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 #2 comments

General Comments

Lee et al. studies the convective transport of pollutants from Sichuan basin to Asian Monsoon Anticyclone (AMA) region during one convective event on Aug. 7 of 2017. Lee et al. (2020) uses a cloud-chemistry model (Meso-NH) and observational data from the StratoClim, IAGOS and satellites. Lee et al. shows in section 3 that the model reasonably reproduced observed concentrations of some chemical tracers including ozone and CO compared during the Aug. 7 convective event. Lee et al. demonstrates using the model that the convection quickly transports CO from boundary layer to 18 km and contributes to 0.5% of CO in the 10-20 km layer for 2 days. Besides, Lee et al. shows that India contributes more than China to the CO in AMA and the Chinese portion is significantly contributed by Sichuan basin. In general, I think the paper reports an important transport pathway from Sichuan basin to AMA, which is constrained by the StratoClim datasets. However, some concerns are needed to be addressed before publication.

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. From Figure 3, we know that the Aug. 7 convective event it reproduced well by the model. In terms of the long-term Chinese/Indian contributions (e.g. 10-days averages in Figure 12), is there any information to show that the clouds/convections are reasonably simulated during the 10-day period?

For the sake of clarity on the long-term model ability to reproduce convective clouds, we have joined here the 12-hourly observed and simulated images of brightness temperature (BT, unit in K) from 1st to 10th August. Figure A shows the composite BT images using SEVIRI/MSG and Himawari, and Figure B shows the simulated BT images using CNTL run.

The figures demonstrate that the spatial coincidence of clouds and deep convection (BT ≤ 210 K) is globally good during the 10-days period. Also it shows that the lifecycle of convective clouds within the ASM (Asian Summer Monsoon, south and East Asia from the tropics to the subtropics) circulation is reasonably reproduced by the model. Compared to observed images, Meso-NH tends to slightly underestimate the horizontal extension and the intensity of convective clouds. This piece of information has been included in the manuscript.

♣ Page 13, lines 411–412

"source regions. Observed and simulated clouds are globally coincident during the 1–10 August period. The model slightly underestimates their extension and intensity. CO distribution [...]"

