Fig. 1b), during 07:30−11:30 UTC on 6 August for flight #6 (Fig. 1c), during 04:30−06:50 UTC on 8 August for flight #7 (Fig. 1d), and during 08:40−12:30 UTC on 10 August for flight #8 (Fig. 1e). Flights #5 and #7 flied in the region almost without tall clouds while flights #6 and #8 flied in the cloudy neighbours."

♣ Page 8, lines 247−252

"[…] for flight #6 it appears that 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 plumes. Also note that flights #6 and #8 flied into populated regions of convective clouds (BT ≤ 210 K; see Figs. 1c, 1e). The location of convective clouds strongly affects the concentrations of chemical properties. […]"

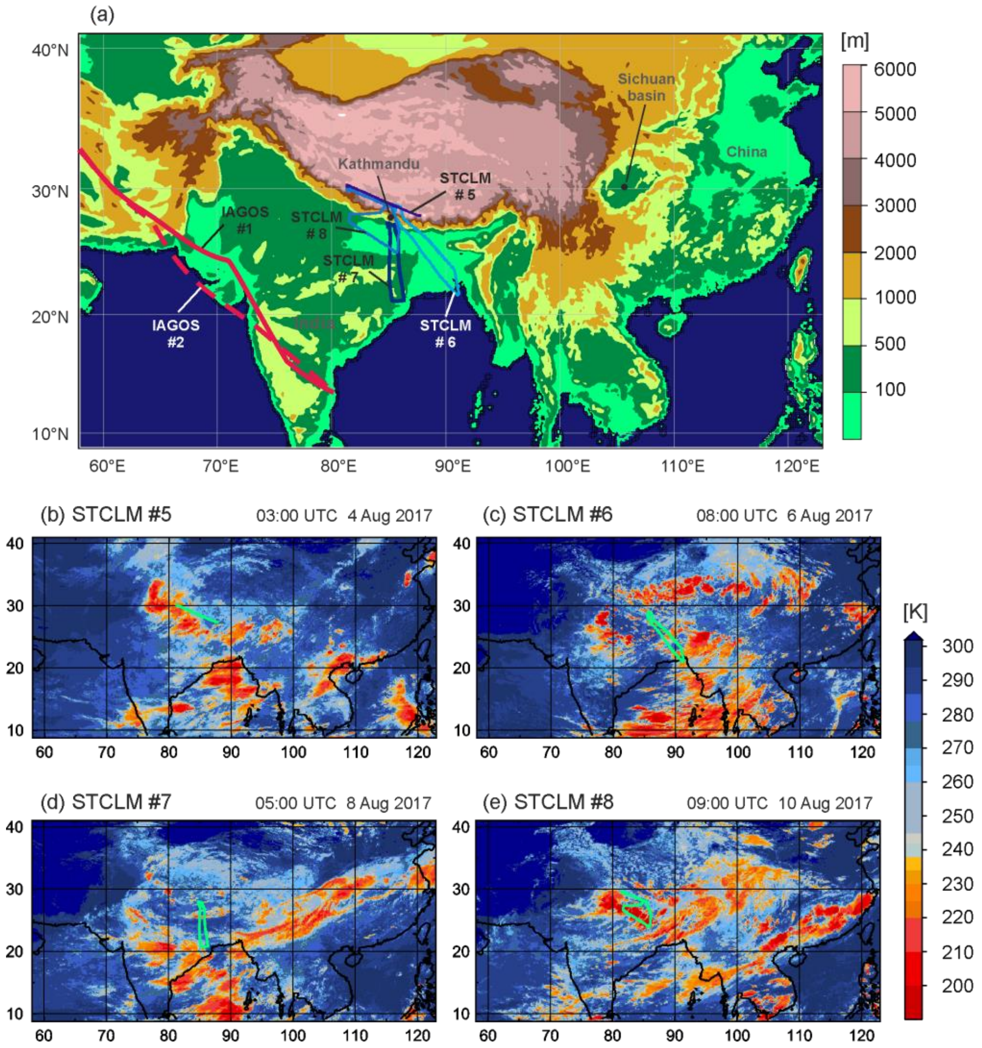

Figure 1. (a) Topography and domain considered in the Meso-NH numerical simulation with a resolution of 15 km. The trajectory of the Geophysica flights #5, #6, #7, and #8 during the StratoClim campaigns (marked with 'STCLM' which is a shortened name of StratoClim) around and south of Kathmandu are shown by the bluish solid lines in (a), while the trajectory is displayed by green solid lines and overlapped on BT distribution at the time around each flight's departure in (b), (c), (d), and (e), respectively. In (a), ~~while~~ two IAGOS flight tracks (to/from Madras in India) are indicted by the red solid and dashed line.

Specific Comments:

1. Line 25: What's IAGOS? Could you show the full name of IAGOS?

Done.

♣ Page 1, line 25–26

"[…] and airborne observations (StratoClim and IAGOS (In-service Aircraft for a Global Observing System) [...]"

2. Line 60: If you decided to use CO to represent carbon monoxide, please keep consistent throughout the paper (e.g in line 102, and in the captions of Figures 2, 4, 6, 7, 9).

Done.

3. Line 62: Please show the full name of CALIPSO.

Done.

♣ Page 2, line 66–67

"[...] observations from the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) spaceborne lidar evidenced [...]"

4. Line 83: Please show the full name of MACCity.

Done.

♣ Page 3, line 88

"emissions from the MACCity (MACC/CityZEN EU project) inventory (https://eccad3.sedoo.fr/) [...]"

5. Line 100: Could you show the aircraft tracks over radar reflectivity or BT to show their relative location to the convections?

Done. See our answer to major comment #2.

6. Line 105: Please show the full name of HITRAN.

Done.

♣ Page 4, line 112

"[...] parameters taken from the HITRAN (high-resolution transmission molecular absorption) 2012 database [...]"

7. Line 124: Please show the full name of CTL.

The *48CTL* is the name of instrument and is not an acronym.

8. Section 2.3: What's the time resolution of the simulation? It would be better if you can use a table to show the model setups.

The time resolution of the simulation had been set to 40 s which is sufficient to simulate the target convective uplift of pollution and to compare it with fine-scale airborne measurement. This piece of information has been included. Also, as suggested, specification of model setup has been summarized in Table 2 as below.

♣ Page 5, line 155

"[...] 7 million grid points; see Table 2 for a detailed description of the model)."

♣ Page 23

Table 2. Specification of model setup.

Horizontal resolution	15 km
Vertical resolution	100 m to 450 m
Temporal resolution	40 s
Emission	MACCity, MEGAN, FGEDv3
Meteorology boundary	ECMWF analyses
Chemical boundary	MOZART-4
Chemical scheme	ReLACS 2
Aerosol modules	ORILAM, ORILAM-SOA

9. Line 181: Are these meteorology schemes are the best combination? Did you try other schemes?

The combination of the selected meteorological schemes (e.g. microphysical scheme, turbulence parameterization, transport scheme) has already shown its capability to simulate heavy precipitation events in both real and idealized framework (Ducrocq et al. 2008; Bresson et al. 2012; Lee et al. 2018, 2019 among others). Authors are confident with the selected meteorological schemes and it has been mentioned in manuscript.

♣ Page 7, lines 210–213

"[…] (Colella and Woodward, 1984). This combination of meteorological schemes has already shown its capability to simulate heavy precipitation events in both real and idealized frameworks (Ducrocq et al. 2008; Bresson et al. 2012; Lee et al. 2018, 2019 among others). […]"

♣ References

Ducrocq, V., Nuissier, O., Ricard, D., Lebeaupin, C., and Thouvenin, R.: A numerical study of three catastrophic precipitating events over southern France, Mesoscale triggering and stationary factors, Q. J. Roy. Meteor. Soc., 134, 131–145, 2008.

Bresson, E., Ducrocq, V., Nuissier, O., Ricard, D., and De Saint-Aubin C.: Idealized numerical simulations of quasi stationary convective systems over the Northwestern Mediterranean comple terrain, Q. J. Roy. Meteor. Soc., 138, 1751–1763, doi:10.1002/qj.1911, 2012.

Lee, K.-O., Flamant, C., Duffourg, F., Ducrocq, V., and Chaboureau, J.-P.: Impact of upstream moisture structure on a back-building convective precipitation system in south-eastern France during HyMeX IOP 13. Atmos. Chem. Phys., 18, 16845–16862, doi:10.5194/acp-18-16845-2018, 2018.

Lee, K.-O., Dauhut, T., chaboureau, J.-P., Khaykin, S., Krämer, M., and Rolf, C.: Convective hydration in the tropical tropopause layerduring the StratoClim aircraft campaign; pathway of an observed hydration patch. Atmos. Chem. Phys., 19, 11803–11820, doi:10.5194/acp-19-11803-2019, 2019.

10. Line 181: How did the model transport the chemical properties in the subgrid scale? See general comments 1 for more details.

See our answer to major comment #1.

11. Section 3.2: Could you compare the simulation with the observation before convection started to prove the accuracy of the initial chemistry condition?

For the initial aerosol species are taken from MOZART-4 (Model for Ozone and Related chemical Tracers, version 4) had been used. Background chemistry condition had been compared using IAGOS which measured the CO and O_3 concentrations and the comparison results (see section 3.2) shows that the model captures most of the general background variabilities within the AMA (Asian Monsoon Anticyclone).

12. Section 3.2: When you compared the simulated and observed chemical properties, did you use a criterion to separate the inside-cloud and outside-cloud region? The CO concentration might be very different inside and outside the cloud.