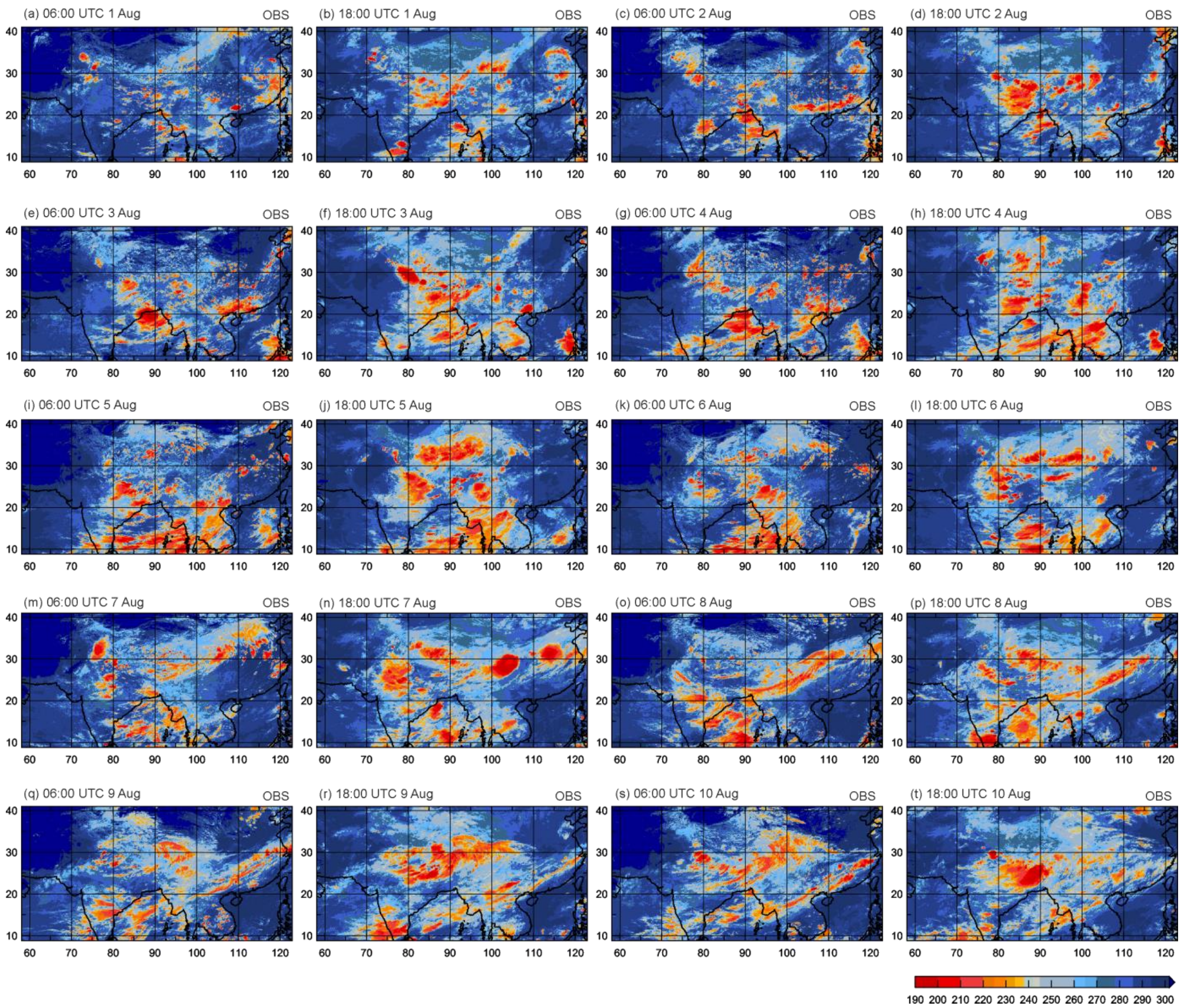

Figure A. BT composite images using SEVIRI/MSG and Himawari at (a) 06:00 UTC, (b) 18:00 UTC on 1 August, (c) 06:00 UTC, (d) 18:00 UTC on 2 August, (e) 06:00 UTC, (f) 18:00 UTC on 3 August, (g) 06:00 UTC, (h) 18:00 UTC on 4 August, (i) 06:00 UTC, (j) 18:00 UTC on 5 August, (k) 06:00 UTC, (l) 18:00 UTC on 6 August, (m) 06:00 UTC, (n) 18:00 UTC on 7 August, (o) 06:00 UTC, (p) 18:00 UTC on 8 August, (q) 06:00 UTC, (r) 18:00 UTC on 9 August, (s) 06:00 UTC and (t) 18:00 UTC on 10 August 2017. The coastlines are marked by black solid lines.

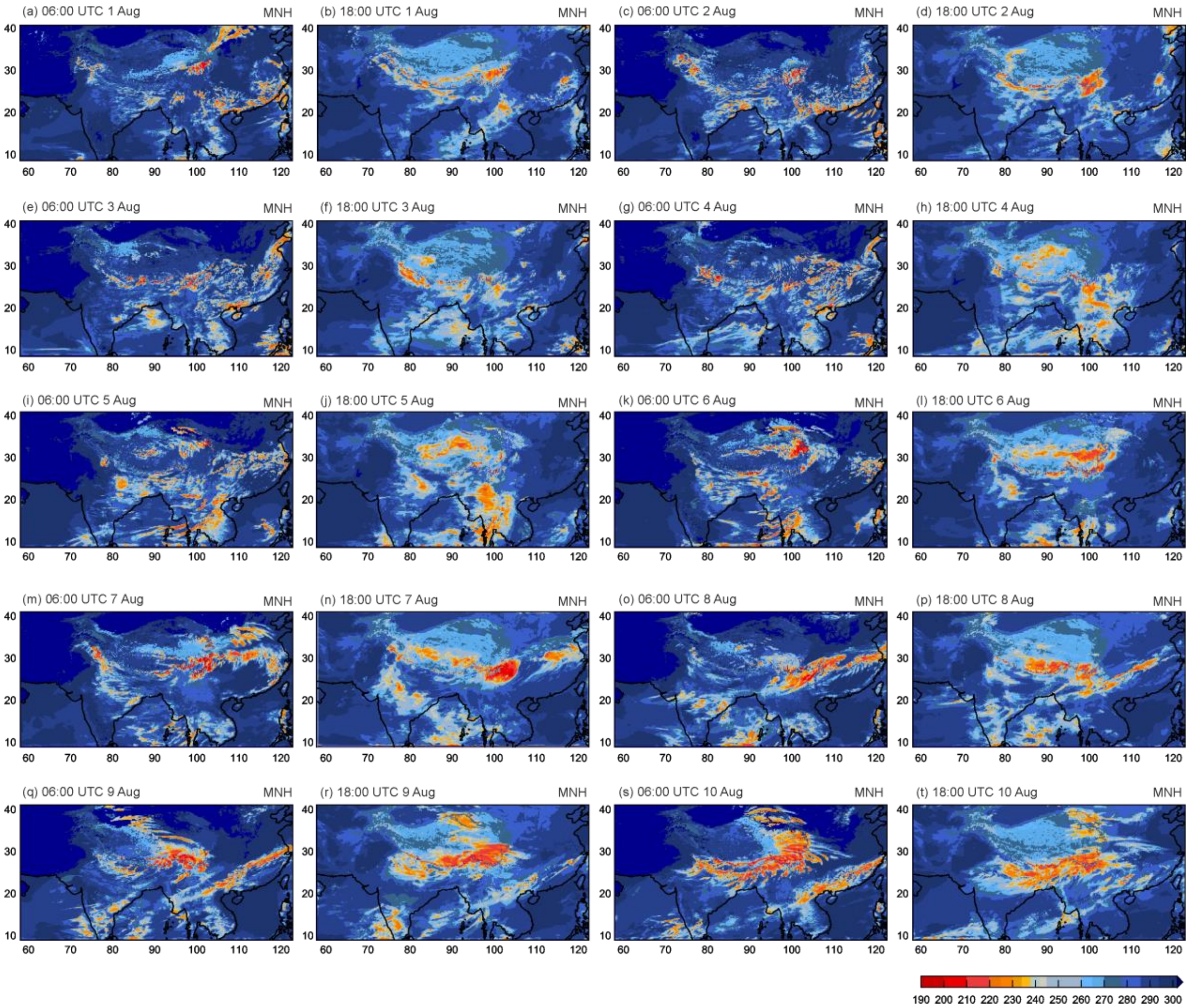

Figure B. Same as Figure A but from the Meso-NH simulation.

2. For the CNTL and other sensitivity simulations, is the multiple convections similar amount those runs (including the starting time, base height, BT, LWC etc)? I am asking this question because we are talking about 0.5% of anomalies due to Sichuan Aug. 7 event. Can we tell the number (i.e. contribution fraction) you derived are statistically significant?
Convection in the CNTL and 4 sensitivity simulations have been identically initialized with ECMWF analyses and identically parameterized with the Kain-Fritsch-Bechtold scheme (Bechtold et al., 2001). The simulated convective systems are thus exactly identical, in terms of lifetime, depth, LWC, etc. For instance, Figure C shows the BT distributions reproduced by the CNTL and the four sensitivity simulations at 18:00 UTC on 7 August, time of matured deep convection over the Sichuan Basin. The figure demonstrates that the horizontal extent and depth of deep and shallow clouds reproduced by CNTL and sensitivity simulations are exactly the same. This is true for the other meteorological variables such as humidity or temperature.