Agreed. The CO concentration is sensitive to cloud appearance. Comparison between BT at 10.8 microns observed by satellite sensors and simulated by Meso-NH highlights the ability of the model to correctly reproduce the life cycle of convective clouds; this allows us to continue to validate the CO concentration in the UTLS. Flights #6 and #8 indeed flied into populated regions of convective clouds (BT ≤ 210 K; see Figs. 1c, 1e), and the model missed the peak of CO concentrations whereas model performance was relatively better for flights #5 and #7 which flied into rather clear sky. Above discussion has been included in manuscript (also see our answer to major comment #2).

♣ Page 8, lines 247–251

"[…] flight #6 it appears that 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 plumes. Also note that flights #6 and #8 flied into populated regions of convective clouds (BT ≤ 210 K; see Figs. 1c, 1e). The location of convective clouds strongly affects the concentrations of chemical properties. […]"

13. Line 265: The height of the tropopause layer might increase in the area of deep convections. Therefore, it is not good to the climatological TTL height to determine whether the pollution affected the lower stratosphere or not. It's better to use the temperature gradient (like the WMO tropopause definition) to determine the height of the TTL.
Agreed. There exist several tropopause definitions in various literature (e.g. WMO, 1957; Maddox and Mullendore, 2018), considering temperature lapse rate, potential vorticity, static stability, and tracer chemicals. As you mentioned, it is true that there is a difficulty of defining a tropopause in convective environment. The term of TTL has been replaced by UTLS.

♣ Page 9, line 285
"[…] tend to overestimate CO by up to 20 ppbv in the UTLS ~~TTL (Tropical Tropopause Layer)~~ especially […].

Maddox, E.M. and G.L. Mullendor: Determination of Best Tropopause Definition for Convective Transport Studies. J. Atmos. Sci., 75, 3433–3446, https://doi.org/10.1175/JAS-D-18-0032.1, 2018.
WMO: Definition of the tropopause, WMO Bull., 6, 136, 1957.

14. Line 290: How do you define the cloud boundary? I see the definition in the caption of Figure 8, could you mention it in the main content of the paper?
Done.

♣ Page 10, lines 309−310
"[…] together with cloud contour (mixing ratio of ice content 10 mg kg^{-1}) corresponding to the locations […]"

15. Line 321: Could you use an equation to describe the contribution of Sichuan?
Done.

♣ Page 11, line 345−346
"relative difference between CNTL and SIC06 simulations (= [CNTL minus SIC06] over [CNTL]) averaged over the AMA domain (20−35°N, 60−120°E). […]"

16. Line 324: Change "The evolution of the difference…" to "The evolution of the contribution of Sichuan…"
Done.

♣ Page 11, line 346
"[…] (20−35°N, 60−120°E). The evolution of the contribution of Sichuan ~~difference~~ is displayed in Fig. 9 with […]"

17. Figure 1. What's STCLM?
Done.

♣ Page 24
"**Figure 1.** […] the StratoClim campaigns (marked with 'STCLM' which is a shortened name of StratoClim) around […]"

18. Figure 2: This is the first time "CNTL", "SIC06", "SIC01", "CHN01", and "IND01" were mentioned in the paper. You may need to explain the meaning of these abbreviations.
This piece of information has been included in caption of Figure 2.

♣ Page 25
"**Figure 2.** The emission map of CO ~~carbon monoxide~~ used for CNTL (control run). Inner boxes indicate the domain of no emission for sensitivity experiments (see also Table 3~~2~~) of SIC06 and SIC01 (101−109°E, 26−33°N, red line), CHN01 (100−122°E, 20−40°N, black line), and IND01 (70−95°E, 10−35°N, green line) simulations. For the name of sensitivity experiments, first three alphabets stand for the area of interest, e.g. SIC for Sichuan, CHN for China, and IND for India while the last two numbers stands for the first day of emission modification. For more details, see Table 3."

19. Figure 7: Change “Horizontal map of BT…” to “Horizontal map of simulated BT…”

Done.

♣ Page 30

“**Figure 7.** Horizontal map of simulated BT […]”

20. Figure 9: Please add the label of the x-axis (i.e. Date).

Done.

♣ Page 32

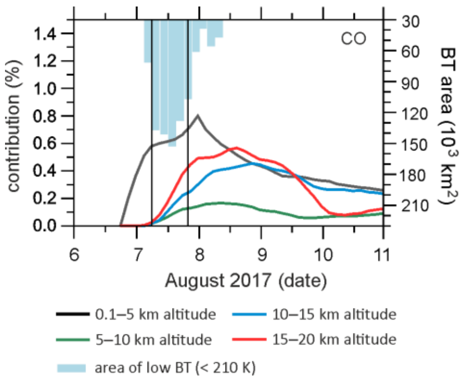

Figure 9. Temporal evolution of contribution of Sichuan emission to CO ~~carbon monoxide~~ concentration of entire AMA region (60−120°E, 20−35°N) every 5 km from 0.1 km to 20 km altitude from 6 to 9 August 2017. The area of low brightness temperature (210 K) in the Sichuan Basin (101−109°E, 26−33°N) are displayed by blue bar.

21. Figure 9: Please show the description of the blue box in the legend and caption.

Done. See our reply to the comment #20.