The only change among the five simulations concerns the surface emissions. From the SIC06 simulation we determined that Sichuan pollution convectively uplifted by the 7 August event was responsible of large and significant CO enhancements (6 to 12 %) over a 1000 km broad region (Figure 7i). The 0.5 % contribution is given to provide an idea about the impact of pollution uplifted by the 7 August convective event over the whole AMA region. It may seem small because of the dilution effect but it is as significant as the enhancement over the Sichuan region. Moreover, this Sichuan CO contribution is still detectable over Nepal as confirmed by the StratoClim observation during F7 (Figure 4c). Figure 9 also shows that this 0.5% contribution remains steadily until 9 August 12:00 UTC in the two uppermost layers (10−20 km).

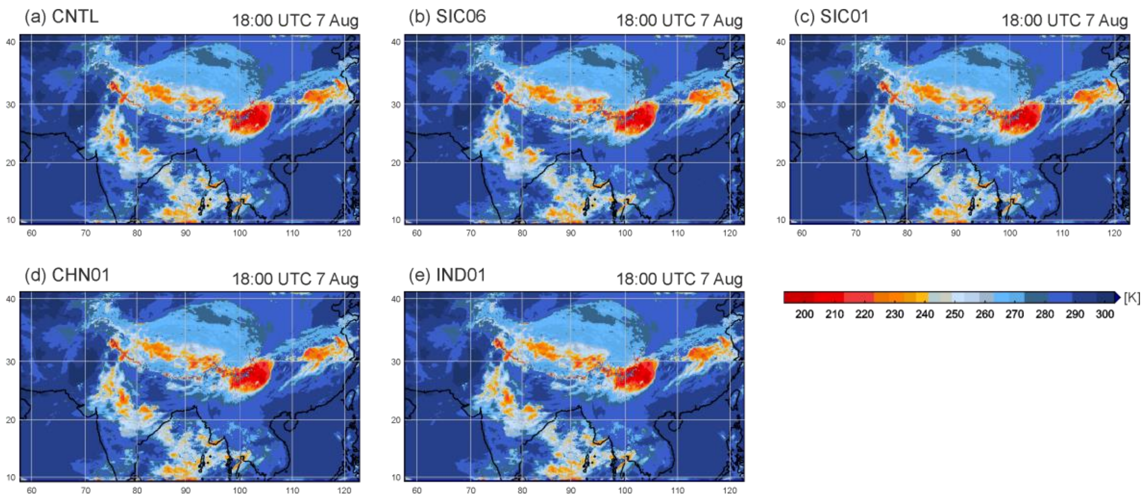

Figure C. Identical BT (K) distributions at 18:00 UTC on 7 August from (a) CNTL, (b) SIC06, (c) SIC01, (d) CHN01, and (e) IND01 simulations.

♣ Page 9, lines 286–287

"[…] All the other environmental conditions are identical to the CNTL run, and the convection activities (i.e. lifetime, intensity) between simulations are as well identical."

♣ Page 12, lines 365–366

"[…] until 00:00 UTC on 8 August. Note the convective activity is identical in all experiments. […]"

3. I am concerned by the analysis on aerosol (POA and BC) and Figure 11. What is the parameterization scheme of the convective removal? Does the secondary activation of aerosols (e.g. Grell and Freitas et al., 2014, ACP; Wang et al., 2013, GMD; Yu et al., 2019, GRL) consider in this study? Convection can quickly remove aerosols in-cloud, which results in fast (in log-scale) decay of aerosols from Figure 8, seems modeled POA and BC can be transported from BC to UT without much loss, which seems not right to me. (note, unlike insoluble species CO in your Figure 8, aerosol even BC and POA can be internal mixed and activated).

The wet deposition scheme is based on Tulet et al. (2010). The kinetic mass transfer between aerosols and cloud or rain drops is considered. The impaction scavenging by raindrops depends mainly on Brownian motion, interception, and inertial impaction following a formula originally described by Slinn (1983). See also Seinfeld and Pandis (1997), Pruppacher and Klett (2000), Tost et al. (2006) for classical parameterization in mesoscale models. Thus the collection efficiency depends on the size of the aerosols (and secondarily on the size of the raindrops). The coarse mode of the particles is strongly leached by impaction. The Aitkin and nucleation modes are collected by Brownian motion.

On the other hand, the accumulation mode is globally little impacted by these two processes and remains preserved in the cloud. This is physically true for insoluble aerosols such as BC, which are not CCN. So the reviewer is right to say that if BC becomes hygroscopic by mixing with soluble secondary compounds (organic for example), it becomes potentially CCN and should be activated into clouds droplets. This process is not taken into account in the simulation and is a source of error. This limitation has been mentioned in the text.

♣ 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 11, lines 355–359

"[...] This result thus implies that aerosol sizes in both modes within the polluted plume are increased during the uplifting within the cloud by gas-particles conversion, condensation of water in the aerosol and coagulation (Andronache, 2003; Tost et al., 2007; Berthet et al., 2010; Tulet et al., 2010). Note that mixing of insoluble aerosols such as BC with soluble secondary compounds to become hygroscopicand potentially CCN (cloud condensate nuclei) that could be activated into cloud droplets is not taken into account in the simulation. [...]"

Minor Comments:

1. For Meso-NH CNTL run, what are the initial conditions for clouds?

Clouds are formed after a saturation adjustment. The model is initialized by ECMWF analyses and the cloud formation will therefore take place in the first time steps of the model (spin-up). Generally, the model is well balanced after 2 hours of simulation. This piece of information has been included in the manuscript.

♣ Page 6, lines 182–183

"The meteorological conditions are initialized by the ECMWF analyses and clouds are formed in the first time steps of the model (spin-up) after a saturation adjustment. Deep convection is parameterised [...]"

2. Are aerosols activated to CCN in Meso-NH, which can influence the cloud droplet number? Since this study heavily relies on the parameterizations of the convections (which shows pretty nice agreement in Figure 3), more information on the aerosol-cloud interaction schemes are needed in the method section.

We do not use aerosol 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. At this resolution, convection, a part of the clouds and precipitation are not explicitly resolved. 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-Fritch-Bechtold (Bechtold et al., 2001) also widely used by the international community and well adapted to this resolution.

In order to study the effect of aerosol activation on clouds, a grid-nesting simulation should be carried out to reach the resolved cloud scale (< 3 km horizontal resolution). 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 (CCN). Thus, and particularly in polluted environments, we can reasonably assume that CCN are not a limiting factor in cloud formation. This has been mentioned in the manuscript.

♣ 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 [...]"

3. Figure 6, the colored circles are extremely difficult to find. Might consider using circles with black boundaries.

Thanks for this suggestion. For the sake of visibility, Figure 6 (a and c) has been improved by closing the circles with black boundaries and reducing the data interval to 5 min from 4 s.

♣ Page 28

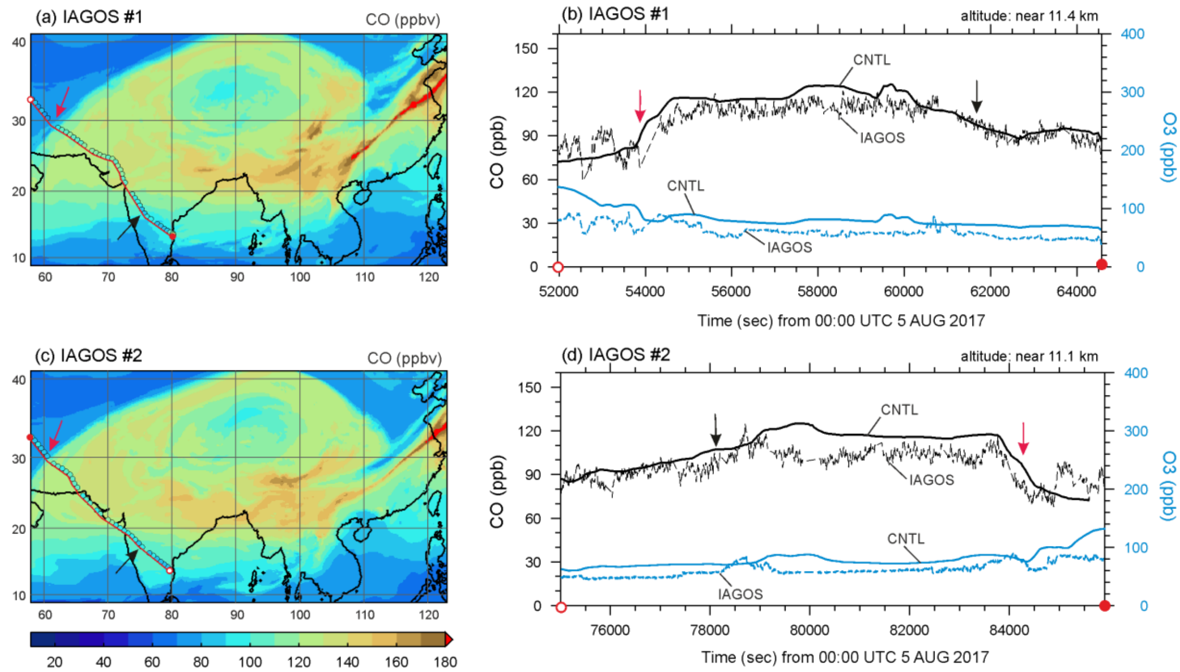

Figure 6. IAGOS-measured (dashed lines) and Meso-NH-derived (solid lines) carbon monoxide (black lines) and O_3 (blue lines) along IAGOS flight tracks on 5 August 2017. In (a) and (c), Meso-NH-derived CO at the altitude of 11.1 km are displayed by shaded areas, while the IAGOS-measured CO every 5 min are displayed by coloured circles along the track (red lines). In (b) and (d), IAGOS-measured CO and O_3 every 4 s are displayed. In (a)–(d), the starting (ending) point of each flight within the domain is marked by open (closed) red circle, while the location of the steep (gradual) change of carbon monoxide is marked by red (black) arrows.

4. Figure 12 caption, AMA region? Altitude info is missing.

Indeed. This piece of information has been included.

♣ Page 33

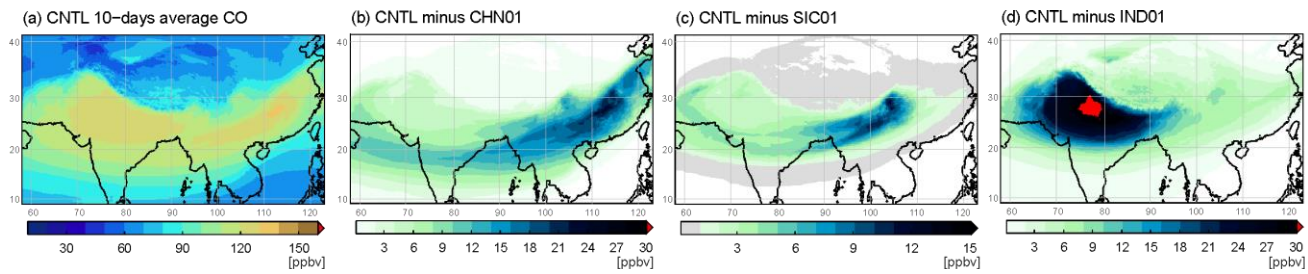

Figure 12. 10-days averaged chemical components of CO (ppbv) at the altitude of 14.8 km from 1 to 10 August 2017 produced by (a) CNTL, (b) CNTL minus CHN01, (c) CNTL minus SIC01, and (d) CNTL minus IND01.